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Flammability of Hydrogen at Sub-Atmospheric Pressures and Reduced Oxygen Concentrations

October 2014

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FLAMMABILITY OF HYDROGEN AT SUB-ATMOSPHERIC PRESSURES AND
REDUCED OXYGEN CONCENTRATIONS

By

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Concentrations

By STEVEN REHN

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Francisco Javier Diez

Prototypes aircraft are currently being built and tested that rely on hydrogen fuel cells to provide power for their electrical demands, and some even use hydrogen to power the entire aircraft. The problem with hydrogen is that it is extremely flammable and has never been used in this capacity before. Therefore, the flammability of hydrogen was tested from the pressure at sea level up to 40,000 feet in a 20 L vessel. The lower and upper flammability limits were found first and compared with previous data. Then, peak explosion pressure was found across all flammable hydrogen and oxygen concentrations. The oxygen concentration started from the concentration found in air and was reduced by adding nitrogen. These tests were performed up to the point where the limiting oxygen concentration was reached for each altitude. In general, as the altitude increased the limits of flammability for hydrogen and oxygen widened, and the peak explosion pressures decreased.

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Table of Contents

Title.....	1
Abstract.....	ii
Acknowledgements	iii
List of Tables	vi
List of Figures.....	vii
Chapter 1: Introduction	1
<i>1.1. Background</i>	<i>1</i>
<i>1.2. Basic Combustion Theory</i>	<i>2</i>
<i>1.3. Comparing Common Experimental Methods.....</i>	<i>4</i>
<i>1.4. Past Hydrogen Combustion Test Results</i>	<i>6</i>
<i>1.5. Objective.....</i>	<i>9</i>
Chapter 2: Experimental Methods.....	11
<i>2.1. Flammability Criterion</i>	<i>11</i>
<i>2.2. Explosion Chamber</i>	<i>11</i>
<i>2.3. Spark Energy Tests.....</i>	<i>13</i>
<i>2.4. Test Procedure</i>	<i>16</i>
Chapter 3: Results and Discussion	28

<i>3.1. Overview</i>	28
<i>3.2. Lower Flammability Limit</i>	29
<i>3.3. Upper Flammability Limit</i>	30
<i>3.4. Flammability in Oxygen-Depleted Air</i>	32
<i>3.5. Limiting Oxygen Concentration</i>	33
Chapter 4: Conclusion	51
References	52

List of Tables

Table 1.1: Summary of the altitudes at which the experiment will replicate.	10
Table 2.1: Summary of the pressure rise caused by the spark alone.	27
Table 3.1: Hydrogen lower flammability limit at each altitude.....	39
Table 3.2: Hydrogen upper flammability limit at each altitude.....	41
Table 3.3: Hydrogen limiting oxygen concentration at each altitude.....	49

List of Figures

Figure 2.1: Combustion chamber diagram.....	20
Figure 2.2: Picture of the outside of combustion chamber	21
Figure 2.3: Picture of inside of combustion chamber	22
Figure 2.4: Voltage and Current data recorded for a single spark test	23
Figure 2.5: Power output comparison between four different spark electrode gaps	24
Figure 2.6: Comparison of the total energy output from the spark as the electrode gap changes.....	25
Figure 2.7: Power output comparison between ignitors with differently shaped tips	26
Figure 3.1: Pressure data from a test near the lower flammability limit.....	36
Figure 3.2: Pressure data from a stoichiometric test.....	37
Figure 3.3: Explosion pressure data near the lower flammability limit at each altitude ..	38
Figure 3.4: Hydrogen lower flammability limit in air as a function of altitude.....	39
Figure 3.5: Explosion pressure data near the upper flammability limit at each altitude ..	40
Figure 3.6: Hydrogen upper flammability limit in air as a function of altitude.....	41
Figure 3.7: Upper flammability limit at 40,000 feet with different spark durations	42
Figure 3.8: Atmospheric pressure hydrogen flammability in air with varying added nitrogen concentrations	43
Figure 3.9: 15,000 feet hydrogen flammability in air with varying added nitrogen concentrations	44
Figure 3.10: 30,000 feet hydrogen flammability in air with varying added nitrogen concentrations	45

Figure 3.11: 40,000 feet hydrogen flammability in air with varying added nitrogen concentrations	46
Figure 3.12: Peak explosion over-pressure at varying altitudes and nitrogen concentrations	47
Figure 3.13: Explosion pressure data near the limiting oxygen concentration.....	48
Figure 3.14: Limiting oxygen concentration of hydrogen as a function of altitude	49
Figure 3.15: Flammability limits of all possible combinations of hydrogen, oxygen, and nitrogen at each altitude	50

Chapter 1: Introduction

1.1. Background

There is a push in the transportation industry towards increased fuel efficiency and lower emissions. Aviation accounts for 13% of all transportation carbon emissions and is the fastest growing source, a trend which is expected to continue into the future [1]. New commercial airliners are already becoming “more electric” by replacing some of the older hydraulic flight systems with electro-mechanical systems. Newer airplanes such as the Boeing 777 or Airbus A330/340 already reach electrical demands of over 500kW [2]. Currently this power is produced from the auxiliary power unit (APU) and generators that run off the main engines. The APU is mostly responsible for providing power to the aircraft on the ground and starting the main engines, while the generators provide most of the power while cruising. However, current gas turbine APUs are only about 20% efficient and the engine generators are only about 30-40% efficient [3].

One of the primary methods being considered to improve this efficiency and reduce emissions is to replace the APU with hydrogen fuel cells, which directly convert hydrogen and oxygen to electricity. The only emission produced from these fuel cells is water vapor. Other advantages to a hydrogen fuel cell system are noise reduction, water production that can be used onboard the aircraft, and nitrogen production which can be used to inert the empty space in the jet fuel tanks [4]. Viable concepts are also being produced to power the entire aircraft on hydrogen by using it as a fuel in the main jet engines [5]. In a comparison between kerosene and hydrogen powered jet engines, their thermodynamic efficiency was about equal, but the hydrogen engine used 2.8 times less

fuel by mass [6], meaning hydrogen powered airplanes could be made lighter, thus more efficient. This is more of a long-term solution than the APU replacement, but hydrogen is looking like a very promising fuel for the future of aviation.

From a safety standpoint, hydrogen poses some issues for the aviation industry. It is a highly combustible fuel that is very easily ignited in a wide range of concentrations. It is also a gas in regular atmospheric conditions, unlike the current liquid jet fuel. This means it is able to displace air in confined areas, and if ignited, causes a very powerful explosion, as opposed to the slower moving fires of conventional liquid fuels. Hydrogen flame speeds easily reach above the sonic velocity in air, over 350 m/s [7]. Therefore, hydrogen combustion properties and inerting strategies need to be investigated in a wide range of conditions that can be applied to the aviation industry.

1.2. Basic Combustion Theory

There are a variety of different properties that can be determined when investigating the flammability of a substance. The flammability limits are an important one, as they state the range of fuel concentrations that can be ignited. The lower flammability limit (LFL) is the minimum concentration of fuel in an environment containing an oxidizer that can still be considered flammable. The upper flammability limit (UFL) is the maximum concentration of fuel in the same oxidizer that can still be considered flammable. The majority of experiments and real-world situations use regular air as the oxidizer.

Other important properties that occur at the limits of combustion are the limiting oxidant (or oxygen) concentration (LOC) and minimum ignition energy (MIE). The

LOC is very similar to the lower flammability limit, but instead of it being the minimum amount of fuel needed for combustion, it is the minimum amount of oxidizer needed for combustion to occur. Each kind of fuel in all different types of conditions, such as varied concentrations, temperatures, and pressures requires a different amount of energy input to ignite as well. The minimum amount of this energy needed to ignite a certain mixture is referred to as the minimum ignition energy. It is usually measured using a spark igniter.

In between all of these limits, the combustion reactions produced become a lot more powerful. As the initial conditions change, the flame speed is an important parameter to measure. Most combustion that occurs is considered a deflagration, which is when the flame front produced from the ignition propagates at a speed below the sonic velocity, or the speed of sound. This speed is 343 m/s or 767 mph in room temperature air at sea level [8]. If the flame speed is above the sonic velocity, it is considered a detonation. This induces a shock wave to form outside of the explosion, which causes a much more abrupt rise in pressure and temperature when compared to a deflagration.

Measuring the pressure and temperature rise caused by a combustion event is very important in the study of combustion. Not only can these properties tell how powerful and dangerous the explosion will be to surrounding people and equipment, but they can also be used to determine whether or not combustion has occurred when testing near the different flammability limits. In this experiment, these combustion properties will apply to hydrogen combustion in air at varying initial pressures and nitrogen concentrations.

1.3. Comparing Common Experimental Methods

Fortunately, hydrogen combustion has been well studied in the last century and most of its properties are well known. However, using different methods to measure flame propagation can yield different results. Two tests being conducted with the same hydrogen concentration in air, at the same temperature and pressure can still produce different flammability limits or minimum ignition energies if the size of the vessel is different, the ignition method is different, or the criteria for determining flammability is different. Therefore, it is a good idea to test using different methods and compare the results to get a comprehensive understanding of hydrogen combustion.

One of the reasons for the differing results of the different experiments at the same conditions is the ignition method and energy. Ono, et al. (2007) showed that spark energy has a large effect on flammability limits [9]. Using a capacitor discharge spark, thousands of times more energy was required to ignite hydrogen at the flammability limits than at stoichiometric mixtures. They also showed that a larger gap between the electrodes at the same energy level widened the flammability limits. The spark duration and the amount of humidity in the air were found to have little effect. Kuznetsov, et al. (2012) performed a direct comparison between the flammability limits using a spark igniter and a glow plug which heated a wire [10]. The exact details of the spark igniter were not given, but the glow plug output a maximum of 12.5J, while the spark was less. The glow plug gave higher upper flammability limits and was able to ignite hydrogen down to a lower initial pressure. Other methods were used such as a pyrotechnic igniter [11], but no other direct comparisons were made.

