

## Abstract

**Berry, Tiffany Leigh.** Measurement of Smoke Point in Laminar Jet Diffusion Flames at Atmospheric and Elevated Pressures. Under the Direction of Dr. William L. Roberts.

Using a Burke-Schumann modeled co-flow burner, a quartz chimney, and a pressure vessel with good optical access, the smoke points in pure and diluted fuels were measured in a laminar jet diffusion flame. Ethylene and methane, burning in a velocity matched, over-ventilated co-flow of air, were tested over the ranges of 1 to 8 atmospheres and 2 to 16 atmospheres, respectively. Various diluents (nitrogen, argon, helium, and carbon dioxide) were added individually to the pure fuels to observe the effects they have on the smoke points and the adiabatic flame temperatures at atmospheric and elevated pressures. These diluents were chosen to allow a wide range of flame temperatures and fuel Lewis numbers to be investigated. For a given fuel flow rate, the dilution level was increased until the flame ceased emitting visible soot (defined as the smoke point). The height of the flame was then measured and the adiabatic flame temperature was calculated based on equilibrium chemistry. While some previous research has focused on the effects of flame temperature (through dilution) on smoke points, the measurements reported here were made to investigate the effects of pressure, different diluents, and varying dilution rates on sooting tendency. The main findings of these experiments were: increasing the amount of diluent to a pure fuel increases the smoke point, the smoke point is a function of the air to fuel velocity ratio, smoke point is strongly dependent on the inverse of pressure, and residence time decreases with increases in pressure.

# **Measurement of Smoke Point in Laminar Jet Diffusion Flames at Atmospheric and Elevated Pressures**

by

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## **Biography**

The author was born Tiffany Leigh Berry on December 31, 1980 in Raleigh, NC, daughter of Billy and Dorothy Berry and younger sister of Adrienne Lynn Berry Bauer. She graduated from Cary High School in 1999, and chose to attend North Carolina State University to pursue a degree in Aerospace Engineering. Upon completion of her undergraduate degree in May 2003, Tiffany decided to continue her education at North Carolina State University in pursuit of a Master of Science degree in Aerospace Engineering. At the conclusion of the first year of her graduate program she received the General Electric Faculty of the Future Program Fellowship for the following academic year, and had the opportunity to aid Dr. Tarek Echehki in preparing and giving lectures for graduate level courses. These experiences further emphasized her desires to seek her doctorate degree in Mechanical Engineering in order to pursue a career in academia. Tiffany has been accepted and will attend North Carolina State University for this degree, continuing her work with Dr. William Roberts.

## Acknowledgements

The author would like to first acknowledge her advisor, Dr. William Roberts, whose knowledge of, interest in, and appreciation for the field of combustion are unparalleled; his support, hard work, and dedication to his student's work are to be commended; Dr. Tarek Echehki for his guidance and interest in her aspirations for a career in academia; Dr. Steven Marley, Laura McCrain, Sean Danby, and Michael Schoen for their advice and assistance.

The author would like to acknowledge most of all her family; her parents, Billy and Dorothy Berry, for always believing in her, supporting every one of her dreams (both emotionally and financially) and teaching her the value of hard work, believing in herself, following her dreams, consideration and care for others, and always emphasizing that her family is forever by her side; her sister, Adrienne Bauer, for watching over her throughout her whole life, dealing with all the emotional highs and lows, and for being her best friend, and also for bringing her a brother-in-law, Jim Bauer, who has always understood how much two sisters need each other's time, and for providing the fastest technical support ever; her dog, Spice, for always greeting her with excitement, love, compassion, and a cold nose at the end of long days; and last, but certainly not least, Will Yelverton, for his never-ending belief in her abilities and dreams, constant support in every aspect of her life, commitment to their current life together and the journey and adventures that they await and anticipate for their future together.

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## 1 Introduction

Due to the continually increasing demand for combustion devices used in the conversion of fossil fuels into work and heat in our society, it is essential to determine ways in which these combustion processes can be made more efficient. Fundamentally, diffusion-flame-driven devices, such as diesel engines, can be more fuel efficient; however, they lack the control that premixed-flame-driven devices have over pollutant emissions, particularly soot and the oxidizing of nitrogen. With fuel efficiency as an utmost concern, as our fossil fuels are a depleting resource, it is in the best interest of everyone to turn to diffusion-flame-driven devices as a means for conservation. However, there is an urgent need for research developments to control the particulate emissions of these devices, as soot is not only an environmental hazard, but also a health hazard to humans (Sydbom et al., 2001; Comstock et al., 1998; Morgan et al., 1997; Scheepers et al., 1992). Meeting the United States Environmental Protection Agency's (EPA) 2010 regulations of 0.01 g/bhp-hr of particulate matter for heavy duty diesel emission, for example, will require a more complete understanding of the soot formation processes at elevated pressure. Since soot is an indicator of the interactions between combustion chemistry and fluid mechanics, a further understanding of these interactions would greatly contribute to the overall understanding of these combustion processes. Further, it would be of great interest to investigate the effects of elevated pressure, as this is a key element of most practical combustion devices because they operate at these elevated pressures to gain thermodynamic efficiency. For example, if the smoke point of a laminar jet diffusion flame was observed, using different fuels, it would be possible to

obtain vital information about each fuel's propensity to soot in a specific controlled environment, including those of elevated pressure. Furthermore, if these fuels observed were then diluted, it would be possible to determine the best mixture of fuel and diluent needed to better control soot formation and growth. With this knowledge of dilution, a fundamental understanding of these interactions could be used to validate current chemical kinetic models used for predicting soot formation, and help develop new, more accurate models. The ability to accurately model complex combustion processes would greatly aid all research and development in the combustion field. The smoke point is one of these fundamental chemical kinetic/fluid mechanic parameters that could be used to validate the models and codes of these processes.

## **1.1 Soot Formation**

The sooting processes are very complex, and not wholly understood. However, if these processes could be investigated for further understanding, in turn gaining fundamental combustion knowledge, the overall efficiency of many combustion processes could improve. Recently investigators have identified the need for information regarding the chemical kinetics, fluid mechanics, and interactions between the two, and have studied the reaction mechanisms and chemical reactions leading to soot formation (Frenklach, 2002; Richter & Howard, 2000).

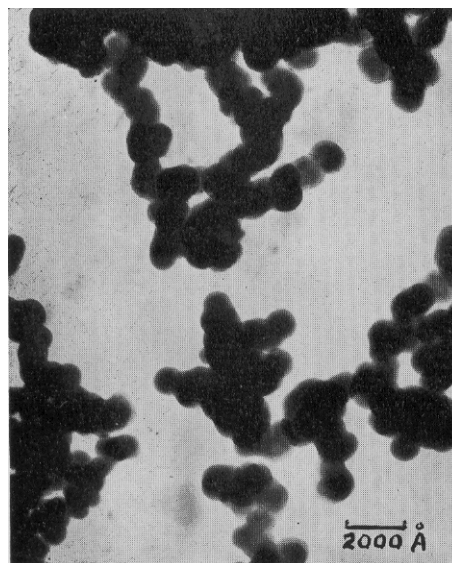
It is important to realize the differences between diffusion flames and premixed flames. Diffusion flames, because of the fundamental controlling mechanisms, typically will produce more soot. In diffusion flames the combustion process is diffusion controlled with the fuel and oxidizer entering the reaction zone from opposite sides of the



primary reaction zone. Fuel and oxidizer diffuse into the reaction zone due to a gradient in concentration (Fickian diffusion). In a diffusion flame the fuel undergoes pyrolysis without the presence of an oxidizer in the mixture, whereas in premixed flames the fuel is heated with the oxidizer present. In premixed flames, the fuel and oxidizer enter the reaction zone together. Moreover, the premixed flame yields soot when the oxidizer is absent, as it is responsible for the converting of hydrocarbons into carbon monoxide and diatomic hydrogen (Wagner, 1981). Wagner experimentally showed if the ratio of carbon to oxygen atoms for the flame is roughly greater than 0.5, the premixed flame will produce soot. Conversely, soot will not be produced if the proper non-soot-yielding ratio of carbon to oxygen atoms is not maintained in the mixture region. However, maintaining this proper ratio of carbon to oxygen atoms in the mixing region of a diffusion flame is typically not possible, therefore making a diffusion flame's environment more capable of producing soot.

The formation of soot begins in the preheat zone, where the large molecules decompose into smaller hydrocarbon pieces, due to the elevated temperatures present in this zone. Because of this pyrolysis, soot precursors, in the form of acetylene ( $C_2H_2$ ) become present. Simple hydrocarbons, such as methane ( $CH_4$ ), are more difficult to thermally decompose into acetylene, and therefore, methane produces little soot. However, a hydrocarbon such as ethylene ( $C_2H_4$ ), which has a high propensity to soot, has a much easier process of thermal decomposition into acetylene. Once these precursors to soot have been created, the acetylene molecules continue on the soot producing path to combine in groups of three, forming benzene rings ( $C_6H_6$ ). Then, several of these benzene rings combine to form polycyclic aromatic hydrocarbons

(PAHs). Next, a process known as polymerization begins, where the rings of benzene lose hydrogen atoms, which form hydroxyl and water, and the rings become unsaturated. As this continues, the carbon to hydrogen atom ratio increases enough to cause the rings to group together to form a soot particle, also called a spherule. The spherule continues to grow in size, until it reaches roughly 30 to 50 nm. The physical reason for the growth to discontinue at this size is still unknown. However, these spherules then begin to join together to first form agglomerates, containing hundreds to thousands of spherules, and then the agglomerates join to form clusters (Figure 1). This process results in soot



**Figure 1: Soot spherules joining to form agglomerates in an ethylene counter-flow diffusion flame (Gaydon & Wolfhard, 1970)**

particles, which have an empirical formula, roughly defined as,  $C_8H$ .

Since this process can produce a large amount of soot, it is fortunate that there is a simple method of soot removal, through oxidation, to combat this soot production and growth. The yellow portion of a flame is the soot incandescence, and if this soot is

oxidized before diffusing across the flame front, then the flame will not emit smoke. This is the difference between what is defined as soot and what is defined as smoke. Carbon that leaves the flame is considered smoke, but carbon that stays within the flame, and is oxidized to CO or CO<sub>2</sub>, is considered soot. Early methods of investigations, by Schalla et al. (1955) examined the smoke and soot production in diffusion flames, and recorded the heights of flame's smoke points for ethylene and ethane flames up to 20 atmospheres. Their studies included wick fed liquid and gaseous fuels. Although there was no metering of the fuel flow rate, they did measure the flame's height at its smoke point. They found that the flame height decreased almost linearly with the reciprocal of pressure for the gaseous fuels. They believed these effects to be caused by diffusion rates, and consequently, the rate of mixing of the fuel and air.

## **1.2 Smoke Point**

A better understanding of smoke point is necessary to the combustion field as it is a fundamental measure of a fuel's propensity to soot. Although the current research was conducted with gaseous fuels, there is an American Society for Testing and Materials (ASTM) method for testing fuels for smoke point, but the testing is primarily for liquid fuels that are wick fed. There is limited experimental work published regarding the smoke points of gaseous fuels, and even fewer publications have focused on smoke points of diluted fuels or the effects of elevated pressure (Glassman, 1988; Gomez & Glassman, 1984; Glassman & Yaccarino, 1981). Slightly more research on smoke points has been conducted and published using numerical simulations (Delichatsios, 1994; Kent, 1986). To the author's knowledge, publications referencing smoke point data with

pressures above atmospheric conditions are not available and minimal information is available at sub-atmospheric conditions (Dai & Faeth, 2000; Urban et al., 2000; Sunderland et al., 1994). Dai & Faeth (2000) investigated air to fuel velocity ratios at sub-atmospheric pressures, using a laminar jet diffusion flame burner, and found the flame's height at its smoke point were twice as long as soot-free (blue) flames under the same conditions. Urban and co-workers (2000) tested laminar jet diffusion flames in microgravity experimental conditions, and reported that contrary to normal-gravity conditions, the laminar smoke point occurred in two configurations: closed-tip flames with soot emissions along the flame axis and open-tip flames with soot emissions from an annular ring around the flame axis. And finally, Sunderland and co-workers (1994) observed smoke points of nonbuoyant jet diffusion ethylene-air flames to find residence times and fuel flow rates at the onset of these smoke points.

In this experimental research conducted on diffusion flames, the flame height and mass flow rates were recorded immediately prior to the flame emitting smoke, for cases of pure and diluted flames. As mentioned above, practical combustion devices operate at elevated pressures, thus making it necessary to not only observe these sooting tendencies at atmospheric conditions, but also at elevated pressure conditions. To further broaden the scope of this research, it was decided that more conclusive results would be established if different fuels with multiple diluents were used.

### **1.3 Effects of Elevated Pressure on Soot Formation and Smoke Point**

It is a well researched and known fact that, in diffusion-driven devices, the production of soot increases, while removal of soot, through oxidation, decreases with

increasing pressure (Flower & Bowman, 1986). However, minimal work has been accomplished at these elevated pressures because it is experimentally extremely difficult, dangerous, and time consuming. Flower (1986) also proved that, not only the physical amount of soot, but also the rates at which this soot is formed increases with increasing pressure. Researchers have agreed that, although it is still not fully understood, increasing the pressure in the environment surrounding a diffusion flame changes the reaction rate and the diffusion coefficient, which lead to increased soot production. Not only relevant to diffusion flames, other researchers have indicated that increasing pressure substantially increases soot production in spray combustion and premixed flames (McArragher & Tan, 1972; Schalla & McDonald, 1955; Kadota et al., 1977; Miller & Maahs, 1977; Millberg, 1959; Fischer & Moss, 1998; Heidermann et al., 1999). Miller and Maahs (1977) found that if a flame was hydrodynamically disturbed then it was more likely to extinguish because diffusion of reactants cannot occur quickly enough to reestablish the flame.