The actual criterion used for determining flammability has an effect as well. The two most commonly used methods are either to measure flame propagation or the pressure increase. The flame propagation method is typically used when the pressure vessel is a long tube. The gas mixture is ignited at the bottom and if the flame propagates to the top, then it is considered flammable. It is either determined visually if a glass tube is being used, or it is determined thermally by a few evenly spaced thermistors to sense the heat from the flame if a metal tube is used [12]. When using the pressure rise method, the percent pressure rise threshold for flammability must be determined. ASTM has attempted to standardize it by using a 7% pressure rise over the initial pressure in their standard test methods [13] [14]. Other experiments have used 1% [15], 3% [11], 5% [16], or 10 times the pressure increase caused by the igniter itself [10]. Obviously, the lower pressure rise thresholds for flammability will produce wider flammable limits. In a comparison between pressure rise and flame propagation criteria, Van den Schoor, et al. (2008) found that the flame propagation criterion produced a lower LFL, while the pressure rise criterion produced a higher UFL [16].

Another problem that needs to be considered when testing the flammability of a substance is the minimum quenching distance. A flame needs to be a certain size in order to propagate or else it will lose too much heat to the surroundings, so if certain dimensions of the vessel being used to test are too small, it will not allow the flame to propagate. This distance increases as the mixture composition becomes leaner or richer than stoichiometric, or as the initial pressure decreases [17]. Tests that are attempted near the upper flammability limit at the lowest initial pressure will have the largest quenching distance for hydrogen.

1.4. Past Hydrogen Combustion Test Results

The majority of the early hydrogen work was summarized in reports by the U.S. Bureau of Mines [18] and the National Advisory Committee for Aeronautics [17] in 1952 and 1958, respectively. They collected lots of useful data about flame temperature, burning velocity, quenching distance, flammability limits, and minimum ignition energy, all at different concentrations, pressures, and temperatures of hydrogen. They also experimented with hydrogen flammability in other atmospheres other than air, including pure oxygen.

Depending on whether the criterion for flammability was upward or downward propagation of the flame, the lower flammability limit (LFL) changed from 4.0% to 9.0%, respectively. The upper flammability limit remained constant at 74% for either criterion however. With increasing initial temperature, the flammability limits widened. With increasing initial pressure, the flammability limits shrunk up to about 20 atm, then widened above that. Decreasing the pressure below atmospheric causes the quenching distance to increase and more spark energy is needed for ignition. Tests were also run to inert hydrogen at atmospheric pressure. With nitrogen or carbon dioxide as a diluent, the oxygen concentration needed to be brought down to 4.9% or 7.5%, respectively, to stop flame propagation.

Since then, further testing was done with hydrogen in pure oxygen environments. Moyle, et al. (1960) [19] and Gealer, et al. (1960) [20] measured the detonation velocities from atmospheric pressure and temperature up to 2000 atm and 5000 K. The velocities increased as the pressure and temperature increased, and as hydrogen concentration increased up to 80%.

Hydrogen's combustion properties at elevated pressures and temperatures have been very well studied. Shebeko, et al. (1995) [21] measured burning velocities and flammability limits of hydrogen and how they changed when diluents were added. The burning velocities increased with increasing pressures and temperatures. The flammability limits changed depending on which diluent was used. The lower flammability limit was found to increase when the diluent has a higher heat capacity than air, and it would decrease when the diluent has a lower heat capacity than air. Kamenskihs, et al. (2010) [22] measured the amount of energy needed to directly initiate a detonation of hydrogen at elevated pressures. It took less energy to produce a detonation event as the pressure increased.

Various experiments with different diluents at atmospheric pressure and temperature have been completed as well. Robin, et al. (1995) [23] found it took 25% Halon-1301 or 24% DuPont FM-200 by volume to completely inert hydrogen in air. Azatyan, et al. (2006) [24] tested four different types of Halons, 1301, 2402, 2501, and NAFS-III, which had inerting concentrations of 25%, 9%, 13.5%, and 20%, respectively. Hexene and octene were experimented with as inhibitors as well and required less than 10% concentration to inert the mixture, but they are flammable themselves, so they are not practical to use as an inhibitor in an actual fire situation. Azatyan, et al. (2005) [25] also ran similar tests with propylene and isopropanol and came to a similar conclusion. Water droplets were tested as well, but required huge amounts of water in order to inert the hydrogen mixture, especially at lower water temperatures. Medvedev, et al. (2002) [26] came to a similar conclusion. Qiao, et al. (2005) [27] found how the laminar

burning velocities change with inert gases helium, argon, nitrogen, and carbon dioxide, but did not test up to the inerting concentration.

The flammability limits of hydrogen have been extensively tested as well. Pfahl, et al. (2000) [28] found the upward propagation limit to be 4.0%, the downward propagation limit to be 8.0%, and the upper flammability limit to be 75% at atmospheric temperature and pressure. It also took a nitrogen concentration of 70% to inert the mixture. Cashdollar, et al. (2000) [11] tested the flammability limits of hydrogen using two different size vessels and two different ignition sources. Using a 7% pressure rise criterion and an electric spark igniter, the LFL was found to be $6.0\pm 0.5\%$ in the 20 L chamber and $6.5\pm 0.5\%$ in the 120 L chamber. Lowering the criterion to a 3% pressure rise changed the LFL to $5.0\pm 0.5\%$ for both chambers. The LFL was tested in turbulent conditions as well and dropped 4% using a 7% pressure rise criterion. Using 5000J pyrotechnic igniters, the LFL was $5.0\pm 0.5\%$.

It wasn't until very recently that the lower and upper flammability limits have been accurately found at pressures below atmospheric. Le, et al. (2012-2013) found the LFL from 1.0 atm down to 0.1 atm [29] and the UFL from 1.0 atm down to 0.05 atm [30]. At 1.0 atm, the flammability limits were $3.95\pm 0.15\%$ to $75.73\pm 0.15\%$ hydrogen in air, then widened to 3.75% to 77.8% at 0.3 atm, then again narrowed to 4.14% to 76.95% at 0.1 atm. The flammability criterion was upward flame propagation in a cylindrical vessel. Kuznetsov, et al. (2012) [10] found that the LFL remained constant at 4% from 1.0 atm down to 0.05 atm. The UFL increased from 75% at 1.0atm to 78% at 0.2 to 0.5 atm, then decreased to 50% at 0.05 atm. However, that sharp decrease was attributed to not having enough spark energy. These tests were all done with the flammability

criterion being a pressure rise of 10 times more than what was produced by the igniter itself. Jones (2009) [31] ran similar tests, but did not have enough spark energy to produce comparable data. Pareja, et al. (2011) [32] tested the laminar burning velocity at 0.947 atm and 0.767 atm and found there was not a significant change between those pressures.

1.5. Objective

The main objective of this experiment is to find the properties of hydrogen flammability in conditions that could possibly be experienced by the commercial aircraft that use it as a fuel. Therefore, testing will be done at air pressures simulating altitudes from 0 to 40,000 feet. Table 1.1 summarizes the altitudes and the corresponding air pressures that will be tested. First, the flammability limits of hydrogen at these air pressures will be found because that can be verified with previous experimental data. Then, further testing will include finding the peak explosion pressure across the full range of hydrogen concentrations.

Nitrogen will be tested as an inerting agent because nitrogen systems are already required to be on most commercial aircraft in order to reduce the oxygen concentration in the jet fuel tanks to a level that will no longer be flammable [33]. Nitrogen will be added to the hydrogen-air mixtures at intervals of 20% to find how the flammability changes as the oxygen concentration decreases. Finally, the limiting oxygen concentration (LOC), which is the minimum amount of oxygen needed for combustion to take place, will be found.

Altitude (ft.)	Pressure (atm)	Pressure (psia)	Pressure (kPa)
0	1.000	14.7	101.3
15,000	0.564	8.29	57.2
30,000	0.297	4.36	30.1
40,000	0.184	2.71	18.7

Table 1.1: Summary of the altitudes at which the experiment will replicate and the corresponding pressures [34].

Chapter 2: Experimental Methods

2.1. Flammability Criterion

This experiment was set up to have the ability to measure the flammability limits of hydrogen along with the higher pressure combustion in the middle of its flammable range. It must be able to do this accurately and consistently, as well as being safe to the user to operate. Since a semi-spherical test chamber was to be used, the pressure rise method is the most practical way to determine flammability. The test method for this experiment was mostly based on the ASTM standards for determining flammability limits [13] and limiting oxygen concentrations [14], except a 3% pressure rise above the initial pressure, instead of 7%, was used as the flammability criterion. This criterion was chosen because hydrogen flames have shown the ability to propagate while still remaining below the 7% pressure rise threshold. A 3% pressure rise would be a much safer option since this experiment is based on aircraft fire safety.

2.2. Explosion Chamber

All of the explosion tests were done in a 20 liter stainless steel chamber based on the Bureau of Mines design for a 20 liter explosibility test chamber for dusts and gases [34]. It is a nearly spherical vessel made of Type 304 stainless steel with a wall thickness of 0.5 inches and maximum pressure rating of 300 psi. It is constructed from a 6" long, 12.75" OD pipe welded to a 6.875" outside radius hemisphere for the bottom. The head is a hinged ellipsoidal piece with an outside height of 2.9375" and a diameter matching the rest of the apparatus. There is a 2.675" diameter sapphire window at the top of the ellipsoidal head to see through. The head is held closed with 6 $\frac{3}{4}$ -10, 4.25" long hex

head bolts and sealed with a rubber o-ring. There are multiple ports around the outside of the chamber to connect sensors, gases, igniters, etc.

There are three types of gases connected to this chamber. They are hydrogen, nitrogen and air, all at about 5 psig. The hydrogen and nitrogen are both 99.999% pure, and the air was fed from a compressor and dried to under 20% humidity. These are controlled by solenoids and the flow rate is regulated by needle valves. Also connected to the chamber is a vent tube leading outside and a vacuum pump (JB Platinum DV-85N) with the ability to reduce the pressure down to 15 microns of mercury, or 2.895×10^{-4} psia. All plumbing to the chamber is done with 1/4" vacuum-rated nylon tubing. Mounted in the center of the chamber are two 1/8" diameter Type 316L stainless steel electrodes used as spark igniters for the gas mixture. One of the electrodes is adjustable so the spark gap can be changed. Connected to the electrodes is a 15,000 V, 30 mA luminous tube transformer (Allanson 1530BPX120) to produce the spark to ignite the gas mixture. A diagram of the explosion chamber showing all of its connections is shown in figure 2.1. Figure 2.2 is an actual picture of the outside of the chamber, showing the high pressure transducer on the left and the low pressure transducer on the right. Figure 2.3 is a picture of the inside of the vessel. The two electrodes, thermocouple, and port for the high pressure transducer can be seen.

For data collection, there is a thermocouple and two pressure transducers also connected to the chamber. The thermocouple (Omega KQXL 1/16") is used to measure the initial temperature. The low-range (0-15 PSIA) pressure transducer (Omega PX409-015A5V-EH) is used measure the partial pressures of the input gases and the explosion pressures at the flammability limits. There is a valve to close off this pressure transducer

before the mixture is ignited because the maximum overpressure it can sustain is 60 psia. The high range (0-150 psia) pressure transducer (Omega PX409-150A5V) is used to measure explosion pressures that exceed the range of the low pressure transducer. These three sensors are all connected to a computer using an IoTech Personal DAQ/3000 Series collecting data at 1kHz per channel.

To ensure the safety and reliability of the hydrogen combustion testing, the combustion chamber was hydrostatically leak tested in accordance with the ASTM E1003-13 Standard [35] by Laboratory Testing Inc. in Hatfield, PA. For this test, the chamber was filled with water and held at 300 psi for 15 minutes. It passed the test by holding the pressure and having a leakage rate of less than 4.5×10^{-7} mol/s.

2.3. Spark Energy Tests

In order to ensure that the energy produced by the spark will not be the limiting factor in determining the flammability limits, the spark energy must be at a maximum. The 15,000 V, 30 mA luminous tube transformer was chosen to power the spark based on two ASTM standard test methods, E681-09 [13] for testing flammability limits of vapors and gases, and E2079-07 [14] for testing the limiting oxidant concentration of vapors and gases. They both state that a 1/4" spark gap should be used, but E681 says to use a continuous spark duration of 0.2-0.4 s while E2079 states a duration of less than 1 s. However, Ono, *et al.* (2007) showed that increasing the spark electrode gap will produce wider flammability limits at the same spark energy level, especially at the upper flammability limit [9]. This means that a larger spark gap should be more adequate than a smaller one at providing enough energy to ignite a combustible mixture.

Therefore, testing was done to measure the actual energy produced by these continuous sparks at different electrode gaps. The instantaneous voltage and current were measured at the high voltage igniter wires using a passive high voltage probe (Tektronix P6015A) and current probe (Tektronix A621). These were connected to a digital oscilloscope (Tektronix THS 730A) and computer to record the data at a speed of 12.5kHz per channel. From the voltage and current data, the instantaneous power could be calculated as well as the total energy produced by the spark.

The gap between the electrodes was varied between 0.186" and 0.75" and the spark timer set to run for 0.05 seconds. For each gap, five tests were run and the energy output was averaged to compare to the other gap sizes. Figure 2.4 shows a graph of the data recorded from a single test at a gap of 0.75". There is a high voltage spike that initiates the spark and then settles down with each subsequent peak in the AC waveform. All the other tests had a similar profile to this one.

After the five tests were run at each of the four electrode gaps, the power output was calculated and averaged over the five runs. Figure 2.5 shows a graph of the average power output over the time of 0.05 s. The average initial power spike at the start of the spark had a very large increase as the electrode gap is increased. The average maximum power for a 0.186" electrode gap is 158 W and this increased to 441 W for the 0.75" gap. The reason the numbers aren't this large on the graph is because the peak doesn't appear at the exact same time for each individual test, so the peaks don't line up perfectly when averaged. The power output after the initial spike also increased as the gap widened, so a larger spark gap produced a much more powerful spark when everything else remained the same. Figure 2.6 shows the total energy output over the 0.05 s for each gap. By

increasing the gap from 0.186" to 0.75" the energy output increased from 1.09 J to 1.88 J, a 72% increase. The increased energy output of the larger spark electrode gap will help to ensure that there is adequate spark energy to ignite gas mixtures near the limits of combustion.

Two different shapes for the tip of the ignitor electrodes were tested as well, a flat tip and a pointed tip. Two identical igniters to the ones used in the explosion chamber were tested at a ¼" gap, first with a flap tip on the electrodes, then with the tips grinded down to a point. Figure 2.7 compares their power profiles averaged over five tests. The pointed electrodes had a slightly higher average energy output of 1.264 J compared to 1.213 J for the flat tip. The bigger difference, however, was that the ignitor with the pointed tip had a much larger power spike at the start of the spark, an average of 731 W compared to 535 W, an increase of 37%. Again, the average peak is not shown on the graph because the peaks for each individual test do not occur at the exact same time.

The final configuration chosen for the ignitor was a pointed tip with a 0.65" electrode gap and a spark duration of 0.5 s. The 0.65" gap was chosen over the larger 0.75" gap because the larger gap was much less consistent because it would sometimes ground out to the explosion chamber itself. The 0.65" gap did not have this problem and still provided a good average energy output of 1.67 J in a 0.05 s spark. The average amount of energy output over the full 0.5 s spark duration is 12.42 J with a standard deviation of 0.8J.

2.4. Test Procedure

There are several steps involved in performing a single hydrogen combustion test in the explosion chamber. Certain procedures are required when starting up the system at the start of each day of use, then less are required between tests. All are done to ensure accuracy and consistency between tests completed at different times on different days.

Before any tests were completed, the amount that the pressure increased in the chamber because of the heat generated by the spark itself needed be accounted for. Therefore, five tests were run at each initial pressure of only the spark being triggered in pure air. Each of the five pressure increases were averaged and then that number was subtracted from the pressure rise for every test at its respective initial pressure. This was done to ensure that the pressure increase recorded came only from the reaction of the gases and not the spark itself. Table 2.1 shows the average pressure increase caused by the heat generated by the spark itself at each test pressure.

To start up the explosibility chamber system, first the valves for the gas cylinders and pressure regulators must be opened and the power to the 28 V circuit and the 115 V circuit must be turned on. The 28V circuit powers the pressure transducers and the relays for the gas solenoids. The 115 V circuit powers the solenoids themselves, the vacuum pump, and the ignitor. The computer must also be turned on and the data acquisition software started. The valve to vent the contents of the chamber outside should be open at this time. Then the hydrogen and nitrogen gas lines must be flushed with their respective gases to ensure the purity of the gases entering the chamber. After this, the air solenoid is opened for at least 15 minutes to flush the chamber with air to ensure it is filled with 100% dry air.

The following steps need to be completed to perform a single explosion test.

1. The valve must be opened to the low pressure transducer so partial pressure measurements of the input gases can be taken.
2. The explosion chamber needs to be evacuated before each test, so the vacuum valve is to be opened and the pump turned on. The pressure should be brought down to 1 psia or less depending on how much air is required for each test. For example, tests performed at the upper flammability limit at a pressure simulating 40,000 feet of altitude only require about 0.63 psia of air, while the rest will be hydrogen. Record the pressure of the air left in the chamber. This amount of air must be factored into the total gas composition.
3. After evacuation, the chamber needs to be filled with the input gases, using the partial pressures of the gases to measure the composition of the final mixture. Each solenoid controlling each gas must be opened separately of each other. The needle valves controlling the flow rate of the input gases may need to be adjusted depending on how much of each gas will be added. The first gas added should be the one that will have the smallest quantity of the final mixture. Then record the pressure after this gas is added. The second gas added should be of the next highest quantity and the pressure must be recorded again. The final gas added should be the largest component of the mixture and should be added at the highest flow rate possible to ensure proper mixing of the gas composition [14]. The actual initial pressure and temperature is to be taken from the output file after the test is completed.

4. If the test being performed is not a test that is at or near the flammability limits of the input gases, the valve to the low pressure transducer must be closed. The low pressure transducer can handle an overpressure of up to 60 psia, but some explosion tests can go well above that. If there is any concern that the pressure might approach anywhere close to this limit, the valve must be closed.
5. Prior to ignition, it is time to start recording pressure transducer data on the data acquisition software. The data should be recording at a speed of 1000 Hz to ensure an accurate recording of the pressure rise curve and peak pressure.
6. In order to get an accurate reading of the initial pressure, the software needs to record for at least 2 seconds, then the spark is to be triggered to ignite the mixture. The initial 2 seconds is for recording the initial pressure and temperature. Continue to record data for at least 10 seconds after the ignitor is triggered to record the full pressure data of the explosion and aftermath.
7. The peak change in pressure caused by the explosion is to be recorded, after subtracting the pressure rise caused by the heat released from the spark itself at the corresponding pressure. If the pressure rises more than 3% above the initial pressure, the gas mixture is considered flammable.
8. In order to flush the combustion products outside and refill the chamber with pure, dry air again, the vent valve and air solenoid need to be opened for at least 15 minutes. Then the next test may be attempted after this is completed.