McCrain and Roberts (2005) worked with a diffusion flame at elevated pressure to measure the effects of elevating pressure. Working with laminar diffusion flames burning methane or ethylene, it was possible for them to measure soot volume fraction,  $f_v$ . Soot volume fraction is just one of the parameters that characterizes mass yield of soot. Using laser induced incandescence (LII) and extinction measurements, they found integrated  $f_v$  of methane scales approximately as  $P^{1.0}$ , and for ethylene approximately as  $P^{1.2}$ , in agreement with previous researchers, and peak  $f_v$  for methane and ethylene scales approximately as  $P^{1.2}$  and  $P^{1.7}$ , respectively. McCrain and Roberts (2005) were able to measure methane flames up to twenty-five atmospheres and ethylene up to sixteen

atmospheres, limited by each fuel's propensity to soot. This research, which was the precursor to the current research, showed that testing methane and ethylene flames, both pure and diluted, at elevated pressures in the co-flow diffusion flame burner and high pressure vessel, was possible. The results obtained in the current research are very different from those obtained in McCrain and Roberts' work. However, both experiments are necessary to understanding the fundamental processes involved in the formation of soot and other pollutants. Smoke point measurements, and the addition of diluents to fuels for these measurements, are two more pieces of information that are critical to understanding these reactions that lead to the formation of soot, as smoke point is a fundamental measure of a fuel's propensity to soot, and how these reactions are effected by elevating pressure. The work by McCrain and Roberts (2005), while very important, and original in the fact that no one had ever measured peak  $f_v$ , was a less sensitive measure of the governing chemical kinetics than the current research, which analyzes the boundary between oxidation and production of soot. This more kinetic-sensitive measure will allow for better kinetic code validation studies.

## 2 Experimental Apparatus

### 2.1 Fuels and Diluents

Methane and ethylene were the two fuels chosen for this experimental investigation. Methane ( $\text{CH}_4$ ) was chosen because it can be used to represent natural gases, commonly used in industry, because it is the simplest hydrocarbon, and has a very low propensity to soot. This allowed investigation at higher pressures (1-16 atm). Ethylene ( $\text{C}_2\text{H}_4$ ) on the other hand was chosen because it is a better representation of more complex hydrocarbons and has a much higher propensity to soot. Propane would have been a very useful fuel to study in these experimental investigations; however, it could not be used because it has such a low saturation pressure at room temperature. Four diluents were chosen: nitrogen ( $\text{N}_2$ ), argon (Ar), helium (He), and carbon dioxide ( $\text{CO}_2$ ). Each of the diluents were chosen in order to determine, at a later time, the effects fuel Lewis number has on soot formation. Air is used as the co-flow in all of the experiments and unlike previous experimental efforts in this area, a method of velocity-matching the volumetric flow rate of fuel and air was employed. Also, some test were run at air to fuel velocity ratios of greater than one to see the effects this had in determining the smoke point of diluted fuels.

### 2.2 Co-flow Diffusion Flame Burner

The burner used for this experimental investigation was modeled as the classic over-ventilated Burke-Schumman (1928) laminar diffusion flame. The burner has been

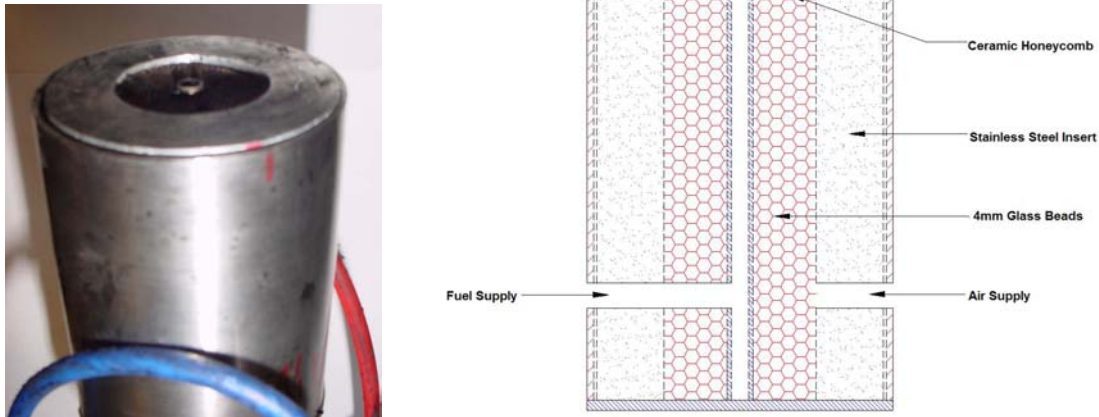
through many iterations of modification to be used for conducting this research. It was modified most recently by McCrain and Roberts (2005) to achieve a stable laminar flame, and then again twice for the current research. The atmospheric pressure data was taken in one burner configuration, and the elevated pressure data in another.

The burner had a height of 104 mm (Figure 2). Originally the inner diameter of the air co-flow was 72.4 mm and the fuel tube's diameter was 4.4 mm. To be able to produce velocity matched air to fuel flow rates it was necessary to design and manufacture a stainless steel insert to reduce the air co-flow diameter to 37.8 mm. This gave the burner approximately an 8.5 ratio between the air co-flow and fuel tube diameters. This allowed for the mixing layer between the fuel and co-flowing air to be undisturbed by the shear layer that forms between the co-flow and ambient air within the pressure vessel.

To further aid in air co-flow straightening, 4 mm glass beads and a tight grid ceramic honeycomb were employed. The glass beads fit into the burner cup and are not visible from the face of the burner as they are under the ceramic honeycomb. The ceramic honeycomb was approximately 51 mm thick, with a diameter of roughly 38 mm.

The fuel tube utilized a very fine grit steel wool as a flow straightener, and this steel wool was packed into the entire length of the fuel tube inside of the burner. This procedure was necessary to make the fuel flow rates less sensitive to pressure fluctuations in the pressure vessel. The fuel tube diameter, and steel wool contents, stayed constant through both of the burner set ups, atmospheric and elevated pressure.





**Figure 2: Co-flow burner**

### **2.2.1 Burner Configuration for Atmospheric Pressure Testing**

For the atmospheric testing the burner was configured as defined in the above section. However, the burner was placed inside of the pressure vessel to avoid outside air disruptions, which caused the flame to be unsteady and caused flickering. The pressure vessel was kept with the exhaust completely open, and the pressure inside was that of the ambient air.

In another attempt to create a stable, laminar flame, a quartz chimney was mounted to the burner. The quartz chimney was optical quality and polished. In order to mount the chimney it was necessary to modify the stainless steel insert to provide a lip to hold the chimney in place. The chimney had an inside diameter of approximately 38 mm and was 381 mm tall. The walls of the quartz chimney were 3 mm in thickness.

### **2.2.2 Burner Configuration for Elevated Pressure Testing**

The burner configuration for the elevated pressure testing was very similar to that used for atmospheric pressure testing, however, in order to achieve the flow rates

necessary to take data at elevated pressures, more air co-flow area was needed. As stated before, the fuel tube diameter and flow straightening remained the same. However, the stainless steel insert, previously in the air co-flow portion of the burner, was removed. This allowed for a diameter of 72.4 mm for the air co-flow region.

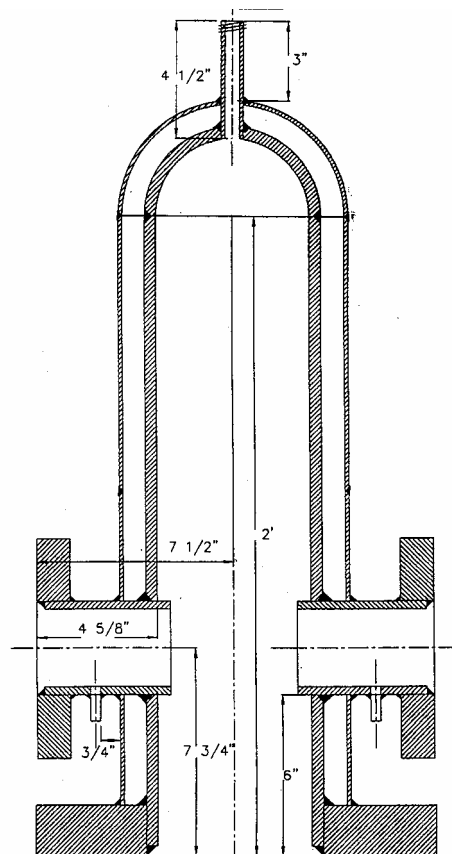
A new piece of ceramic honey comb had to be cut to fit the new air co-flow area. The same material with the same cell size and grid was used. The ceramic was also 51 mm in thickness, but now had a diameter of approximately 65 mm.

A new quartz chimney was purchased in order to fit the new air co-flow diameter. It was constructed from the same material, and was also optical quality and polished. This chimney had an inside diameter of 65 mm, an outside diameter of 71 mm, with a wall thickness of 3 mm. To protect the quartz chimney, and to assure that no air co-flow passed along the outside edge of the chimney, felt (roughly 1mm thick) was placed around the inside of the burner cup.

## **2.3 High Pressure Vessel**

The vessel in which the burner is housed is a water cooled pressure vessel that is rated at pressures up to 30 atmospheres and is shown in Figure 3. It was designed and originally built by Li (2001). The vessel is one meter in height and has four flanges extended beyond its circular body. Three of these flanges house non-intrusive glass (BK-7) windows (7.6 cm diameter by 2.5 cm thick) to allow for optical viewing and diagnostics. The fourth flange houses fittings for pressure readings. Because of the sooting tendencies of the fuels used, and the effect pressure has on these tendencies, the vessel has air ports at two of the windows to purge the area, in order to prevent

accumulation on the window surface and thus allowing optical access. The burner has the capability to be translated vertically inside of the vessel, thus allowing optical access to all portions of the flame. While measuring the smoke points of some diluted flames, the flame became rather tall, and since it is critical to view the tip of the flame for measurements, this became a very important feature. Since the flame was small in size, in comparison to the volume of the vessel, problems with air entrainment and circulation coming off of the optical ports, were negligible. The pressure vessel was removed frequently in between tests for cleaning. Because of the high volume of soot produced this was necessary, as was the disassembling, cleaning, and reassembling of the window ports (Appendix 7-1).

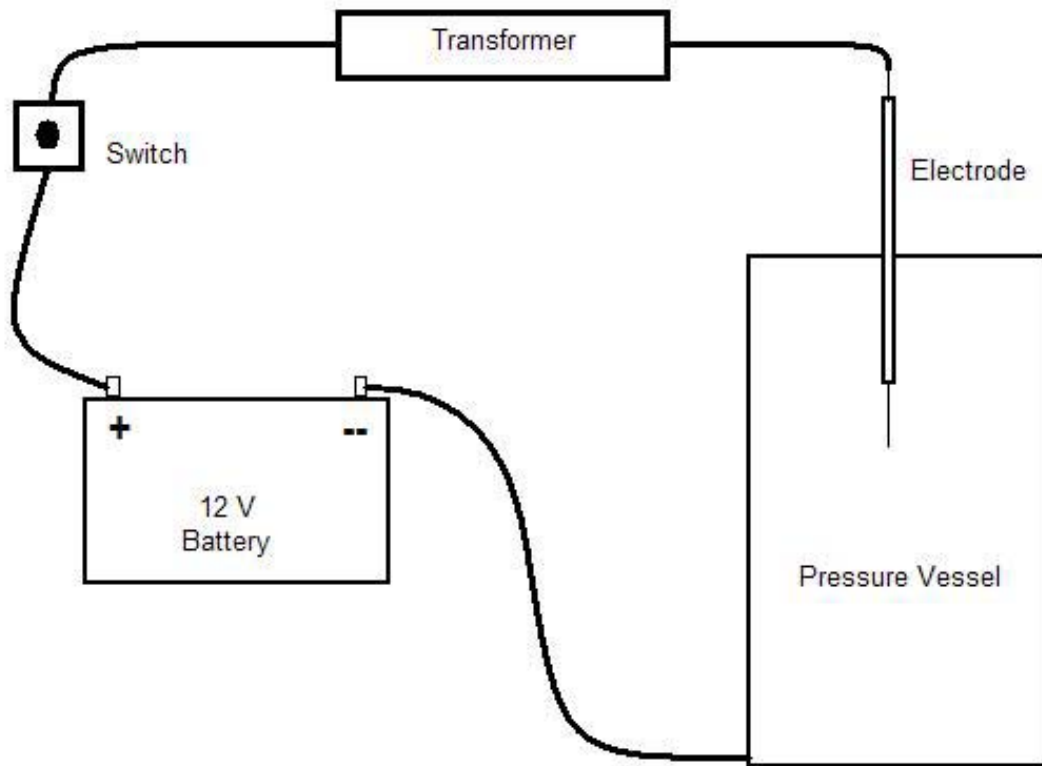


**Figure 3: High pressure vessel**

## 2.4 Ignition System

The ignition system was the simple design of a copper wire electrode. A piece of solid copper wire was encased by a ceramic rod to provide electrical insulation. The copper wire extended out approximately 25 mm from either end of the ceramic rod. The ceramic rod had an approximate inside diameter of 2 mm, an outside diameter of 6 mm, and was 36 mm long. The ceramic rod was coated in electrical tape to ensure that the path of least resistance to ground for the spark was not reached until it was near the fuel tube.

The electrode was placed inside of the exhaust tube of the pressure vessel, entering the top of the chamber above the open end of the quartz chimney, and passing through the chimney until the exposed copper tip of the electrode was roughly four millimeters from the stainless steel fuel tube. Since the electrode passed through the exhaust tube it was necessary to remove it once ignition had occurred. The piece of copper wire extending outside of the pressure vessel was attached to an electrical cable that ran to a low-to-high voltage transformer. The transformer was powered by a 12 volt deep cycle marine battery. The transformer was attached to the positive lead of the battery, while the negative lead was grounded to the experiment. A toggle switch between the battery and the transformer provided an electrical arc inside the pressure vessel from the copper wire to the fuel tube. The apparatus is shown below in Figure 4. The small, but strong spark was plenty to ignite the burner repeatedly and reliably.



**Figure 4: Ignition system schematic**

## **2.5 Metering Fuel Flow and Pressure**

### **2.5.1 Flow Meters**

Two ports existed in the bottom flange of the pressure vessel, stainless steel fittings and plastic fuel line carried the fuel and the air into the burner from these two ports. Two more ports existed, as mentioned before, in two of the window flanges of the pressure vessel. These ports provided only air to the window ports to help prevent soot

and condensation from building up on the windows and preventing optical and diagnostic access.

For all air access, into the pressure vessel, for the burner co-flow and the window ports, stainless steel, high pressure, braided fuel line was used. These stainless steel lines supplied air from four tanks of dry, compressed, hydrocarbon-free air with a purity level of 99.95%. This air ran from the tanks into a two-stage high pressure regulator which was diverged into two branches, each of which going to a separate Hastings mass flow meter (Model 201). Each of these flow meters were calibrated with nitrogen for the range of 0 to 100 standard liters per minute (SLPM).