Then to shut down the system after a group of tests has been completed for the day, close all solenoids and valves, close all gas cylinders and regulators, turn off the 28 V and 115 V circuits, and turn off the computer.

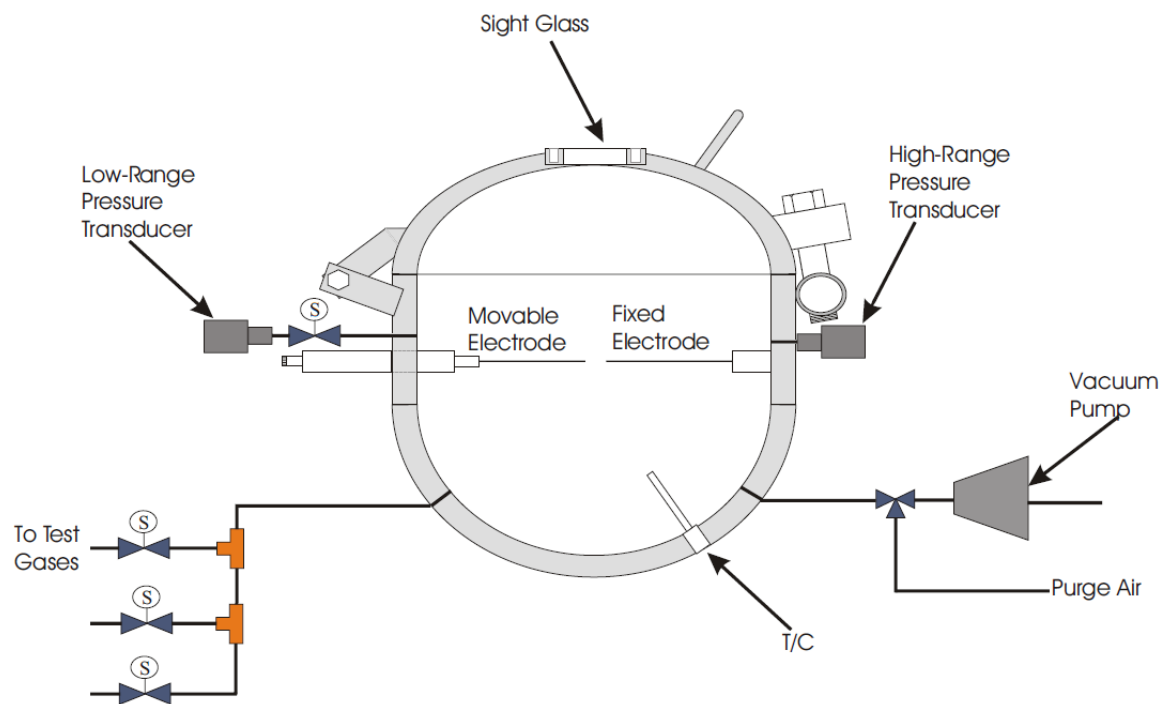


Figure 2.1: Combustion chamber diagram showing all physical connections and sensors for data collection.

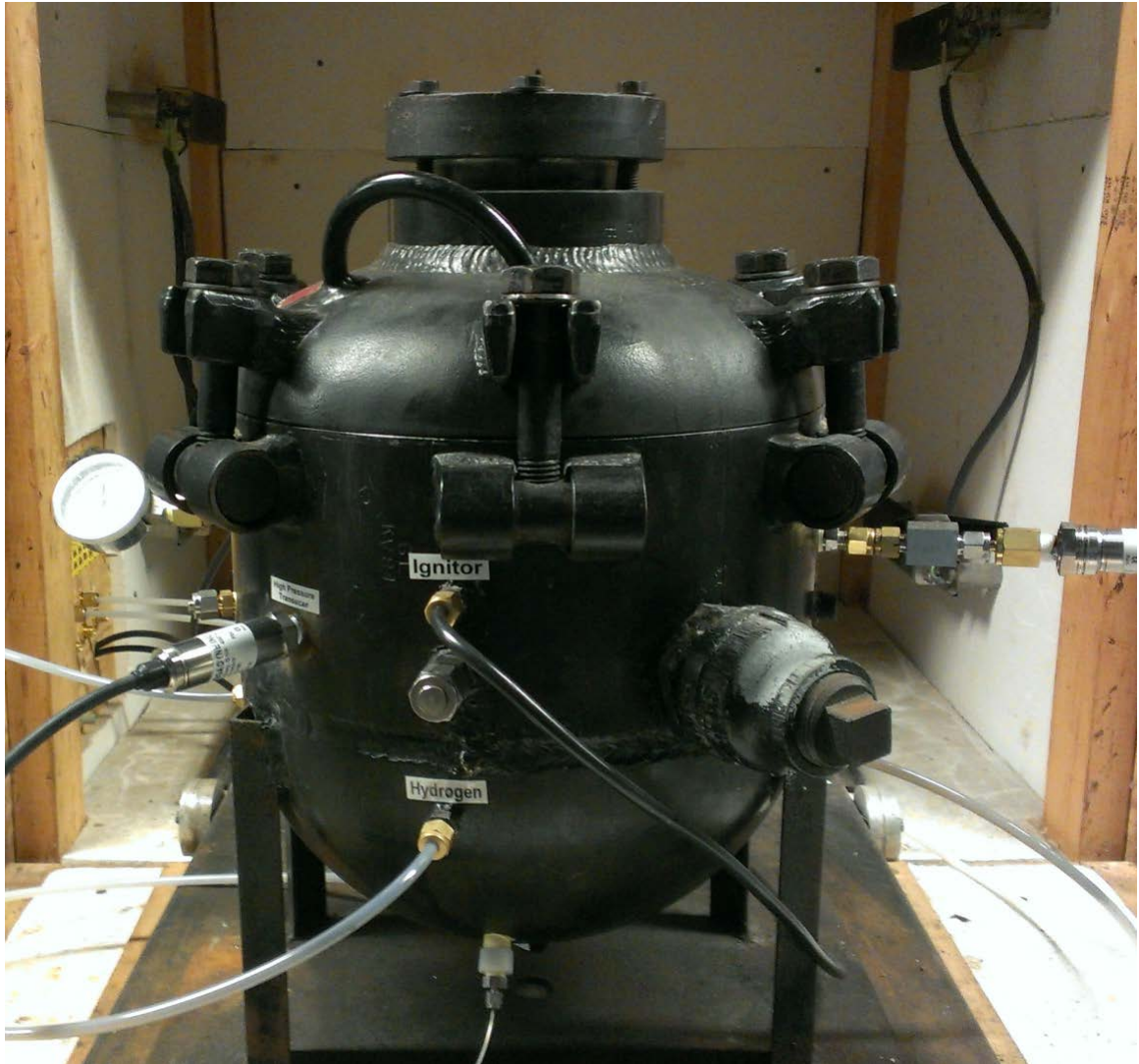


Figure 2.2: Picture of the outside of combustion chamber. Shown in this picture are the high pressure transducer (left), low pressure transducer (right), thermocouple (bottom), hydrogen tube, and one of the two igniter wires (middle).

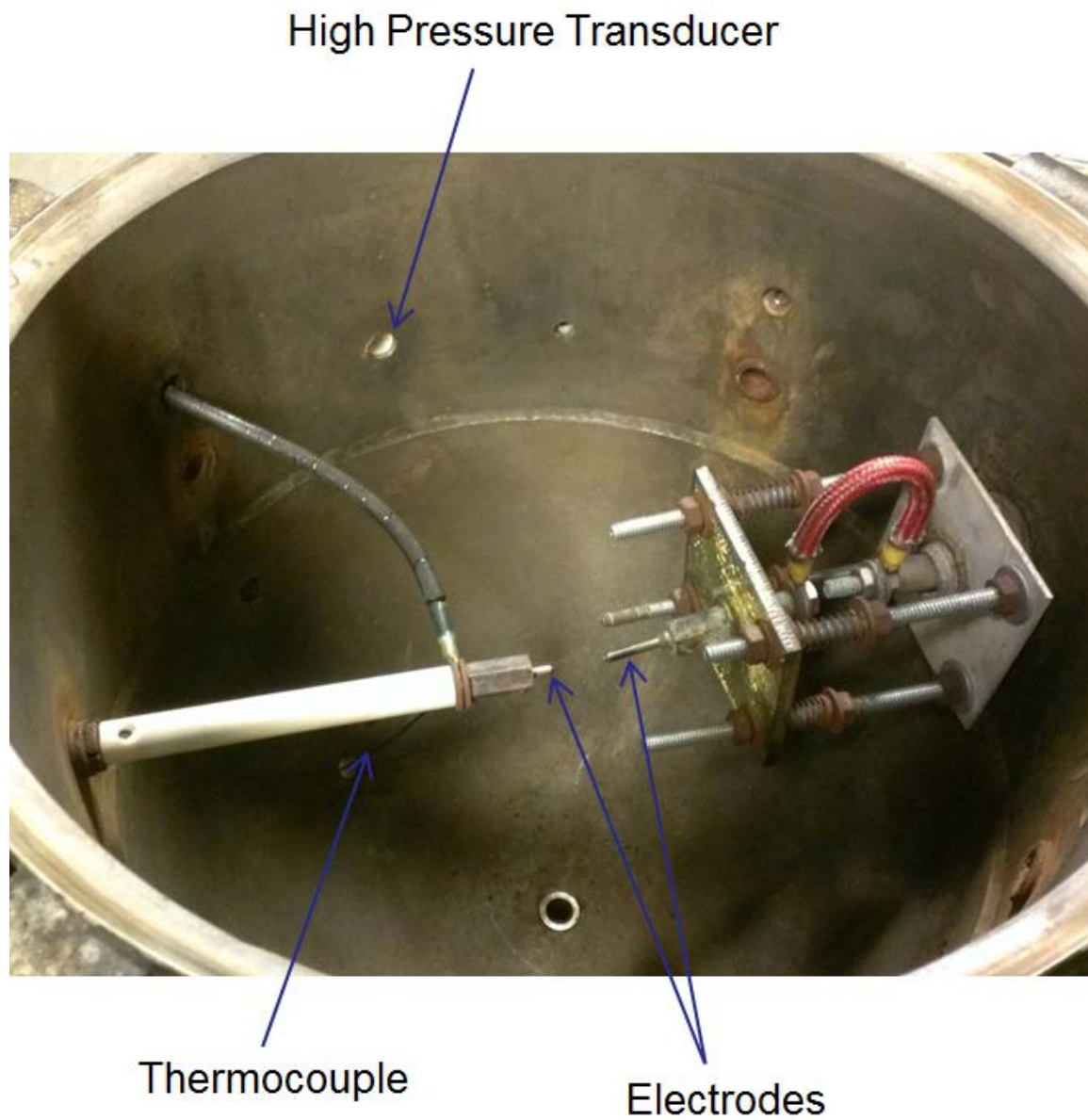


Figure 2.3: Picture of inside of combustion chamber. Shown in this picture are the fixed and movable electrodes, thermocouple, and the position of the high pressure transducer.

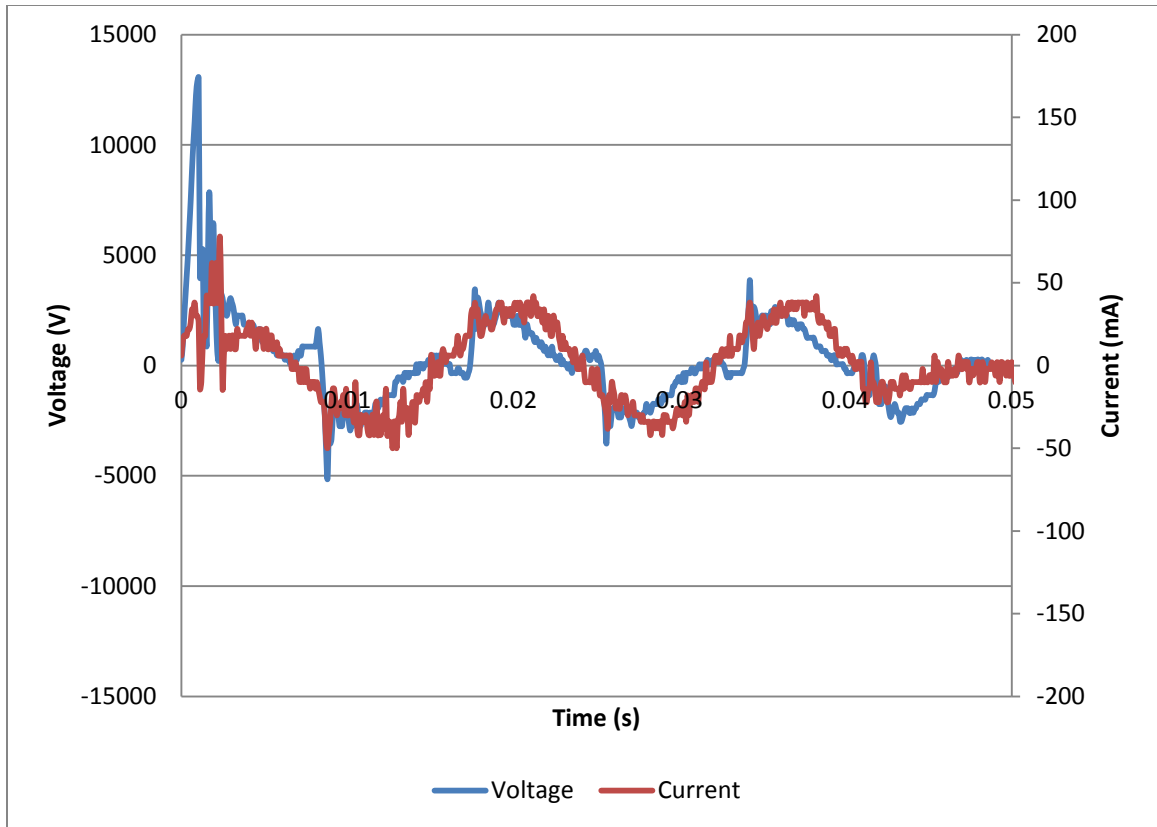


Figure 2.4: Voltage and Current data recorded for a single spark test with a $\frac{3}{4}$ " electrode gap and 0.05 s spark duration.

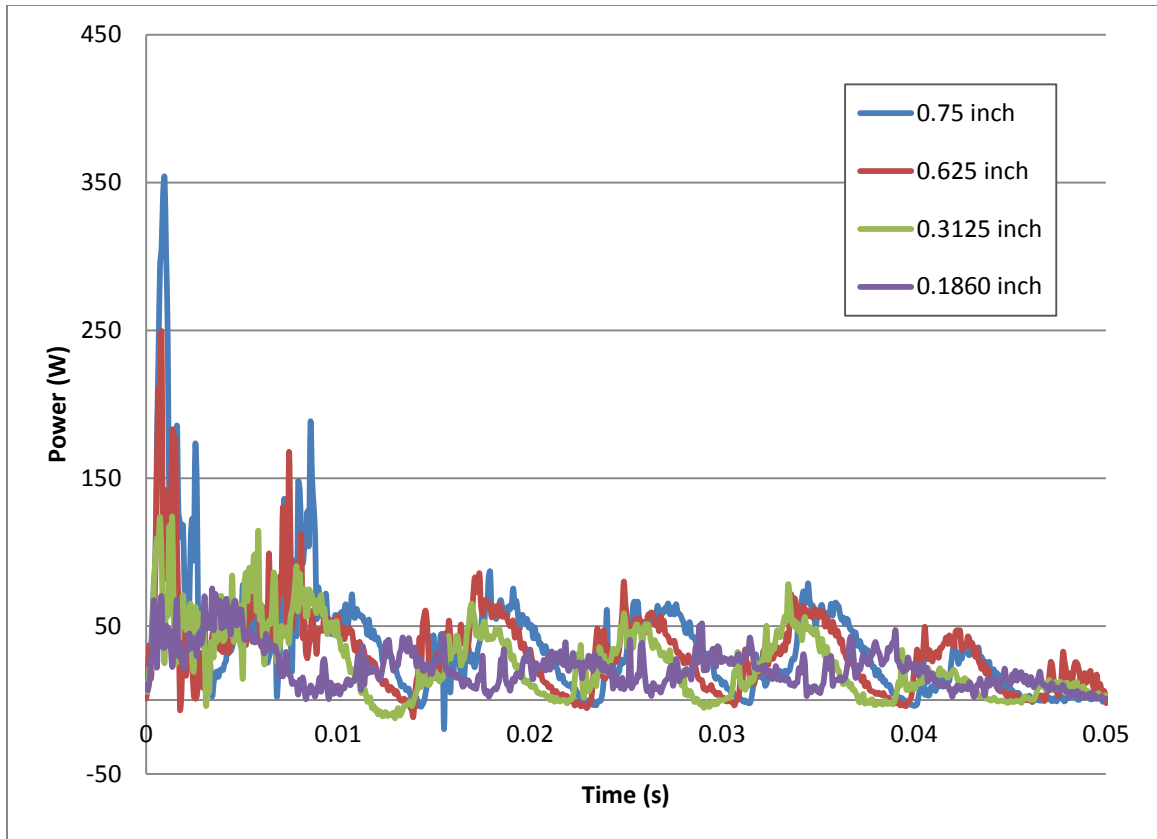


Figure 2.5: Power output comparison over time between four different spark electrode gaps. The data for each gap is from the average of five individual spark events of 0.05 s duration.

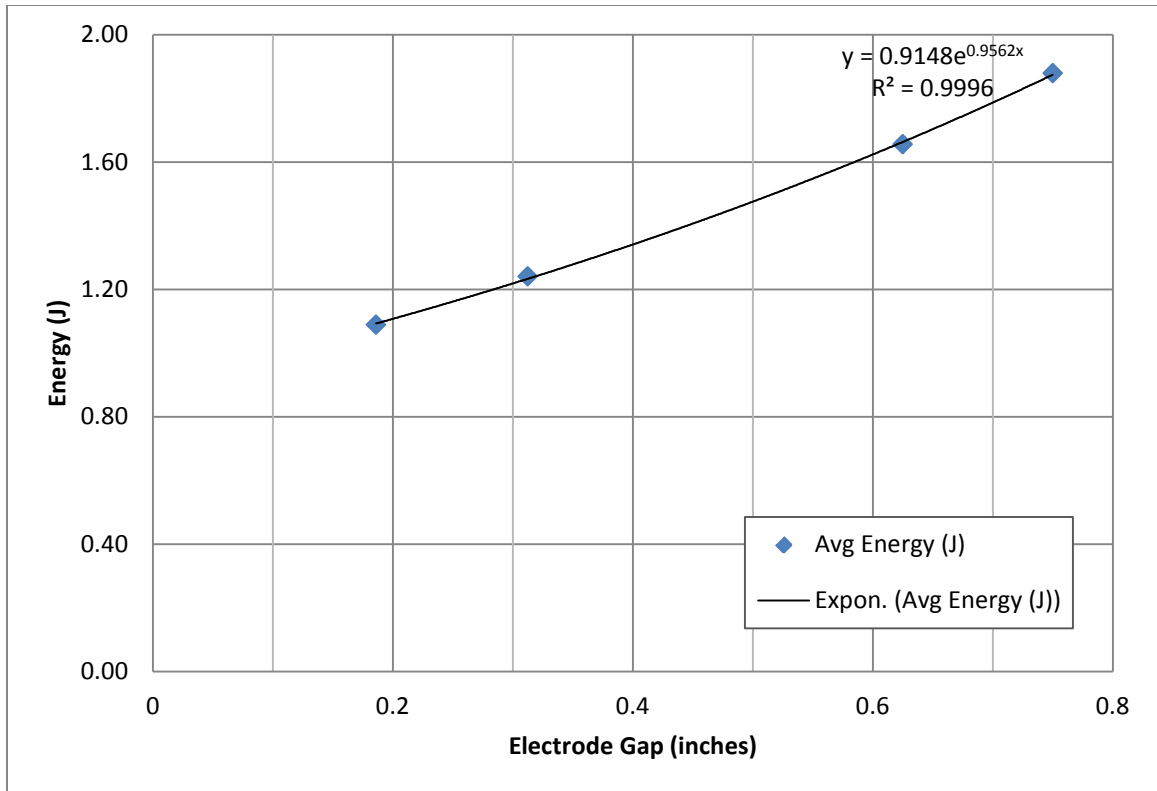


Figure 2.6: Comparison of the total energy output from the spark as the electrode gap changes. The data for each gap is from the average of five individual spark events of 0.05 s duration.