Stainless steel, high pressure, braided line was also used for all fuel and diluent access into the pressure vessel. The lines for the fuel and diluent individually each ran from one bottle, as their flow rates were much less than that of air, and into a separate Hastings mass flow meters (Model 200). Each of the flow meters was calibrated with nitrogen from 1 to 1000 standard cubic centimeters (sccm). Each fuel (methane or ethylene) or diluent (nitrogen, argon, helium, and carbon dioxide) used were at least 99.0% pure.

Each of the four Hastings flow meters that were used was powered by a Hastings power supply (Model 40). This power supply not only provided power, but also provided a digital read-out of the flow rates of each meter. However, since each of the flow meters was calibrated using nitrogen, a correction factor had to be taken into account for each species other than nitrogen. These corrections were referred to as gas correction factors and were provided by the manufacturer for air, ethylene, methane, argon, helium, and carbon dioxide. The gas correction factors were 0.998, 0.604, 0.770, 1.430, 1.430, and

0.73, respectively. In some instances a bottle of fuel diluted by volume from the factory was used in experimental investigation. When this instance arose, the following equation was used to calculate the correction factor to be used.

$$\left[ \frac{1}{\left( \frac{\%volume}{fuel\_correction\_factor} \right) + \left( \frac{\%volume}{diluent\_correction\_factor} \right)} \right]$$

### 2.5.2 Elevating Pressure

Achieving elevated pressure in the vessel was time consuming, and very sensitive. Building pressure was based on a simple concept of reactants flowing in verses the amount of products that were allowed to flow out of the vessel. Air and fuel flows from the bottles, being read digitally from the mass flow meter provided the metering of air into the vessel while a needle valve, located on the exhaust port provided the metering of products out of the vessel. The pressure inside the vessel was displayed on a gauge, externally, with a range of 0 to 1000 psig. The mass flux of co-flowing air was increased, while the needle valve on the exhaust port was nearly closed to build pressure. Trying to maintain a velocity matched air to fuel ratio contributed to the time consuming nature of this process. Window air was kept flowing (to prevent soot deposits on the windows) a very minimal flow rate (no more than 1.5 SLPM) to allow for clear optical access, but was considered negligible at pressures where it was needed, with consideration to the co-flow air rate. One most important aspect of building pressure within the vessel was to be certain never to completely close the exhaust needle valve, as this extinguished the flame very rapidly.

As the pressure increased inside of the vessel it had a few impacts on the appearance of the flame. Since the reaction rates increased in proportion to the pressure, and the diffusion rates increased as the gradient became steeper, the flame became: much thinner, less stable more easily, and shorter. These same findings about pressure effects to the flame were reported by Miller and Maahs (1977), as mentioned previously, by reporting on hydrodynamics disrupting the flame and making it more sensitive to extinction because diffusion of reactants cannot occur quickly enough to restore the flame.

## **2.6 Appearance of Flames**

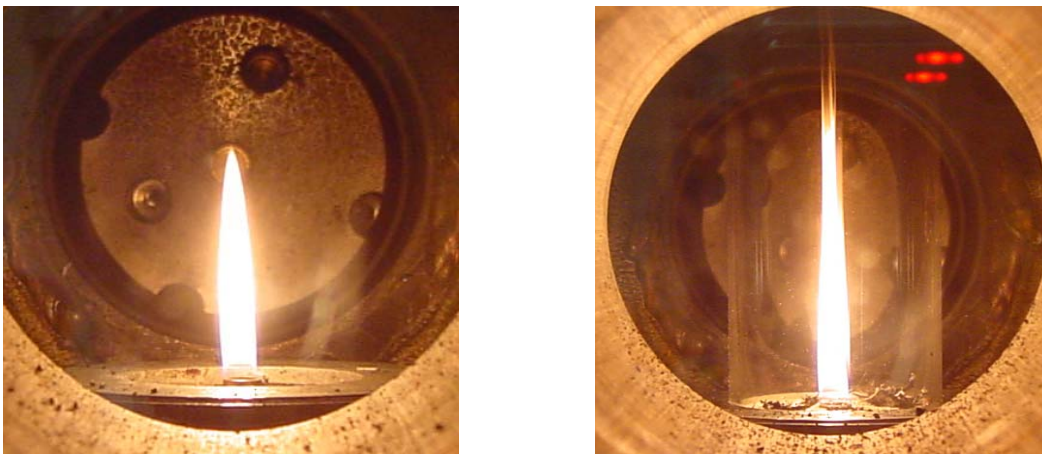
### **2.6.1 Smoke Point Measurements**

Originally efforts were made to keep all flames having a velocity matched air to fuel ratio. If a velocity matched environment was achieved then axial components of diffusion could be assumed negligible, meaning only radial direction components of diffusion would need to be considered. Also, it was originally believed that velocity matching in an over-ventilated flame would have an effect on the flame's smoke point that would be more decisive than just assuming a heavily over-ventilated flame condition. The current flames visually appeared to be "candle-like" and thus over-ventilated, however, to be certain that the flame was over-ventilated at all times throughout the experimental investigation, a quick stoichiometric calculation was performed. Since all of the data taken, both at atmospheric and elevated pressure conditions was taken at an air to fuel velocity ratio of no less than 0.5, this velocity ratio was used for calculation. At stoichiometric conditions for ethylene burning in air there was a 14.28:1 air to fuel mole



ratio. With the burner configuration at atmospheric conditions, and an air to fuel velocity ratio of 0.5, if there had been 100 sccm of fuel, there would need to be greater than 1428 sccm of air to make the justification for an over-ventilated flame. For the current configuration there was 3570 sccm of air for 100 sccm of fuel for a 0.5 air to fuel velocity ratio. The same calculation was performed for the elevated pressure burner configuration and it was found to also yield an over-ventilated flame. Therefore it was safe to assume that all of the current data, both atmospheric and elevated pressure conditions were taken in an over-ventilated flame, and most of this data was taken at velocity matched conditions; however, some data was taken specifically at air to fuel velocity ratios between 1.5 and 3 to compare the actual experimental effects of having a velocity ratio of one verses that of one greater than one. These effects were considered at atmospheric as well as elevated pressure conditions, and did have effects on the smoke points (discussed later in § 3.1.2 and 3.2.2).

There is a distinct difference in a diffusion flame that is just at its smoke point and one that is slightly above its smoke point. The difference is very clear to see without use of any complicated laser or optical set-up. Figure 5 shows two images;



**Figure 5: Flames at and above their smoke points**

the image on the left is an undiluted ethylene flame at 2 atmospheres, at its smoke point, with a volumetric fuel flow rate of 76 sccm, an air flow rate of 16.6 SLPM, and a smoke point height measured to be 36.8 mm. The image on the right is also an undiluted ethylene flame at 2 atmospheres, just slightly above its smoke point with a volumetric fuel flow rate of 98 sccm, and air flow rate of 21.7 SLPM. It is very obvious that the flame imaged on the right is smoking. Not only is there visible smoke being emitted, but the feathering of the flame tip is also an indicator of it being above its smoke point.

### **2.6.2 Smoke Point Data Collection**

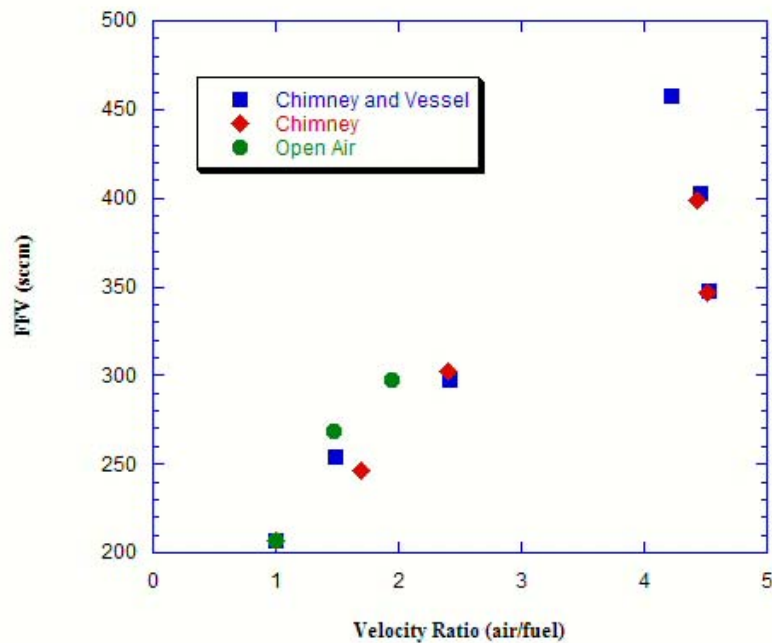
Initially, the smoke point data was to be collected using a laser and optical set-up, similar to that of an extinction measurement set-up. A helium-neon (HeNe) laser was utilized to create a beam to be split in two. The beams were to pass into the pressure vessel optical port with one beam going through, but slightly above, the plane of the flame, while the other beam going just to the side, and slightly above, the plane of the flame. The beams then pass through the other optical port to exit the pressure vessel and into photodiodes. Each of the photodiodes was connected to an oscilloscope to read intensities of the laser beams. Knowing the ratio of the two beam's intensities for a flame that is not emitting smoke would have been the baseline for comparison. Once smoke started to form, the laser beam would scatter off and be absorbed by the soot particles and the intensity of the beam going through, but slightly above the plane of the flame would decrease, therefore predicting the presence of smoke, and thus the flame's smoke point.

After a great deal of effort in the experimental set-up with the HeNe laser and photodiodes, it was found that the set-up was too sensitive to light. As the images above

show, the appearance of smoke from the flame is a very distinguishable characteristic, and does not need a laser extinction measurement set-up to predict accurate smoke points. Therefore the following data was collected through visual observation methods, looking for signs of visible smoke and feathering of the flame tip to provide an accurate position of smoke point.

### 2.6.3 Smoke Point Data Apparatus Determination

In the initial phases of data collection, the methodology to be used was questioned for atmospheric data collection. The options for set-up, for atmospheric data, included running the tests in: (1) open air, (2) with the quartz chimney, or (3) with the quartz chimney inside of the pressure vessel. From above it is known that the atmospheric data was taken with a quartz chimney inside of the pressure vessel with the exhaust valve completely open. This option was chosen after a simple test of velocity ratio (air/fuel)



**Figure 6: Undiluted ethylene at 1 atm to determine testing apparatus**

verses the volumetric fuel flow showed very little difference in the effects on the smoke point of each of these set-ups, as shown in Figure 6. From the plot it is easy to see that for velocity ratios of less than three, the smoke point is insensitive to the experimental apparatus. However, because of outside air perturbations, it was impossible to run higher volumetric fuel flow rates, or air to fuel ratios, without the chimney or vessel. In trying to maintain a constant environment throughout all data collection, both atmospheric and elevated pressures, it was determined that working with the quartz chimney inside of the pressure vessel was in the best interest.

#### **2.6.4 Smoke Point Data Reduction**

Images of the flames, at their smoke points were taken with a digital camera on a still mount. These images were then examined with computer software, MGI PhotoSuite, to determine the number of pixels present from the base to the tip of the flame. With a baseline image of a ruler instead of a flame, it was possible to determine the number of pixels per millimeter for a given optical magnification. With a simple calculation it was possible to determine the actual height of each flame, at its smoke point, in millimeters. For the majority of all images taken, the camera was mounted in such a way as to have 5.4 pixels per millimeter; thus the minimum uncertainty in the flame height at the smoke point is 0.2 mm.

### **3 Smoke Point Measurements**

Smoke point data was taken in several different configurations. As mentioned previously, ethylene and methane were tested. In addition to testing these pure fuels, these fuels were tested individually with four diluents: nitrogen, argon, helium, and carbon dioxide. These fuels were tested at dilution levels up to 40% by volume for each diluent. Also, two bottles of each ethylene and methane specialty gases were premixed using 20% and 40% nitrogen by volume. Each of these pure and diluted fuels were tested in atmospheric and elevated pressure conditions.

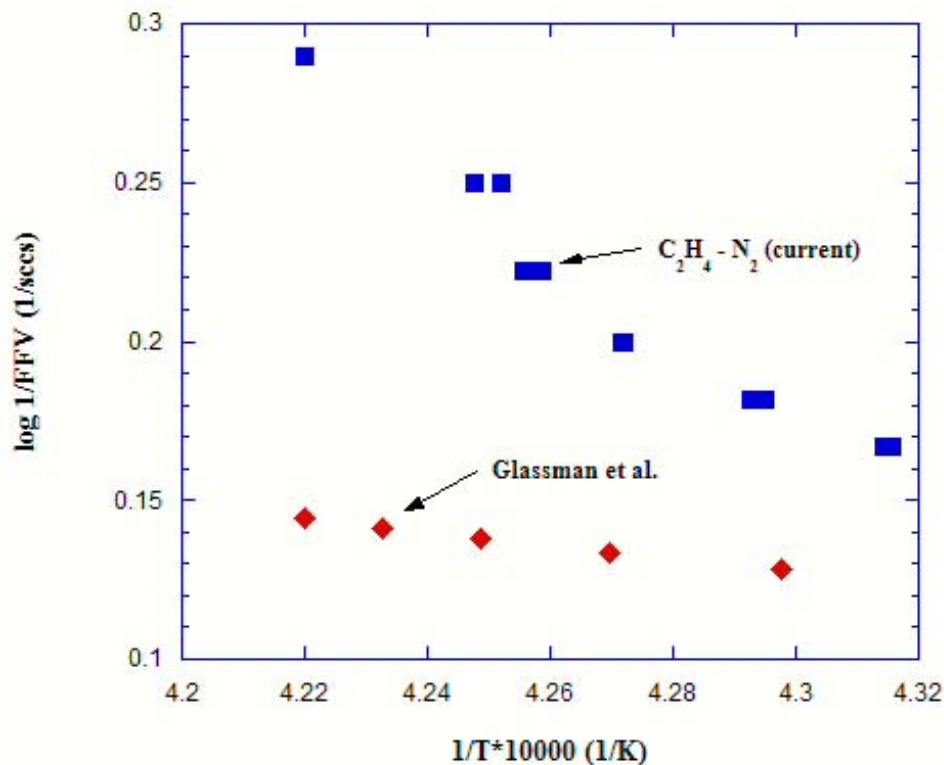
In most instances of data collection, velocity matched air to fuel were maintained. However, in an effort to see how changes in velocity ratio affect the smoke point, varying velocity ratios were also investigated.