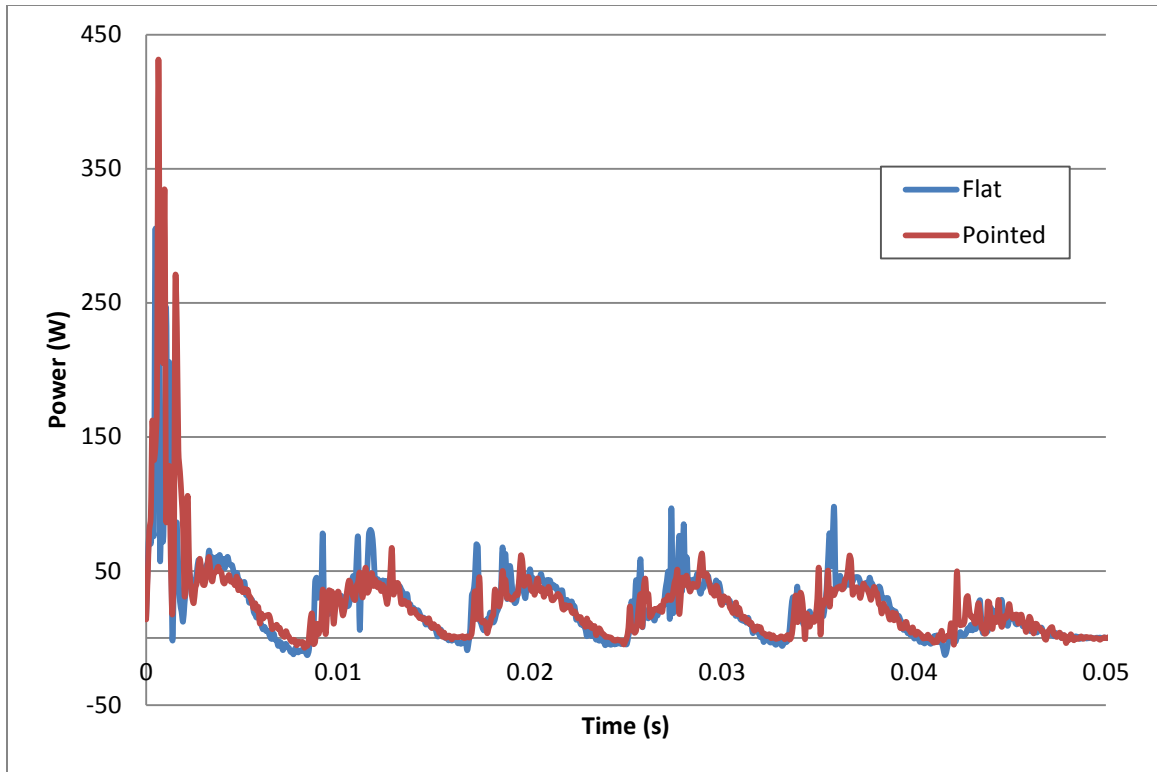


Figure 2.7: Power output comparison over time between two igniters with the shape of the tips of the electrodes either flat or pointed. The data for each shape is from the average of five individual spark events of 0.05 s duration.

Altitude (ft.)	Pressure (atm)	Pressure (psia)	Pressure (kPa)	Pressure Rise from spark only (psi)
0	1.000	14.7	101.3	0.0461
15,000	0.564	8.29	57.2	0.0382
30,000	0.297	4.36	30.1	0.0268
40,000	0.184	2.71	18.7	0.0220

Table 2.1: Summary of the initial pressures to be tested and the pressure increase caused by the spark alone in air at those pressures. The pressure rise caused by the spark alone is from an average of five individual tests each.

Chapter 3: Results and Discussion

3.1. Overview

The flammability of hydrogen was determined for many different conditions that may be experienced when hydrogen systems are used at high altitude. The upper and lower flammability limits, limiting oxygen concentration, and maximum explosion pressures were found across the full range of hydrogen concentrations, oxygen concentrations, and pressures simulating 0 to 40,000 feet in altitude. Testing at the limits of combustion used a 3% pressure rise over the initial pressure as the criterion for determining flammability, while testing between these limits determined how dangerous hydrogen can be at the conditions specified.

Each test produced a pressure curve with 1000 data points per second. Two examples of the pressure data recorded from a single test are shown in figures 3.1 and 3.2. Figure 3.1 is a test near the lower flammability limit at an altitude of 15,000 feet. It shows the pressure of the mixture with respect to time for a period of 3 seconds. The spark is triggered 500 ms into the graph and the pressure increases from there. The spark then turns off another 500 ms later, at which point the pressure continues to rise for another 323 ms, meaning that the explosion still propagates on its own even near the flammability limits. The initial pressure was 8.292 psia and the peak pressure reached was 8.630 psia, giving a pressure rise of 3.62% after subtracting for the pressure rise caused by the spark alone.

The pressure data from a stoichiometric hydrogen-air test looks much different. This condition at atmospheric pressure is shown in figure 3.2. This test produced the highest peak pressure out of the entire experiment. The spark was triggered 25 ms into

this graph and the peak pressure is reached 12 ms later. The pressure raised from the initial pressure of 14.704 psia to a peak of 115.52 psia, giving a pressure rise of 686%, a very dangerous condition.

3.2. Lower Flammability Limit

Several tests were completed near the lower flammability limit (LFL) at each altitude. As the hydrogen concentration increased, the peak explosion pressure increased. This produced the curves shown in figure 3.3. Each point represents the peak pressure increase as a percentage of the initial pressure at different altitudes and hydrogen concentrations. The points where the parabolic best fit lines intersect a 3% pressure rise are the lower flammability limits for each altitude. As shown by the graph, the LFL decreases as the altitude increases, meaning less hydrogen is needed at high altitude to still be flammable.

The graph also shows how hydrogen explosions can become much more powerful even with very small increases in hydrogen concentration. This effect then becomes even more pronounced as the initial pressure drops like it would at high altitude, as shown by the steeper slopes for the curves at 30,000 and 40,000 feet. The same hydrogen concentration at the lower flammability limit at sea level will produce about a 10% pressure rise at 40,000 feet.

The actual lower flammability limit decreased from 4.70% hydrogen at sea level down to 3.89% at 40,000 feet. Figure 3.4 summarizes the lower flammability limit as the altitude changes. The graph also shows how the limit changes if the flammability criterion was changed to a 2% or 4% pressure rise. The difference is more pronounced at

sea level, giving a change of almost $\pm 0.3\%$. The difference at 40,000 feet is much smaller, with a change of only $\pm 0.09\%$.

These numbers compare favorably to previous studies. The majority of them had the LFL at atmospheric pressure from 3.95% to 5.0%, the lower bound of that having upward propagation as the flammability criterion, which typically produces a lower LFL. One experiment that tested at sub-atmospheric pressures had the LFL remaining constant at 4% as the pressure dropped, and the other had it drop from 3.95% to 3.75% at its lowest point at 0.3 atm. This was a very similar trend to the data in this experiment.

3.3. Upper Flammability Limit

Similar tests that were run for the LFL were also completed around the upper flammability limit (UFL) of hydrogen. The peak pressure data for each altitude is shown in figure 3.5. The same 3% pressure rise criterion for flammability is applied to this graph, but the exact point cannot be seen on the graph because of the much larger vertical scale. The actual upper flammability limits as the altitude changes are shown in figure 3.6. Figure 3.5 is the same type of graph with the same span of hydrogen concentrations (4%) as the LFL, but the trends are completely different. Small changes in the hydrogen concentration near the UFL can produce a much greater explosion pressure increase than what was observed near the LFL. For example, dropping the hydrogen concentration just one percentage point from 78.0% to 77.0% at atmospheric pressure, the explosion pressure increased from 3.75% to 167%.

It is also shown in the graph that as the altitude increased, the drop-off in the pressure rise at the UFL becomes much steeper. It is fairly gradual at atmospheric

pressure, becomes a little steeper at 15,000 feet, but then becomes completely vertical at 30,000 and 40,000 feet. This is because at the lower pressures, more spark energy is required near the UFL to ignite the mixtures. There is a sharp drop from 250% pressure rise or more down to just over 2% at the UFL of 30,000 and 40,000 feet. These limits are actually the limits of the spark energy, and not the true upper flammability limits.

Increasing the duration of the spark produced higher UFLs at high altitudes because of the increased spark energy. Figure 3.7 shows the upper flammability limit at 40,000 feet with 0.5s, 1.0s, and 10s spark durations. Leaving the spark on for a longer amount of time puts more energy into igniting the mixture. Increasing the spark duration from 0.5s to 10s raised the UFL to about 78% H₂, up from 76.87%. This still does not appear to be the true UFL either because there is no gradual drop to the non-flammable mixture, like what happens at atmospheric pressure.

The previous experiments that have been done have produced similar upper flammability limits to this test. In the two experiments that tested the UFL at sub-atmospheric pressures, it was found to increase until it got to the pressure at about 40,000 feet. This trend started to show itself as the UFL increased at 15,000 feet, but then reversed at the altitude increased further. This shows that the amount of spark energy specified by the ASTM standards is not adequate for tests at the UFL at low pressure and more spark energy is needed. However, the spark energy was adequate for all other situations, so determining how much more spark energy was needed was beyond the scope of this project.

3.4. Flammability in Oxygen-Depleted Air

The flammability of hydrogen was also tested in air with varying concentrations of nitrogen added. The full range of hydrogen concentrations were tested with pure air, 20%, 40%, and 60% nitrogen added at each altitude. The peak explosion over-pressure was found at all concentrations in order to quantify the danger posed by hydrogen in air and in lower oxygen environments.

The peak percent pressure rise across all hydrogen concentrations at the four different nitrogen concentrations at atmospheric pressure, 15,000 feet, 30,000 feet, and 40,000 feet was found. The graph for atmospheric pressure is shown in figure 3.8. It shows that the highest pressure rise is reached at a stoichiometric mixture of hydrogen and air, which is 29.5% H_2 . The actual pressure rise data vs. time for this test was shown earlier in figure 3.2. Each subsequent nitrogen concentration added shifts the stoichiometric point to a lower hydrogen concentration because there is less oxygen available to react with it. The highest pressure rise for 20%, 40%, and 60% N_2 added occur at 23.6%, 17.7%, and 11.8% H_2 , respectively.

For each altitude, the basic trend in the data is the same. The added nitrogen has no effect on the lower flammability limit at any altitude because the LFL is limited by the amount of hydrogen. As the amount of hydrogen increases, the percent pressure rise increases exponentially until about 10% H_2 . In the lean mixtures, the amount of nitrogen added does not change the peak explosion pressure reached. The differences in the nitrogen concentrations only start to show when each mixture reaches its respective stoichiometric point. Then the peak pressure decreases until the upper flammability limit

is reached. Figure 3.9 shows this data for 15,000 feet, figure 3.10 is 30,000 feet, and figure 3.11 corresponds with 40,000 feet.

The differences between each graph are more subtle. Atmospheric pressure has the highest percent pressure rise for a stoichiometric mixture at 0%, 20%, and 40% N₂, but the lowest for 60%. As the altitude increases, the percent increase of the peak pressure decreases at 0%, 20%, and 40% N₂, but increases at 60%. Also, the flammability limits get wider as the altitude increases for all concentrations of nitrogen, except for the situations that were limited by the spark energy, which were 0% and 20% N₂ at 40,000 feet, and 0% N₂ at 30,000 feet. For example, the flammability limits with 60% N₂ go from 4.70% - 19.76% H₂ at sea level up to 3.89% - 22.84% H₂ at 40,000 feet.

Showing the actual peak explosion pressure reached instead of as a percentage of the initial pressure, a much bigger difference between each altitude can be seen. The data from the four graphs above are combined into one for figure 3.12. It is much easier to compare how the flammability limits change at each altitude and how much more powerful a hydrogen explosion can be at sea level than at altitude. The highest over-pressure reached at sea level was 5.75 times higher than the highest over-pressure at 40,000 feet. The added initial pressure means there is a much higher mass of hydrogen and oxygen occupying the same volume, creating a much more powerful explosion.

3.5. Limiting Oxygen Concentration

The limiting oxygen concentration (LOC) is the minimum concentration of oxygen required for hydrogen to still be flammable. This test was done by reducing the amount of oxygen in the air by adding nitrogen, then adding just enough hydrogen to

make a stoichiometric ratio with that amount of oxygen. For example, a test run with a 4.00% oxygen concentration would have 8.00% hydrogen and the rest (88%) would be nitrogen.

The peak pressure data was recorded for each test run near the limiting oxygen concentration at each altitude. The graph produced is very similar in appearance to the lower flammability limit graph (figure 3.3); however the x-axis compares different oxygen concentrations instead of hydrogen concentrations. It is shown in figure 3.13. A 3% pressure rise is still used as the flammability criterion. As shown, the amount of oxygen required decreases as the altitude increases, following the same trend of wider limits at high altitude.

The LOC at sea level is 4.21% and drops to 3.49% at 40,000 feet. Figure 3.14 shows the limiting oxygen concentrations as a function of altitude. Also shown in the graph is how the limit would change if flammability criterion was adjusted to a 2% or 4% pressure rise. Lowering it to 2% caused a much bigger change in the LOC than raising the criterion to 4%. The fact that the LOC decreases as the elevation increases also gives credence to the idea that the tests at the upper flammability limit were limited by the spark energy at high altitude. The UFL is also a limiting oxygen concentration test; however it is done in an atmosphere of mostly hydrogen instead of nitrogen.

The flammability limits for all possible combinations of hydrogen, oxygen, and nitrogen from sea level to 40,000 feet can be summarized into a single graph. This is shown in figure 3.15. Everything inside the curve is considered to be a flammable mixture. The amount of nitrogen and oxygen shown on the graph would be the amount in the original atmosphere before the hydrogen is added. The nitrogen on the x-axis is

only the amount that needed to be added to air, it does not count the amount of nitrogen already in the air. The amount of hydrogen added would then displace part of the nitrogen-air mixture. This is why the lowest oxygen concentration shown on the graph for 40,000 feet is 3.75%, when the actual limiting oxygen concentration is 3.49%. When the hydrogen is added, it would reduce the oxygen concentration from 3.75% to 3.49%.

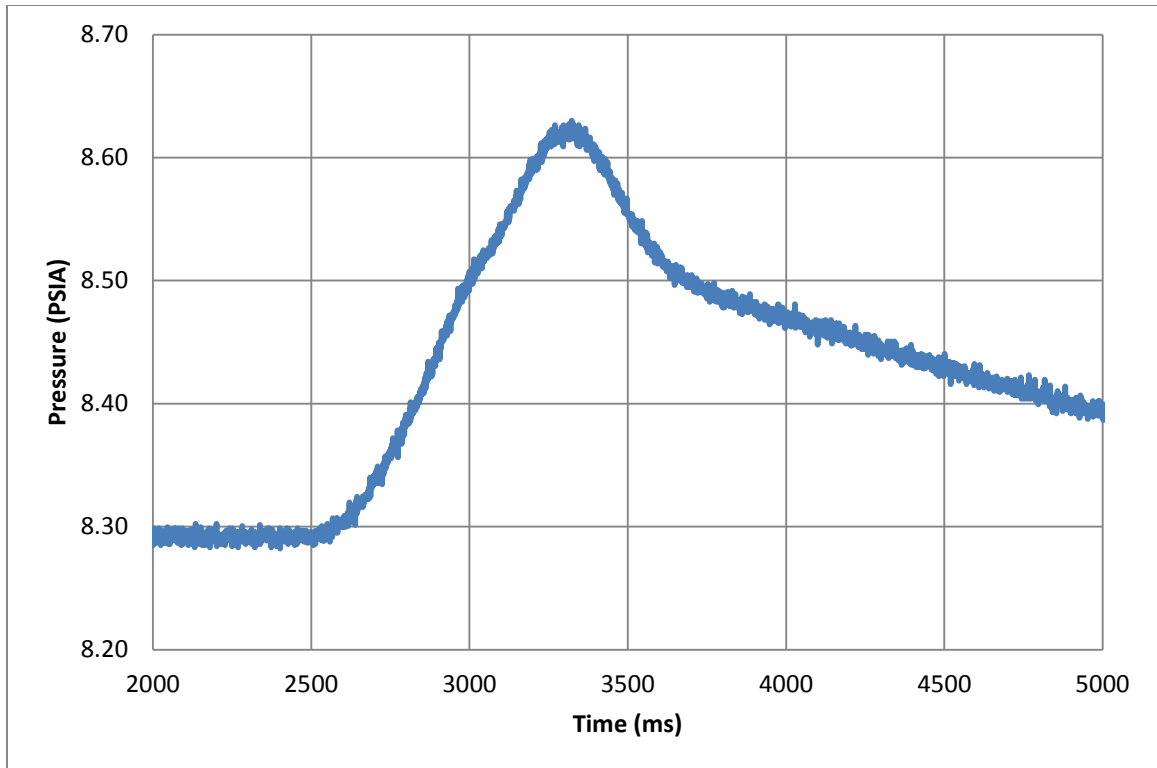


Figure 3.1: Pressure data from a test near the lower flammability limit at 15,000 feet. This specific test had an initial pressure of 8.292 PSIA a gas composition of 4.55% H_2 and 95.45% air. There was a pressure rise of 3.62% over the initial pressure, meaning this mixture is considered flammable.