#### **3.1 Ethylene Data at Atmospheric Pressure**

##### **3.1.1 Nitrogen Diluted Ethylene Justification**

Researchers in the past have worked with the sooting tendencies of different fuels and the effects that nitrogen dilution has on these fuels (Gomez et al., 1984; Glassman & Yaccarino, 1981); however, they have approached their experiments in slightly different manners from the current research. Instead of using flames that are just classified as “highly over-ventilated” laminar jet diffusion flames, as were used by Glassman and co-workers, the current research uses an over-ventilated, velocity matched, co-flowing laminar jet diffusion flames.

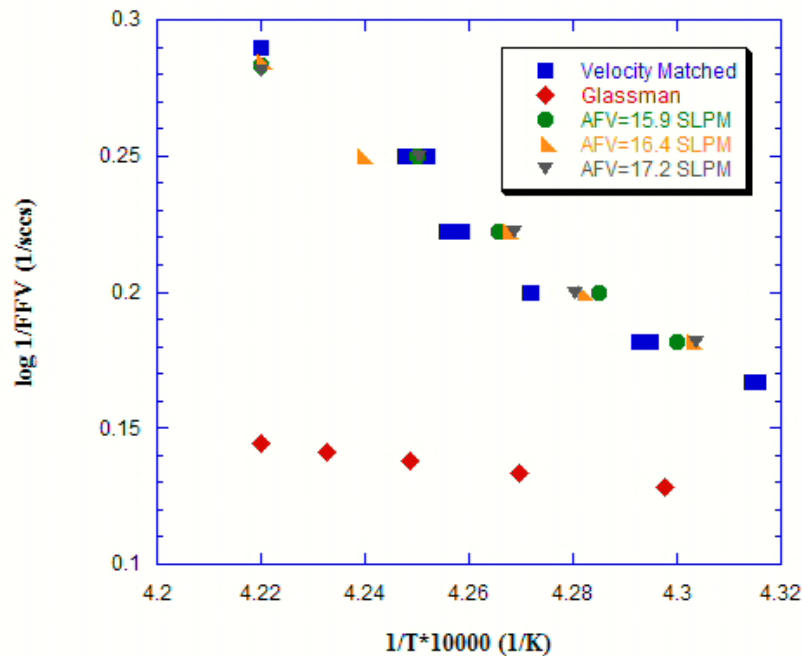
Testing began with ethylene at atmospheric conditions, and an undiluted smoke point was measured. Smoke points were then measured as a function on nitrogen dilution up to 40% by volume. Upon running these diluted cases and reducing data, the results were compared to diluted cases of ethylene completed by other researchers (Yaccarino, 1981; Glassman, 1988). Temperatures for each of the diluted cases were calculated using a website created by graduate students at Colorado State University ([www.grashof.engr.colostate.edu/tools/SoftwareTools.html](http://www.grashof.engr.colostate.edu/tools/SoftwareTools.html)). The data was plotted as the log inverse volumetric fuel flow ( $\log 1/\text{FFV}$ ) verses the inverse temperature ( $1/T$  in degrees K) multiplied by 10,000, along with the results of Glassman and co-workers and is shown below (Figure 7)



**Figure 7: Smoke point fuel flow rates for nitrogen diluted ethylene flame at 1 atm as a function of temperature (through dilution)**

The points for Glassman's data were extrapolated from previous plots published with his work, allowing only a few points to be plotted. However, the current research ran the nitrogen diluted ethylene data set twice in an effort to assume repeatability of points.

In an attempt to decipher the differences in slope between the current data and the previously published data by Glassman and co-workers, two hypotheses were derived. The difference in slope could be caused by burner geometry or the air to fuel velocity ratio used by Glassman. Glassman's data was only reported as a "highly over-ventilated" flame, not distinguished as velocity matched or non-velocity matched, meaning that an over-ventilated, velocity matched flame might yield a different slope from the published data's slope. In order to determine if this was a reasonable hypothesis, the nitrogen diluted data was re-run using several different, but constant, air co-flow rates (Figure 8).

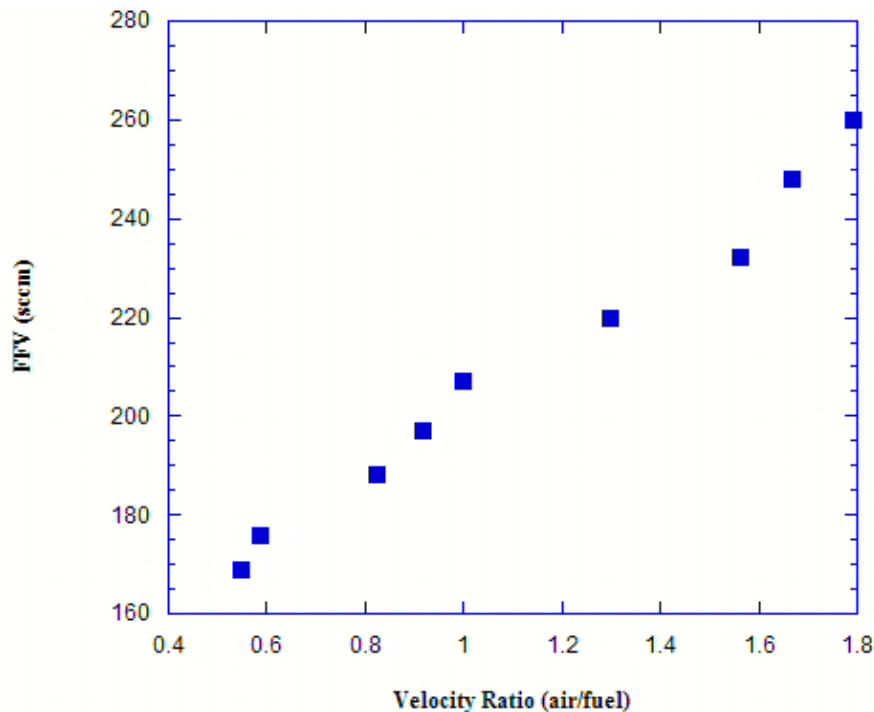


**Figure 8: Smoke point fuel flow rates with constant air flow rates for nitrogen diluted ethylene at 1 atm as a function of temperature**

From the plot it was apparent that the hypothesis that the slopes of the current data and Glassman and co-workers data were different because of the effect of having velocity matched volumetric air co-flow was not valid. This led to the conclusion that the difference in slope is caused by burner geometry. However, it was decided that the slopes were similar enough to assume the current data, and method of obtaining this data was accurate and data collection continued for atmospheric testing.

### 3.1.2 Undiluted Ethylene

In a further effort to understand the effects of non-velocity matched air to fuel ratios on smoke point, undiluted ethylene was tested, at 1 atmosphere, with varying volumetric fuel flow rates and velocity ratios. The resulting data was then plotted as volumetric fuel flow verses air to fuel velocity ratios ranging from 0.5 to 1.8 (Figure 9).



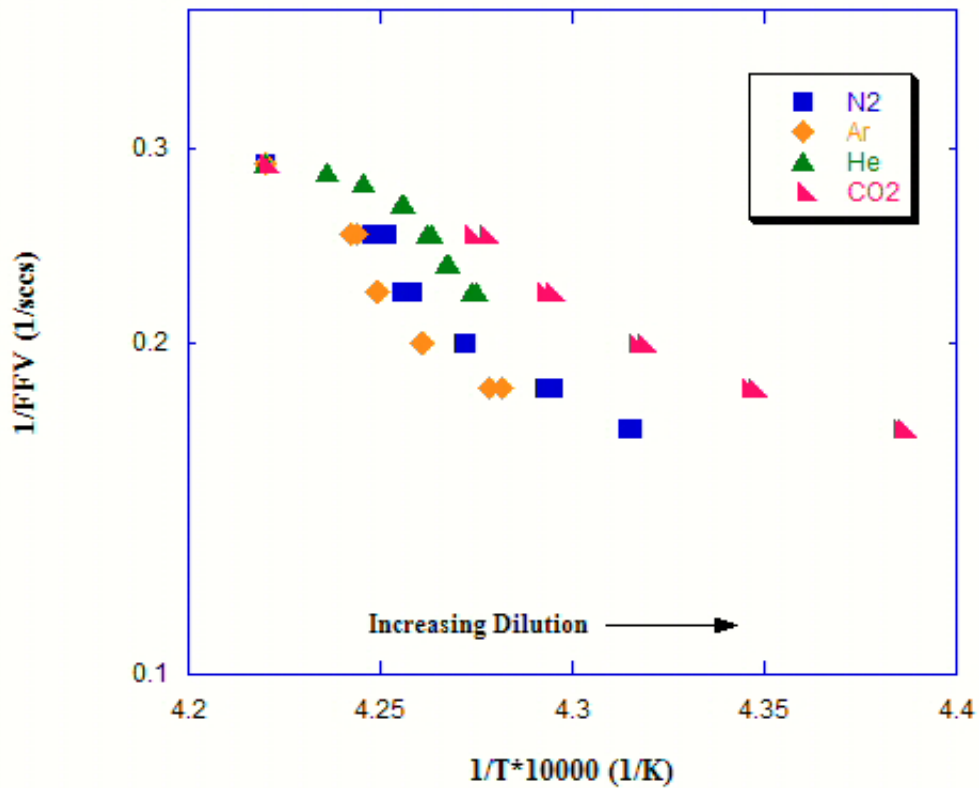
**Figure 9: Smoke point fuel flow rate for undiluted ethylene at 1 atm as a function of velocity ratio**



The data showed a relatively linear relationship between volumetric fuel flow rate and an increase in air to fuel velocity ratio.

### 3.1.3 Diluted Ethylene

Once the current research with ethylene, both pure and nitrogen diluted, with velocity matched air to fuel ratios, proved itself to be in agreement with other published data the experimental investigation continued with diluents other than nitrogen. Argon, helium, and carbon dioxide were tested, mixed in ethylene, up to 40% dilution by volume. This data was then plotted to check for noticeable trends (Figure 10).



**Figure 10: Smoke point fuel flow rates for ethylene diluted individually with four diluents at 1 atm as a function of temperature**

The data shown above were taken twice for each diluent as a means to assure repeatability. Each of the diluents seemed to follow the same trend, but because of differences in the chemical composition of each diluent, the trends were slightly offset from one another, as expected. It should also be noted that the undiluted point, at which each of the diluent trends begins (approximately  $1/\text{FFV}=0.29$  1/sccs), matches the velocity matched case seen in Figure 9. The current trends, and slight differences in offset, between each diluent's effect on the ethylene flame is believed to be an result of the fuel Lewis numbers; however, more research is needed to verify this hypothesis.

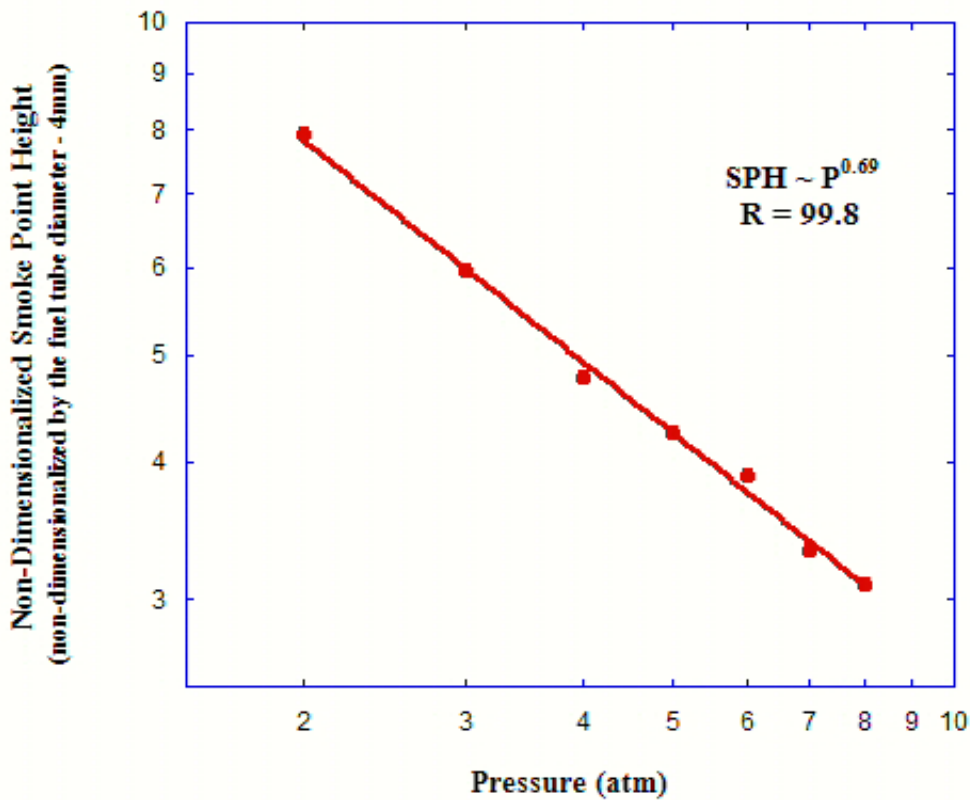
### **3.2 Ethylene Data at Elevated Pressures**

Smoke point data at elevated pressures has had very little attention from researchers in the combustion field. To the author's knowledge there is no published experimental data on pressures greater than atmospheric, where fuel and air flow rates were collected, and only a small amount of research focusing on sub-atmospheric conditions (Sunderland et al., 1994) recorded these flow rates. The published data at pressures other than atmospheric (0.1 to 1.0 atm) has not focused on the effects of dilution.

This research had the opportunity to focus on ethylene flames, pure and diluted individually with four diluents, at not only atmospheric, but also at elevated pressures ranging from 2 to 8 atmospheres. With the current burner and pressure vessel configuration, the ethylene flame, either pure or diluted, was tested only to eight atmospheres. This is due mainly to the flame's height above the burner at pressures greater than eight atmospheres, which is roughly 1 to 1.5 times the fuel tube diameter.

### 3.2.1 Undiluted Ethylene

One of the most important aspects of the data collected was the effects that elevating pressure had on the smoke point. Since this research has not been investigated, it was decided that data would be taken with the same procedure as atmospheric data. Data was taken with undiluted ethylene in one atmosphere increments from two to eight atmospheres. The resulting data was plotted (Figure 11) and showed a steady trend.

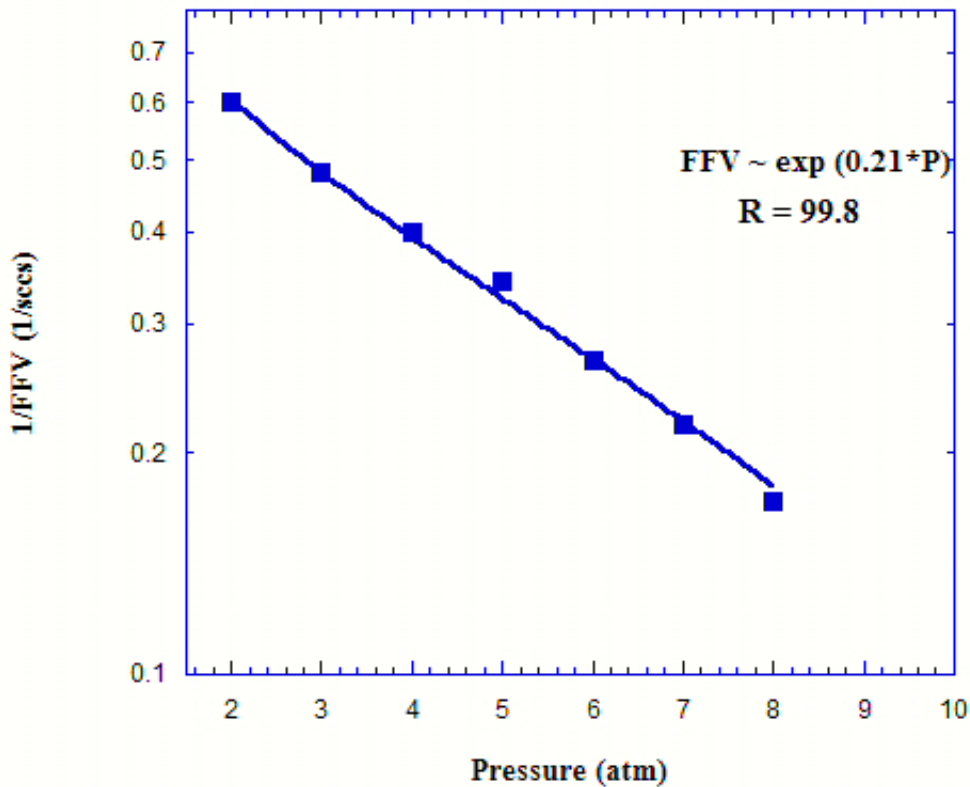


**Figure 11: Non-dimensionalized smoke point heights of undiluted ethylene flames at elevating pressures**

For the plot, the smoke point was non-dimensionalized by the fuel tube diameter to give a more consistent measurement as to not be burner specific. The above data was plotted in

log-log space, with the data fitting a power law such that smoke point height scales as pressure to the 0.69, with a residual of 99.8.

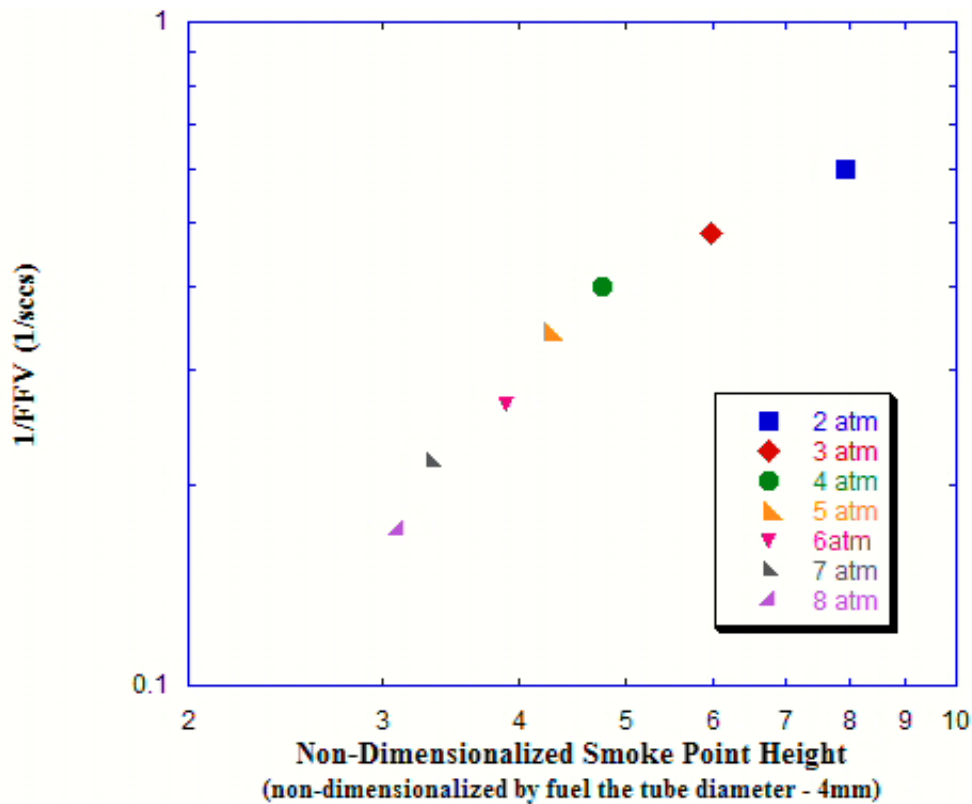
Originally it had been assumed that volumetric fuel flow rate and a flame's smoke point scaled linearly with one another. In order to decide the relevancy of this assumption, another plot was generated (Figure 12).



**Figure 12: Volumetric fuel flow of undiluted ethylene at the flame's smoke point for elevating pressures**

In this plot, in order to find the data's trendline, the inverse volumetric fuel flow was plotted as a function of pressure in log-linear space. The data followed a logarithmic trend with volumetric fuel flow scaling as the exponential of 0.21 times pressure. This data was then compared to the plot of smoke point as a function of pressure (Figure 11).

There were noticeable differences in the trends of the two plots. This established that the smoke point of the flame and the volumetric fuel flow rates of the flame did not have an exact linear relationship with one another. Therefore, a third plot was created (Figure 13) to find the relationship between these two important parameters of the flame.



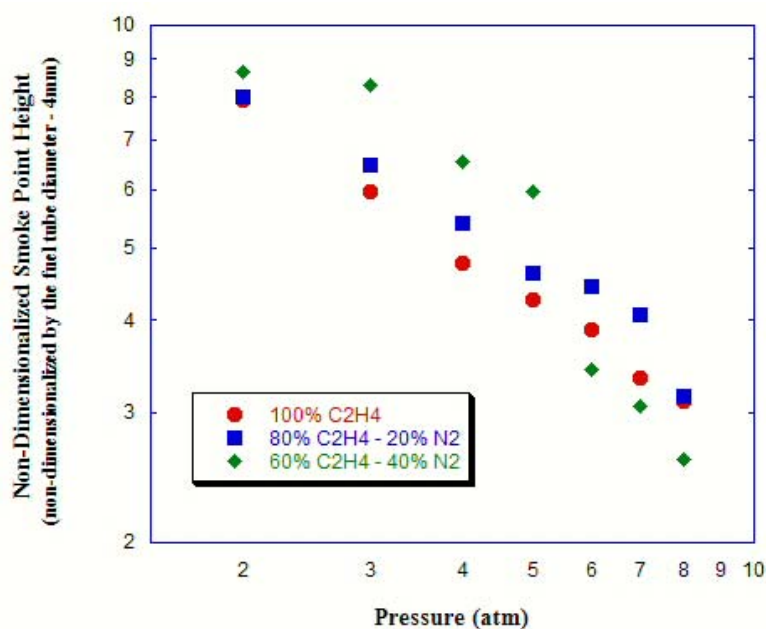
**Figure 13: Relationship between the smoke point height and volumetric fuel flow for undiluted ethylene flames at elevating pressures**

Plotting inverse volumetric fuel flow versus non-dimensionalized smoke point height in log-log space did however yield a relatively linear trend. Once satisfied with the undiluted ethylene data at elevated pressures, it was necessary to move on the nitrogen diluted ethylene.

### 3.2.2 Nitrogen Diluted Ethylene

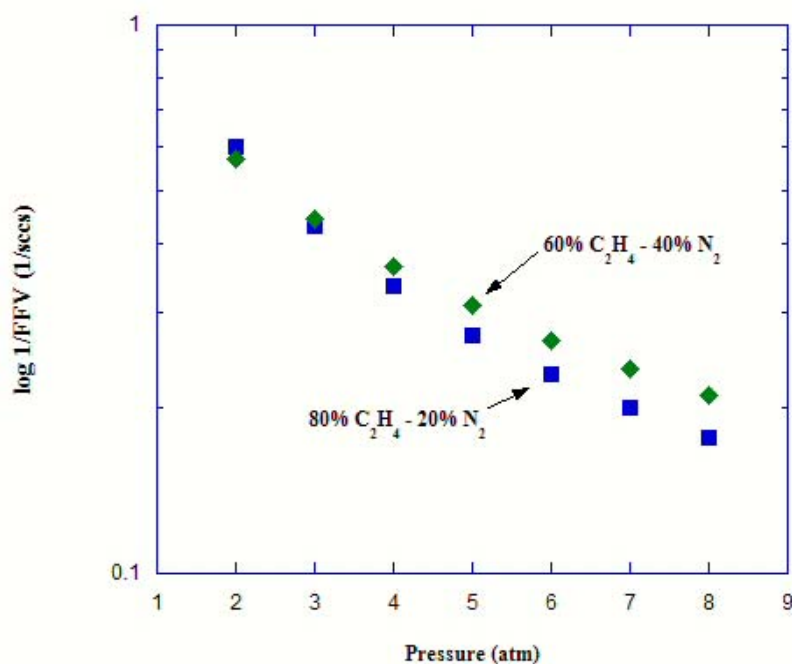
The next step involved diluting ethylene with nitrogen. Instead of mixing the fuel and diluent in the laboratory, bottles of premixed ethylene, diluted by nitrogen, were purchased. This made the data taking process more convenient, and also gave a couple of reference points to make sure our “in house” mixing of fuel and diluent was a reliable procedure. The bottles of premixed diluted fuel were: (1) 80% ethylene with 20% nitrogen and (2) 60% ethylene with 40% nitrogen, and had a purity rating of at least 99.0%.

With these bottles, the effects of pressure were again tested from two to eight atmospheres. Once this data had been collected, it was plotted as the non-dimensional smoke point against pressure (Figure 14). It was also plotted with the undiluted ethylene, elevated pressure data, shown previously, for comparison.



**Figure 14: Smoke point heights of nitrogen diluted ethylene at elevating pressures**

It should be noted that the 60% ethylene, 40% nitrogen data points seem to show higher smoke points than the other two fuels, as it was expected, until after five atmospheres of pressure. This behavior is troubling; however it should also be noted that an error is possible in measuring smoke points at pressures nearing eight atmospheres. As mentioned earlier, the flame heights as ethylene reaches eight atmospheres came to be on the order of 1 to 1.5 time the diameter of the fuel tube. Therefore, there is some ambiguity over the data points collected at six, seven, and eight atmospheres due to the shortness of the flames. Therefore a plot of the volumetric fuel flow as a function of pressure was created (Figure 15). This was considered of interest as the fuel flow rates collected, using the mass flow meter, were more accurate than the measured heights of the flames.



**Figure 15: Fuel flow rate of smoke points for nitrogen diluted ethylene as a function of increasing pressure**

This plots shows the 40% nitrogen diluted ethylene flames require greater fuel flow than that of the 20% nitrogen diluted ethylene. The above plot also proves that small errors could exist in the measurements of the flames at their respective smoke points. As expected, the plot does not show the two data paths crossing at or around the five atmosphere point.

Because the data in Figure 14 and Figure 15 was taken at velocity matched air to fuel ratios, it became important to see how air to fuel velocity ratios affected the smoke points of these flames at elevated pressures, with dilution. For that reason, experimental investigation was completed for velocity ratios ranging from 1.3 to 2.9. These experiments were tested with each bottle of nitrogen diluted ethylene and each at four and eight atmospheres of pressure. This data was then plotted as volumetric fuel flow against velocity ratio (Figure 16).

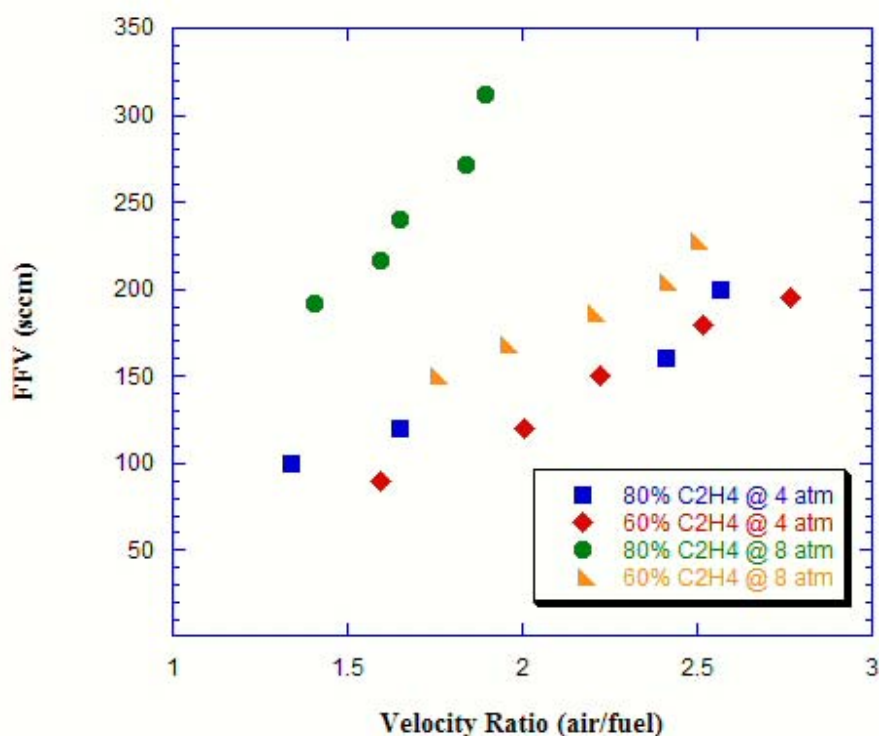
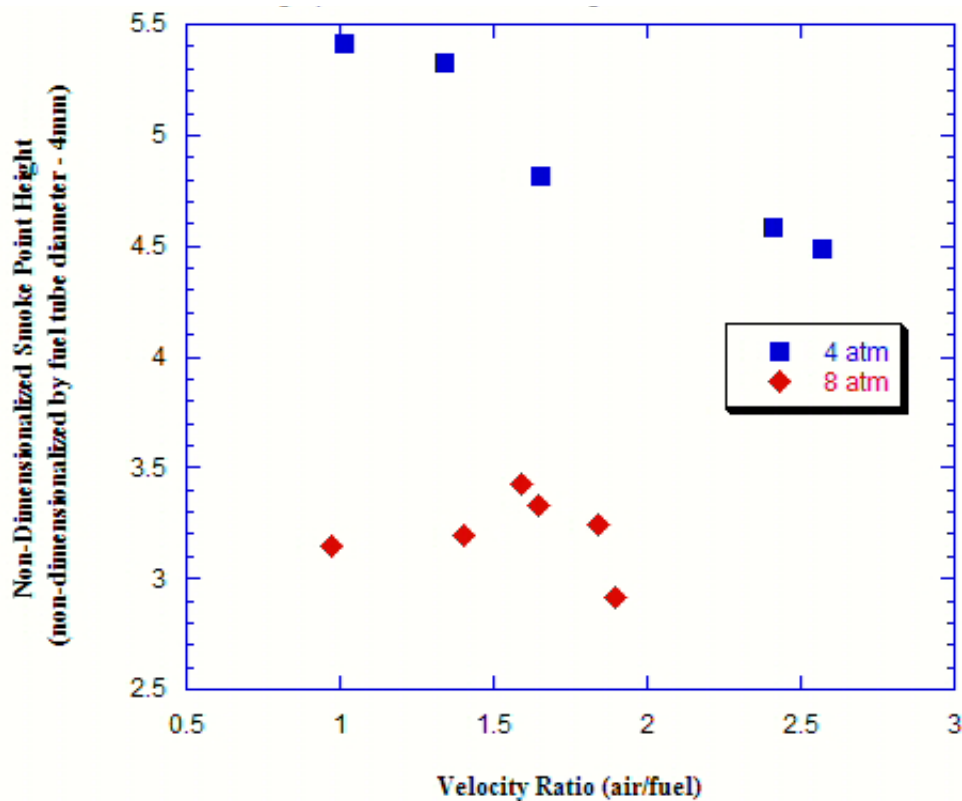


Figure 16: Diluted ethylene at varying dilution and pressure



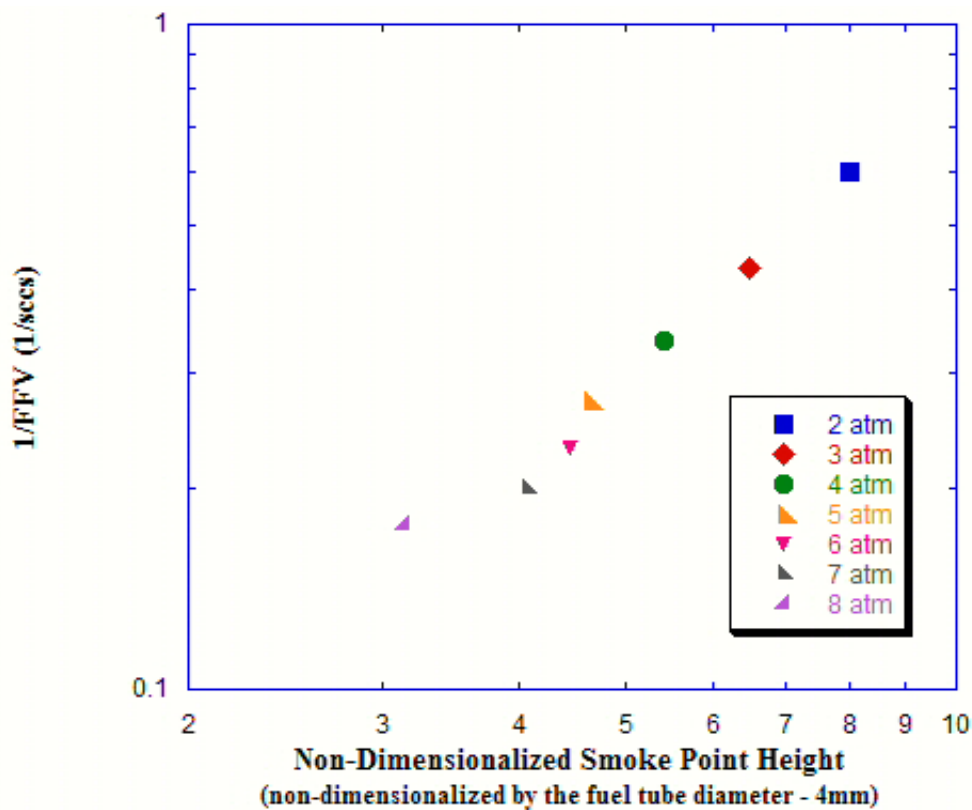
This plot, similar in trend to the plot shown earlier in Figure 9, of velocity ratio verses volumetric fuel flow of undiluted ethylene at one atmosphere, behaves as expected. The data taken at eight atmospheres require higher fuel flow rates, and seemed to have slightly steeper slopes. From Figure 16 it appeared that the higher the pressure, the more sensitive the flame was to velocity ratio.

With the trends shown in Figure 16, it was questioned as to how the smoke point height is affected at these dilution levels and elevated pressure. Therefore the 80% ethylene, 20% nitrogen bottle data was analyzed a bit further and showed how smoke point height was affected by these changes in velocity ratio (Figure 17).



**Figure 17: Smoke point heights of diluted ethylene as a function of velocity ratio**

From this plot, it was obvious that the actual smoke point of the flame had been affected only slightly by the change in the air to fuel velocity ratio, whereas the volumetric fuel flow seemed to be more sensitive. This is substantially different from the trends shown by plotting the volumetric fuel flow against the velocity ratio. In order to establish a reason to explain this difference it became necessary to find the relationship between smoke point height and volumetric fuel flow at elevating pressure for the 20% nitrogen diluted ethylene. Therefore, similar to the plot seen earlier for undiluted ethylene at pressure, the volumetric fuel flow was plotted as a function of smoke point (Figure 18).

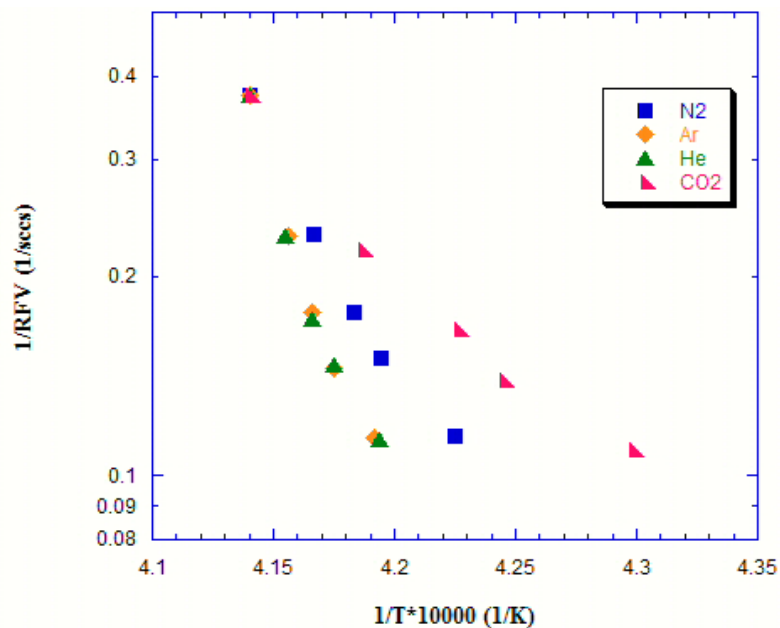


**Figure 18: Relationship between smoke point height and volumetric fuel flow of 20% nitrogen diluted ethylene at elevating pressures**

The above plot showed a slightly different trend in slope than previously shown in the Figure 13 plot for the undiluted case at elevating pressure. It is believed that this slight difference in slope can be attributed to the dilution by nitrogen. The data does show a somewhat linear trend, in log-log space, with the exception of the eight atmosphere case. The point at eight atmospheres was difficult to achieve as the flame was on the order of two to three times the fuel tube diameter, and therefore has the lowest confidence.

### 3.2.3 Diluted Ethylene

After analyzing the data for ethylene diluted by nitrogen, the spectrum was broadened to encompass other diluents. As before ethylene was diluted individually with nitrogen (a non-premixed bottle), argon, helium, and carbon dioxide. This data was collected and plotted (Figure 19) as the inverse of volumetric reactant (including the fuel and diluent) flow versus inverse temperature.



**Figure 19: Smoke point fuel flow rates for ethylene diluted individually with four diluents at 4 atm as a function of temperature**

The data was taken at a constant pressure of four atmospheres with velocity matched air co-flow. This plot appears very similar to the previously shown plot of ethylene diluted individually with these four diluents taken at one atm. From this data, it was hypothesized that dilution rate, rather than diluent, is the determining factor in dictating smoke point, and thus residence time is more important to the smoke point of a flame than temperature effects.

### **3.3 Methane Data at Atmospheric Pressure**

Just as smoke point data was taken for ethylene at atmospheric pressures, methane smoke point data at atmospheric conditions was desired. However, in the atmospheric pressure burner configuration taking methane data for smoke point proved to be impossible. It was impossible to achieve a stable, laminar, methane flame that smoked, mostly because of methane's low propensity to soot.

The burner configuration would have needed altering in order for atmospheric methane data to even be possible. Possible alterations to the burner, such as decreasing the fuel tube diameter, might have made the data collection possible. However, it was decided that the limited value of the atmospheric methane data did not warrant the substantial changes in burner geometry necessary to provide this data. Thus, data continued by moving on to methane cases at elevated pressures.

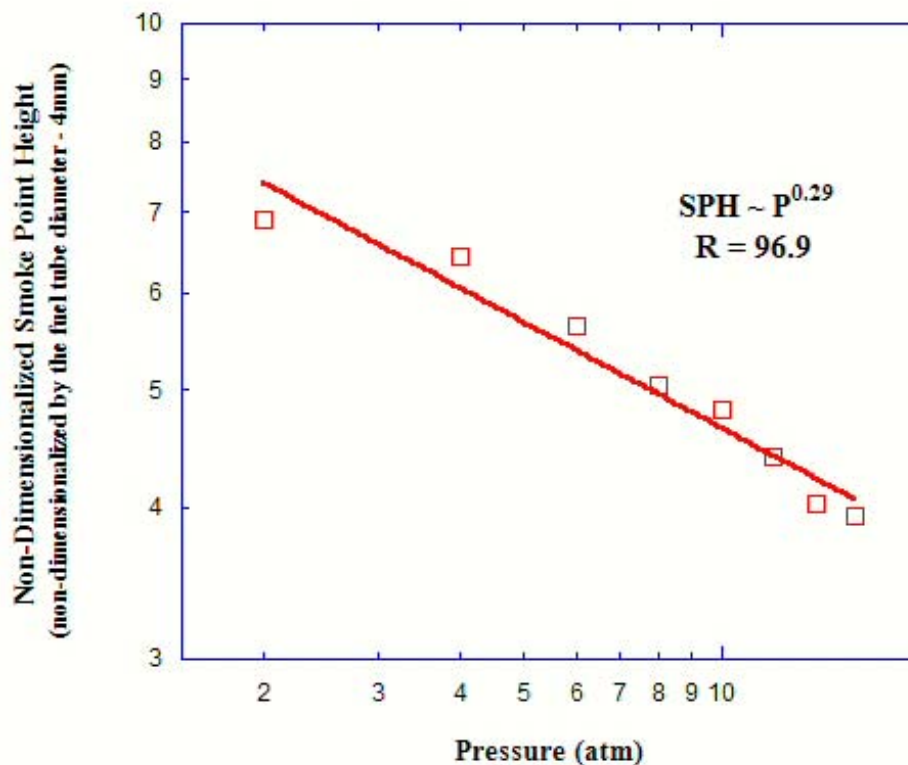
### **3.4 Methane Data at Elevated Pressures**

The research for methane smoke point data was successful at elevated pressures. Similar to the elevated pressure tests completed with ethylene, methane was tested in conditions both pure and diluted individually with four diluents ranging from 2 to 16

atmospheres. Methane, as mentioned previously, was also tested with two individual premixed bottles with nitrogen at 20% and 40% dilution. Just as the ethylene cases were completed to eight atmospheres, the methane data was completed to sixteen atmospheres; at these pressures, the flame's smoke point height was only 1 to 2 times the fuel tube diameter and thus too short to be dominated by soot kinetics.

### 3.4.1 Undiluted Methane

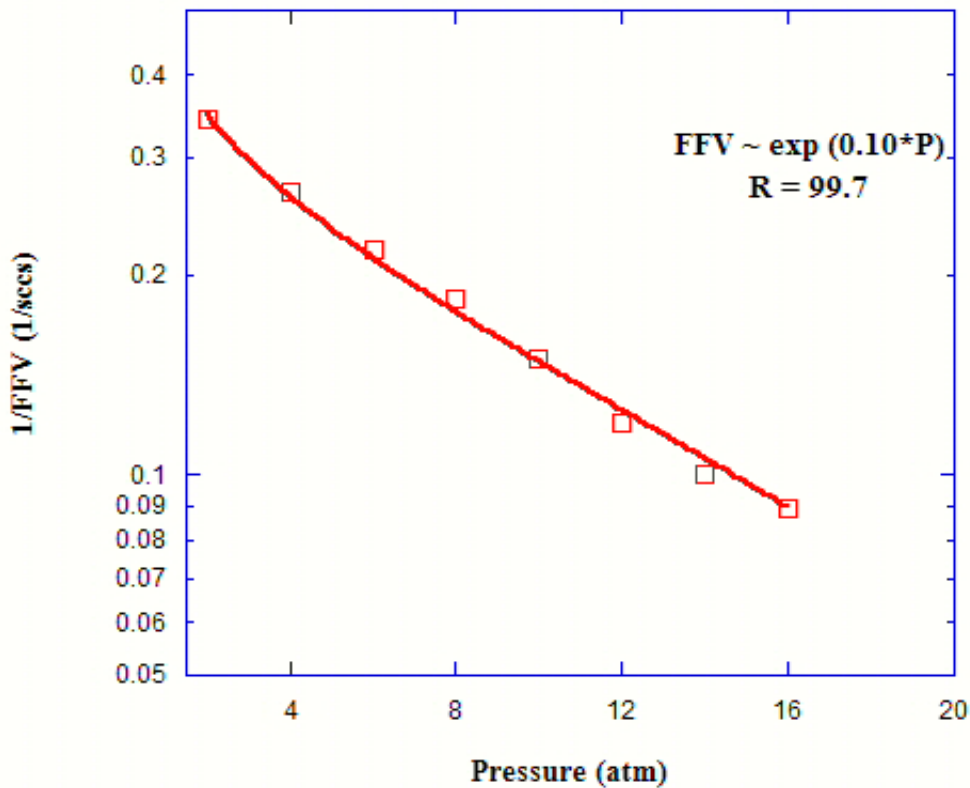
As with the undiluted ethylene data, the elevated pressure data is of utmost importance, and was collected in the same manner. The smoke point was measured from two to sixteen atmospheres in two atmosphere increments to quantify the pressure effect on the smoke point of these flames (Figure 20).



**Figure 20: Non-dimensionalized smoke point heights of undiluted methane flames at elevating pressures**

The smoke point of each flame was measured, giving the smoke point height, and then this height was non-dimensionalized by the fuel tube diameter, 4 mm. The trend, shown above in Figure 20, is very similar to that of the ethylene data shown in Figure 11. By plotting the non-dimensionalized smoke point height verses pressure in log-log space, as done with ethylene, it was possible to fit the data to a power trend such that smoke point height scales as pressure raised to the 0.29 with a residual of 96.9.

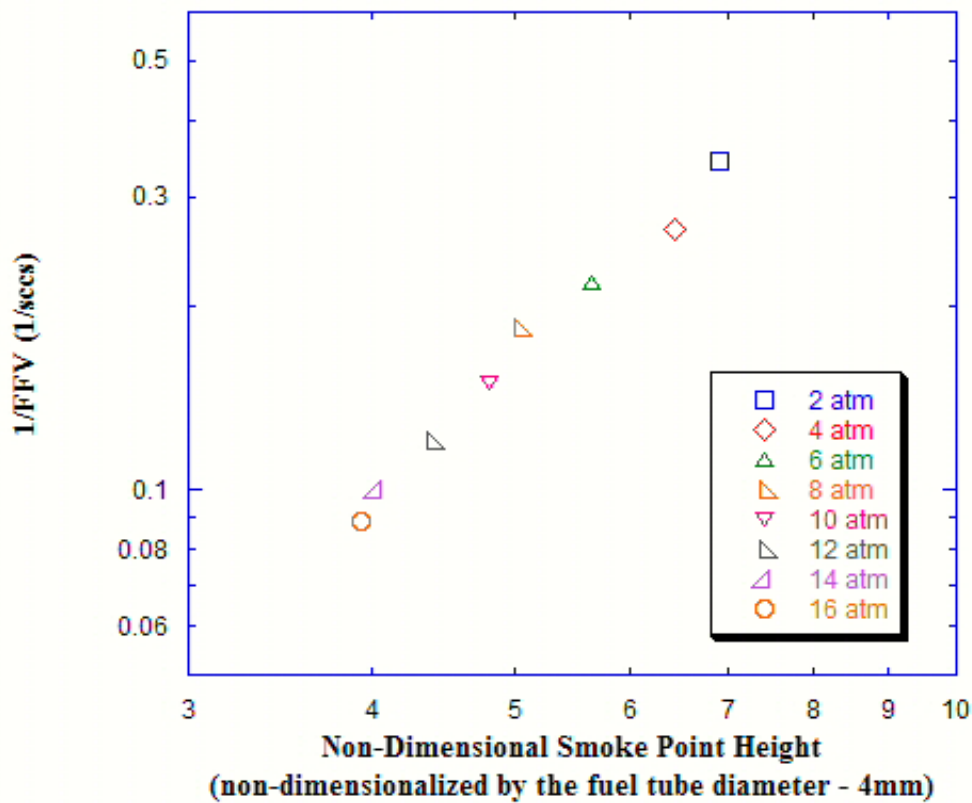
In order to see how the height of the smoke point varies with the volumetric fuel flow it was first necessary to see how the volumetric fuel flow at the smoke point was affected by pressure (Figure 21).



**Figure 21: Volumetric fuel flow of undiluted methane at the flame's smoke point for elevating pressures**

This data showed a very similar trend to that of Figure 20, of undiluted methane, as well as the trend to that of Figure 12, of undiluted ethylene. The inverse of volumetric fuel flow of methane, when plotted in log-linear space, fell into a logarithmic trend such that the volumetric fuel flow scaled as the exponential of 0.10 times pressure with a residual of 99.7.

It was necessary to find the relationship between the volumetric fuel flow and the height of the smoke point for the undiluted methane flames tested (Figure 22).



**Figure 22: Relationship between volumetric fuel flow and smoke point at elevating pressures for undiluted methane**

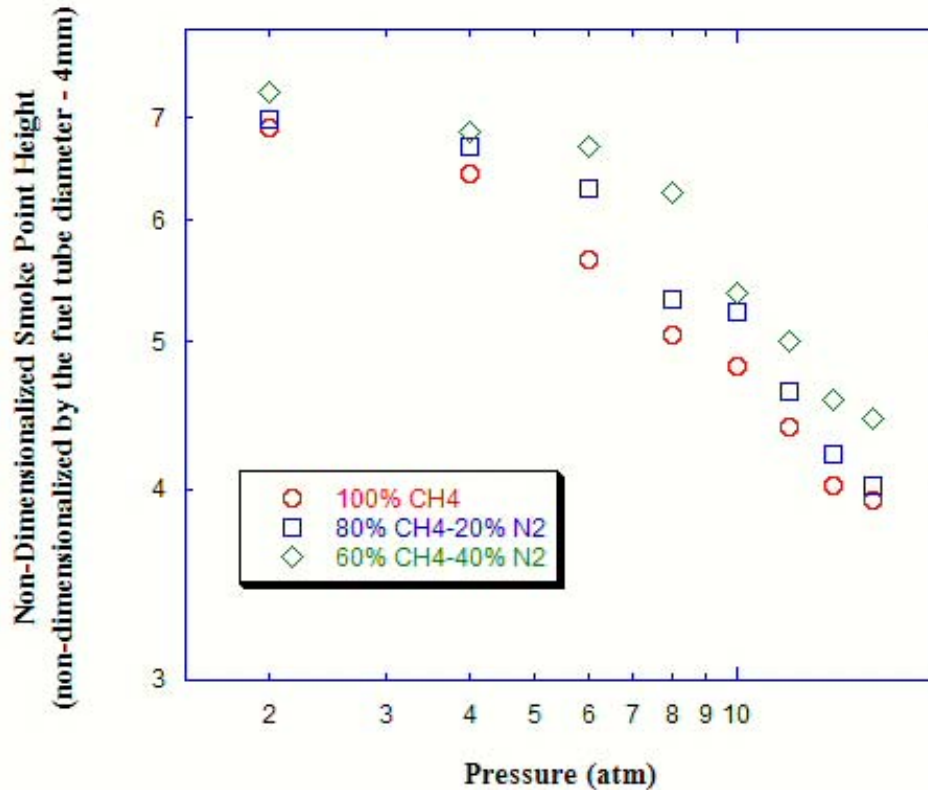
As expected, when plotted in log-log space, this plot was in agreement with the trend on the similar plot (Figure 13) for pure ethylene data at elevated pressures. However, the

data taken for methane does seem to have more of a linear trend than that of ethylene. The two atmosphere data for undiluted methane did seem to be slightly off of the relatively linear trend of the other data. This could be attributed to methane's low propensity to soot, and not being able to create a smoke point in the current burner geometry at atmospheric conditions. Therefore it might also be difficult to find the most accurate smoke point at low pressures such as two atmospheres.

### **3.2.2 Nitrogen Diluted Methane**

As mentioned before, two bottles of premixed nitrogen and methane gas, 20% and 40% nitrogen by volume, were purchased. With these bottles of diluted methane, the effects of pressure were tested again from two to sixteen atmospheres. The data from each premixed bottle and from the undiluted methane cases previously taken were then plotted, in log-log space, as the non-dimensional smoke point height as a function of pressure (Figure 23).





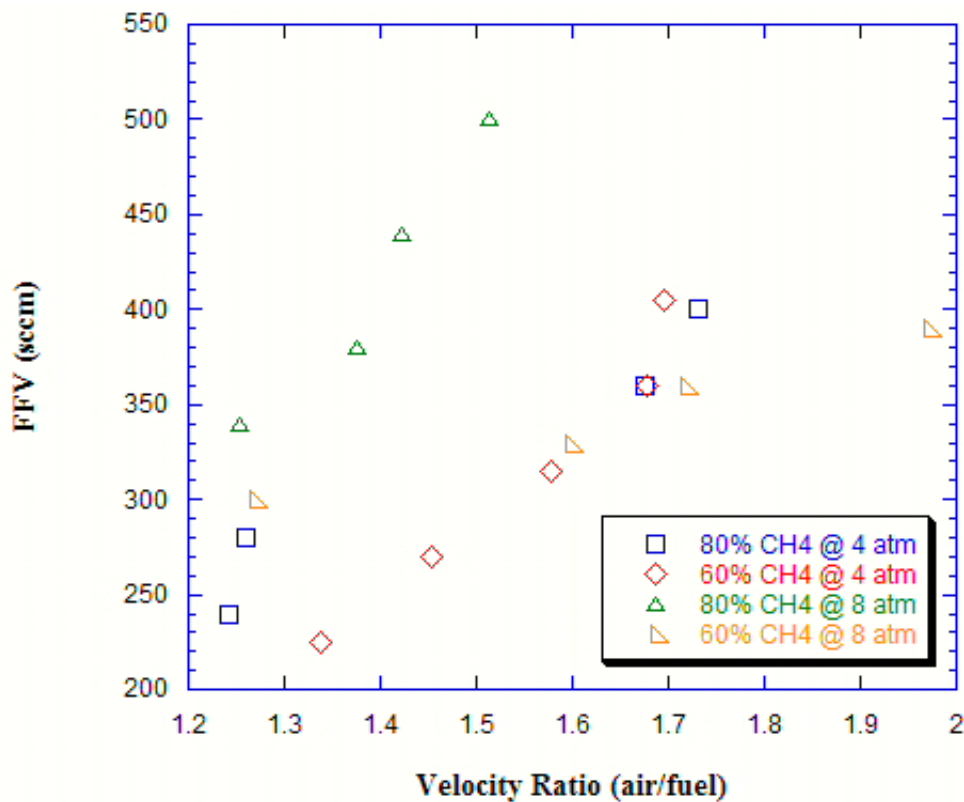
**Figure 23: Smoke point heights of nitrogen diluted methane at elevating pressures**

As expected, the more heavily nitrogen diluted, 60% methane, 40% nitrogen flames had taller smoke points, followed by the 80% methane, 20% nitrogen flames, and finally the undiluted methane flames. This plot behaves as the similar plot, Figure 14, of nitrogen diluted ethylene was expected to behave. The above plot, showed no signs of a change in paths for either of the 20% or 40% nitrogen diluted cases at or around five atmospheres. Again this proved that some error in imaging and measurement of a flame at its smoke point might exist for data shown in Figure 14.

Since the data shown in Figure 23 was all taken at unity air to fuel velocity ratios, it was desired to explore the effects that might have been possible in cases, other than velocity matched, with nitrogen dilution. For this reason, data sets were completed at

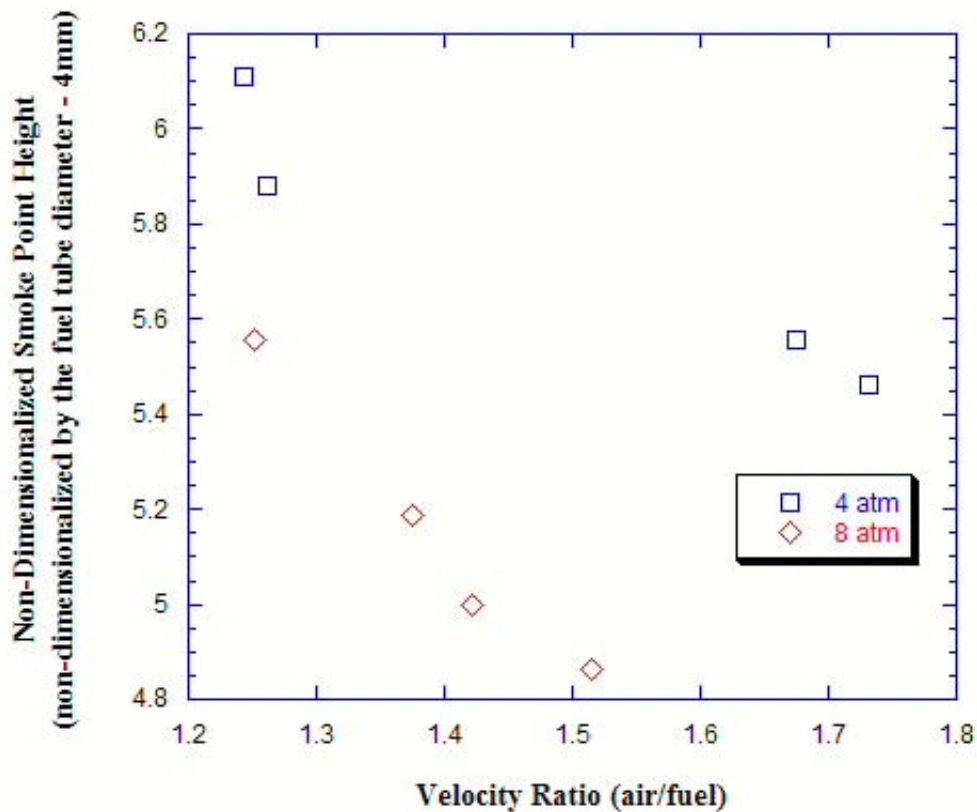
four and eight atmospheres, with each premixed bottle, with velocity ratios ranging from approximately 1.3 to 2.8. This data was plotted below as the volumetric fuel flow against the varying velocity ratio (Figure 24).

The trends of the fuels, although slightly different in trend from that of the ethylene data, shown in Figure 16, still behave as expected. The data taken at eight atmospheres tended to have a greater volumetric fuel flow rates, as anticipated. Also, like ethylene, the 80% methane at eight atmospheres was the most sensitive to increased velocity ratio. However, unlike ethylene, the 60% methane at eight atmospheres was the least sensitive to increased velocity ratio.



**Figure 24: Diluted methane at varying dilution and pressure**

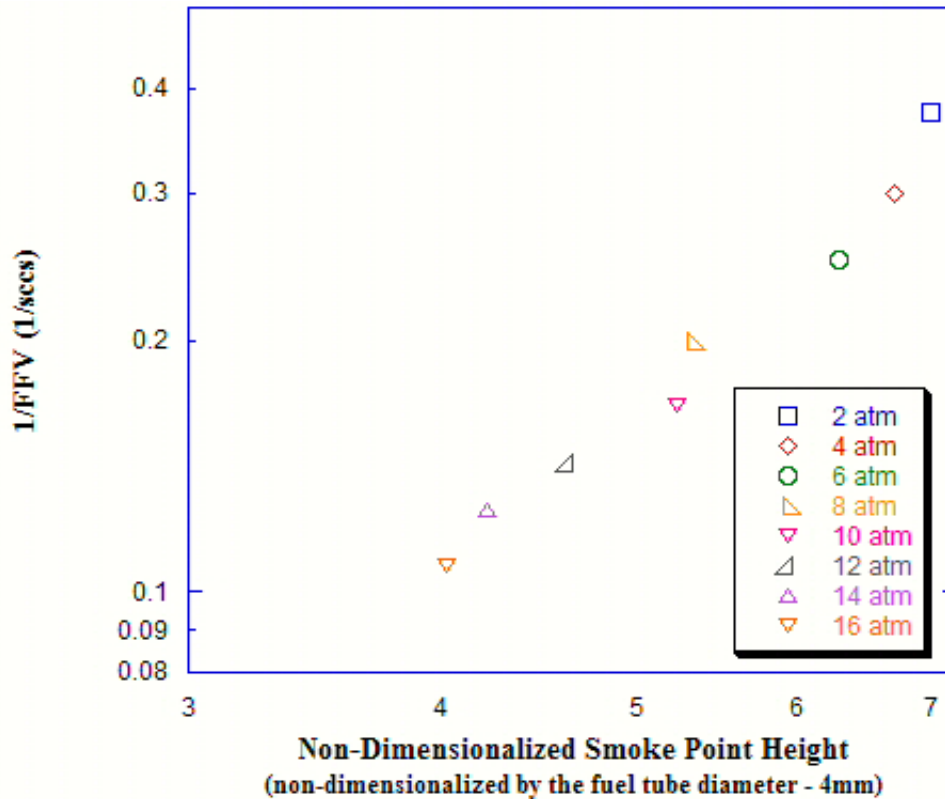
The next step was to see how the heights of the flames at their smoke points were affected by this varying of velocity ratios. In order to study this more extensively, the 80% methane, 20% nitrogen bottle was tested at four and eight atmospheres with the same range in velocity ratio, 1.3 to 2.8, and the flame height was measured at its smoke points (Figure 25).



**Figure 25: Smoke point heights of diluted methane at varying velocity ratios**

This data, although much better behaved, shows a very different trend than that of the ethylene data at similar conditions (Figure 17). The slopes in the above plot for 80% methane, 20% nitrogen, are much steeper than those of 80% ethylene, 20% nitrogen.

In order to find the relationship between the smoke point and the volumetric fuel flow of these nitrogen diluted methane flames it was necessary to plot the log inverse of volumetric fuel flow against the heights of the flames at their smoke points (Figure 26). It was hoped that this plot would give reason to the differences between the previous two plots, and show the relationship in question.

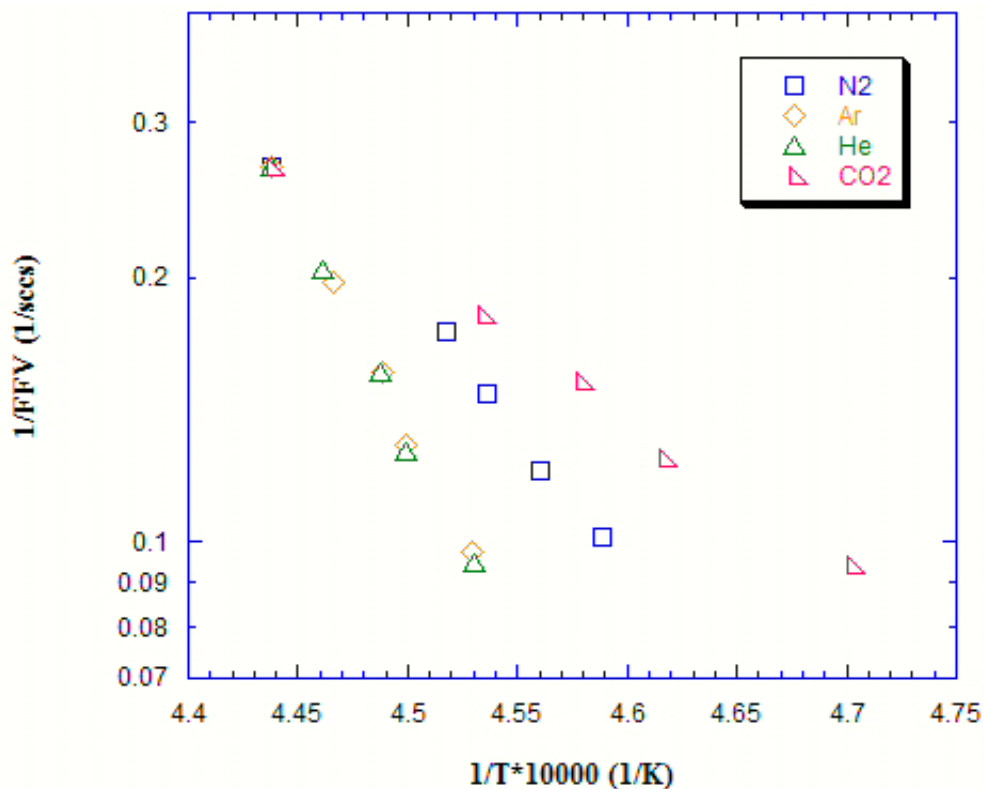


**Figure 26: Relationship between smoke point height and volumetric fuel flow of 20% nitrogen diluted methane at elevating pressures**

This data, plotted in log-log space, was only slightly different than the data of the undiluted methane cases (see Figure 22), which was expected, due to the addition of 20% nitrogen dilution. There was an increase in volumetric fuel flow required, and a slight increase in the height of the flames at their smoke points.

### 3.4.3 Diluted Methane

Upon completion of the undiluted and nitrogen diluted methane cases, other diluents were used to see how each diluent individually affected the methane flame's smoke points. The methane was diluted with each of four different diluents: nitrogen (a non-premixed bottle), argon, helium, and carbon dioxide. Once this data had been collected, it was plotted as the inverse of volumetric fuel flow against inverse temperature at a constant elevated pressure of four atmospheres (Figure 27).



**Figure 27: Smoke point fuel flow rates for methane diluted individually with four diluents at 4 atm as a function of temperature**

This plot appeared to be very similar to that of ethylene data taken under similar conditions (Figure 19). Here again, the trend is very similar to the atmospheric diluted ethylene behavior shown previously (recall that atmospheric pressure methane smoke

point was not collected due to the hydrodynamic instabilities at the necessarily higher fuel flow rates). Again, it was hypothesized that dilution rate, rather than diluent, is the determining factor in this data, and thus residence time is more important to the smoke point of a flame than temperature effects.

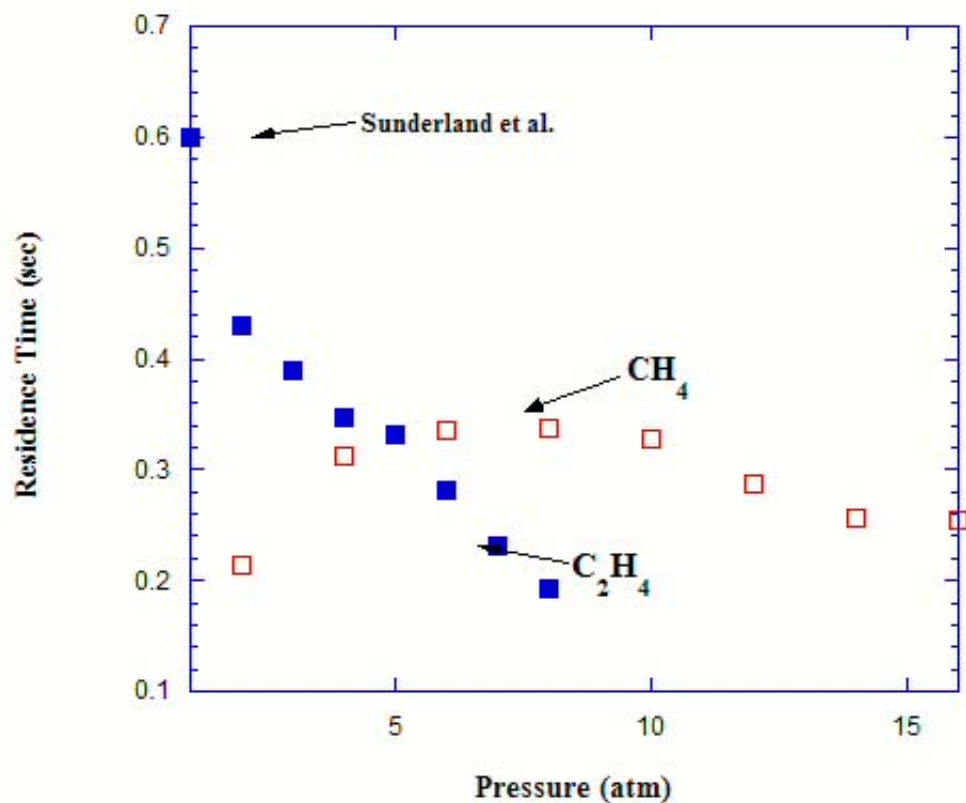
### 3.5 Residence Time Calculations

Residence time was calculated for each fuel, ethylene and methane, for varying pressures of two to eight and two to sixteen atmospheres, respectively. The residence time in a diffusion flame is the time from when the fuel leaves the fuel tube to the time it reaches the tip of the flame, in this case, at the flame's smoke point.

In order to calculate this value, a height or length, in this case the smoke point height and a fuel velocity exiting the area of the fuel tube are needed. The following equation was used to calculate the residence time:

$$\frac{(Smoke\_Point\_Height) * (Area)}{(Volumetric\_Fuel\_Flow) * \left(\frac{1}{Pressure}\right)}$$

where smoke point height is measured in centimeters, area is in squared centimeters, volumetric fuel flow is in standard cubic centimeters per minute, and pressure is in atmospheres. Solving for these values and reducing the units gives a time in seconds. The following plot (Figure 28) shows the effect that increasing pressure had on residence time. There was a vast difference between these effects for ethylene and methane flames. Some differences were expected, as the chemical kinetics of the two hydrocarbons are dissimilar.



**Figure 28: Residence time as a function of pressure**

The current residence time data collected for ethylene was consistent with previously published data (Sunderland, 1994) for residence time of ethylene at one atmosphere. However, to the author's knowledge, this was the first measure of residence time for methane, or for a fuel as a function of pressure, thus no other comparisons are available.

## 4 Conclusions

Soot formation has been studied for at least the last fifty years, but is yet still not understood. In order to gain more knowledge of the many, and often competing, chemical kinetic processes and their resulting emissions, it is necessary to gather a full understanding of all of the fundamental processes involved in forming and producing these soot emissions. The current work focused on fundamental combustion knowledge that is very much under researched, smoke point. The research focused on pure fuels, diluted fuels, effects of dilution rates, and how pressure affects each of these elements. Examining soot formation at these elevated pressures is of utmost importance as this is where most practical combustion devices operate. The current work is unique because it not only measured the smoke point of the pure fuels ethylene and methane, but also of fuels diluted with nitrogen, argon, helium, and carbon dioxide, as well as performing all of these experimental measurements at both atmospheric and elevated pressure conditions up to eight atmospheres for ethylene and sixteen atmospheres for methane. The conclusions obtained from the current research are as follows:

- 1.) Increasing the dilution level in a pure ethylene or methane flame increases the height at which the flame reaches its smoke point at both atmospheric and elevated pressures.
- 2.) A flame's smoke point is a distinct function of the air to fuel velocity ratio, at elevated pressures.
- 3.) Increasing pressure in a pure or diluted flame decreases the height at which the flame reaches its smoke point, as the smoke point is strongly dependent of the inverse of pressure as follows:



a.) for ethylene –  $SPH \sim P^{0.69}$  and  $FFV \sim \exp(0.21 \cdot P)$

b.) for methane –  $SPH \sim P^{0.29}$  and  $FFV \sim \exp(0.10 \cdot P)$

4.) Residence time, for ethylene, decreases with respect to increases in pressure, and for methane, follows a parabolic trend, first increasing, then decreasing, and then seemingly reaching an asymptote with increasing pressure.

## 5 Future Work

The high pressure vessel and laminar jet diffusion flame burner allow for a great deal of interesting and informative research in the near future. In addition to the smoke point data collected at atmospheric and elevated pressure conditions, the current research will continue for the next three years investigating the following items listed below.

- 1.) Further investigation into the residence time and into the effects of fuel Lewis numbers, from current data, will begin immediately.
- 2.) Soot surface temperatures measurements will be made using a three color filter set-up and a black body temperature calibrator.
- 3.) The work of McCrain and Roberts (2005), measuring the soot volume fraction, will be repeated but will incorporate dilution into the pure fuels of ethylene and methane.
- 4.) Three classes of PAH will be measured, using PLIF and spectral filtering, as a function of pressure, diluent and dilution, and fuel.
- 5.) Benzene, acetylene and other major species will be measured using the method of Fourier Transform – Infrared (FT-IR).
- 6.) Once the temperature measurements are complete and the H concentrations are known, and the O and OH atom concentrations are estimated, it will be possible to use the Li/LiOH technique to find hydrogen atom concentration.

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## 7 Appendices

### 7.1 Pressure Vessel Window Assembly

The pressure vessel windows were cleaned with ethanol. Once the windows were taken completely apart, it was necessary to place a layer of high vacuum grease around the circumference of the glass and inside the window's Teflon holder. The window was then placed into the Teflon holder. The Teflon holder was then inserted into the middle flange (shown below), and then all three flanges were pressed together and mounted back onto the vessel with eight threaded bolts.