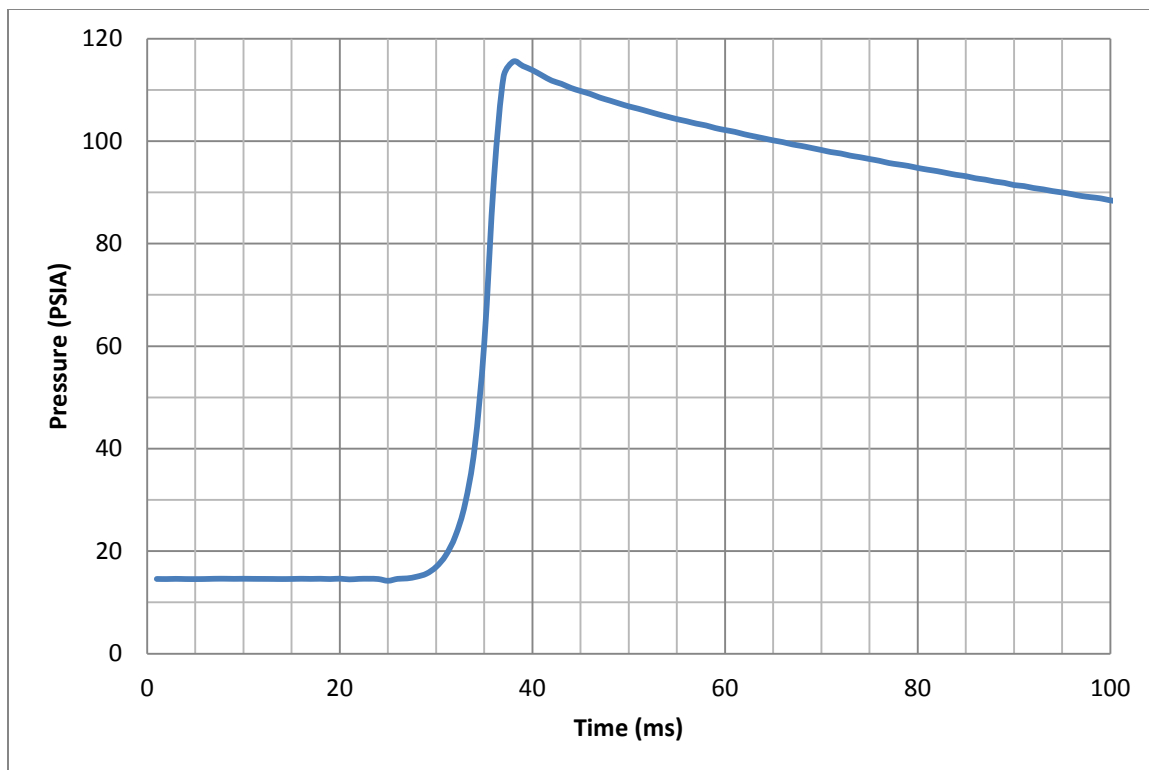


Figure 3.2: Pressure data from a stoichiometric test at atmospheric pressure. This specific test had an initial pressure of 14.704 PSIA, with a gas composition of 29.73% H_2 and 70.27% air. It had a 686% pressure rise and a rise time of 12 ms.

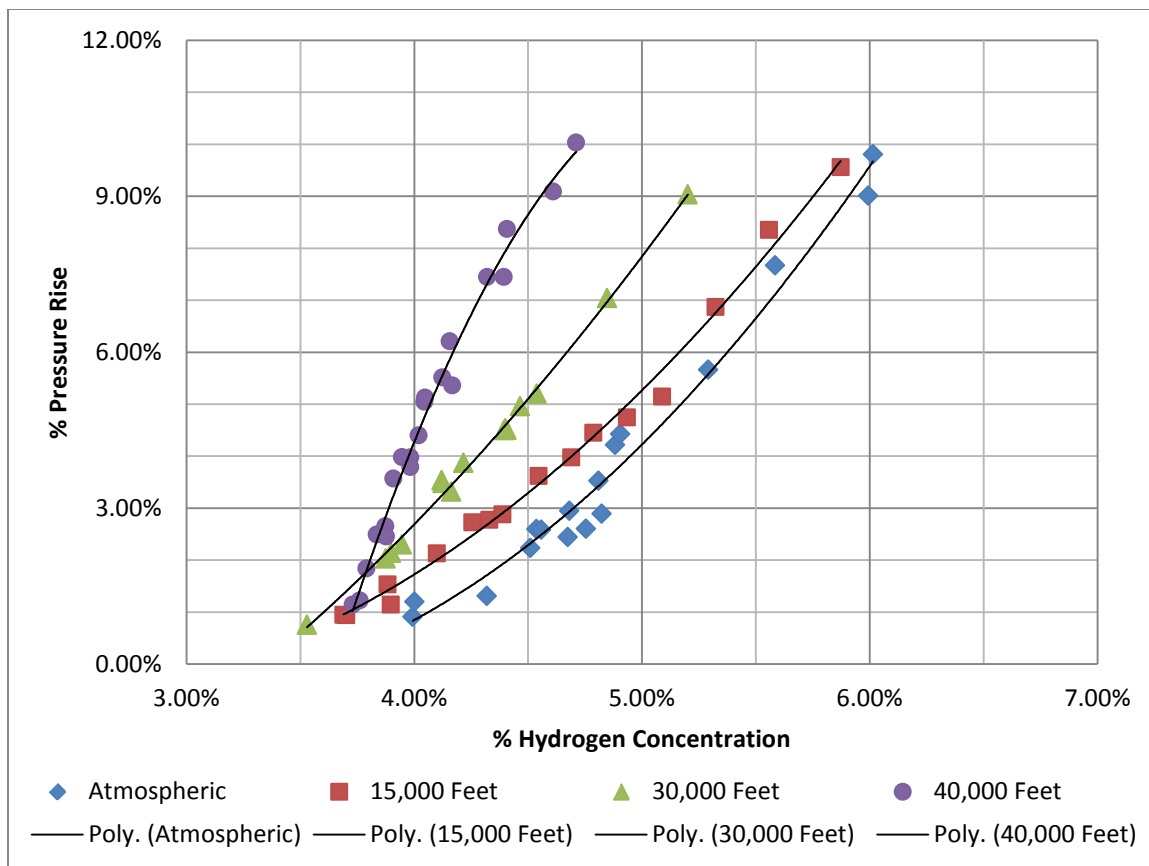


Figure 3.3: Explosion pressure data near the lower flammability limit at each altitude. Each point shows the peak pressure rise of a single test as a percentage of the initial pressure. A 3% pressure rise or more is considered flammable.

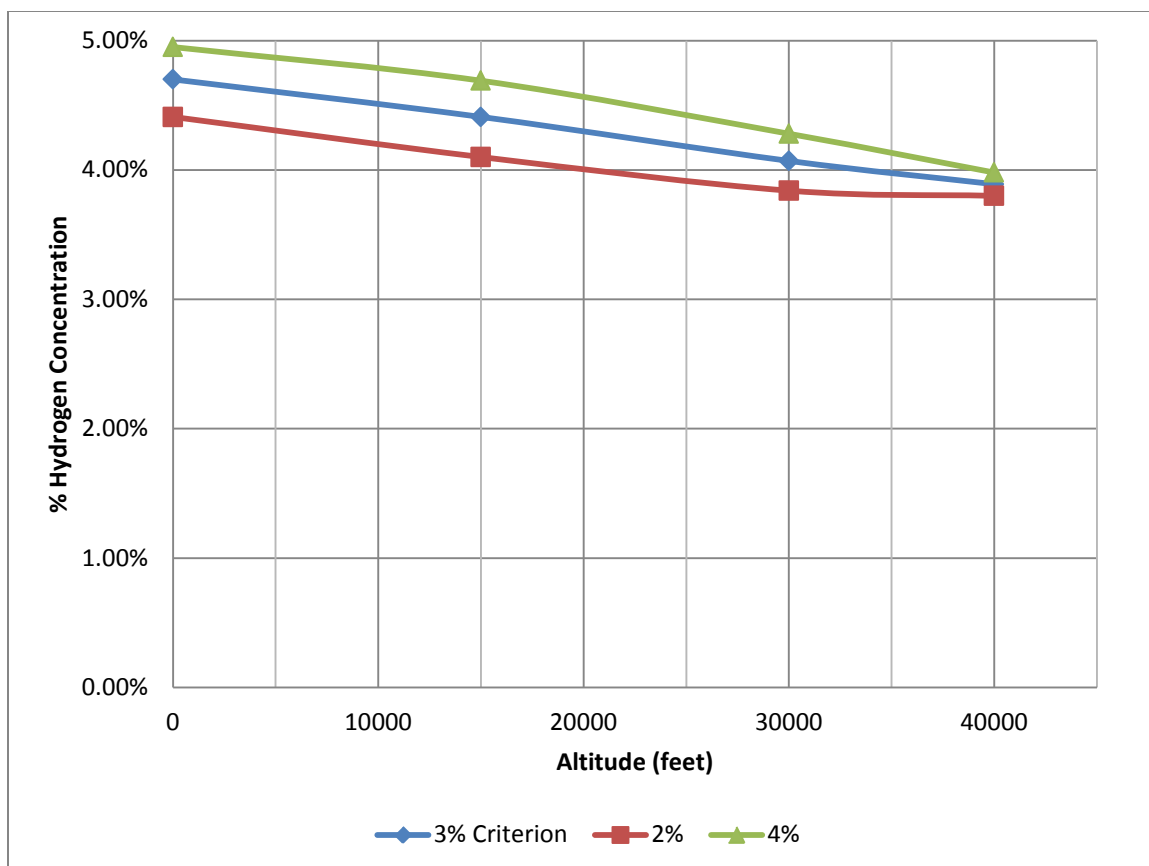


Figure 3.4: Hydrogen lower flammability limit in air as a function of altitude using three different flammability criteria.

3% Criterion	
Elevation (ft.)	Lower Limit
0	4.70%
15,000	4.41%
30,000	4.07%
40,000	3.89%

Table 3.1: Hydrogen lower flammability limit at each altitude using a 3% pressure rise criterion.

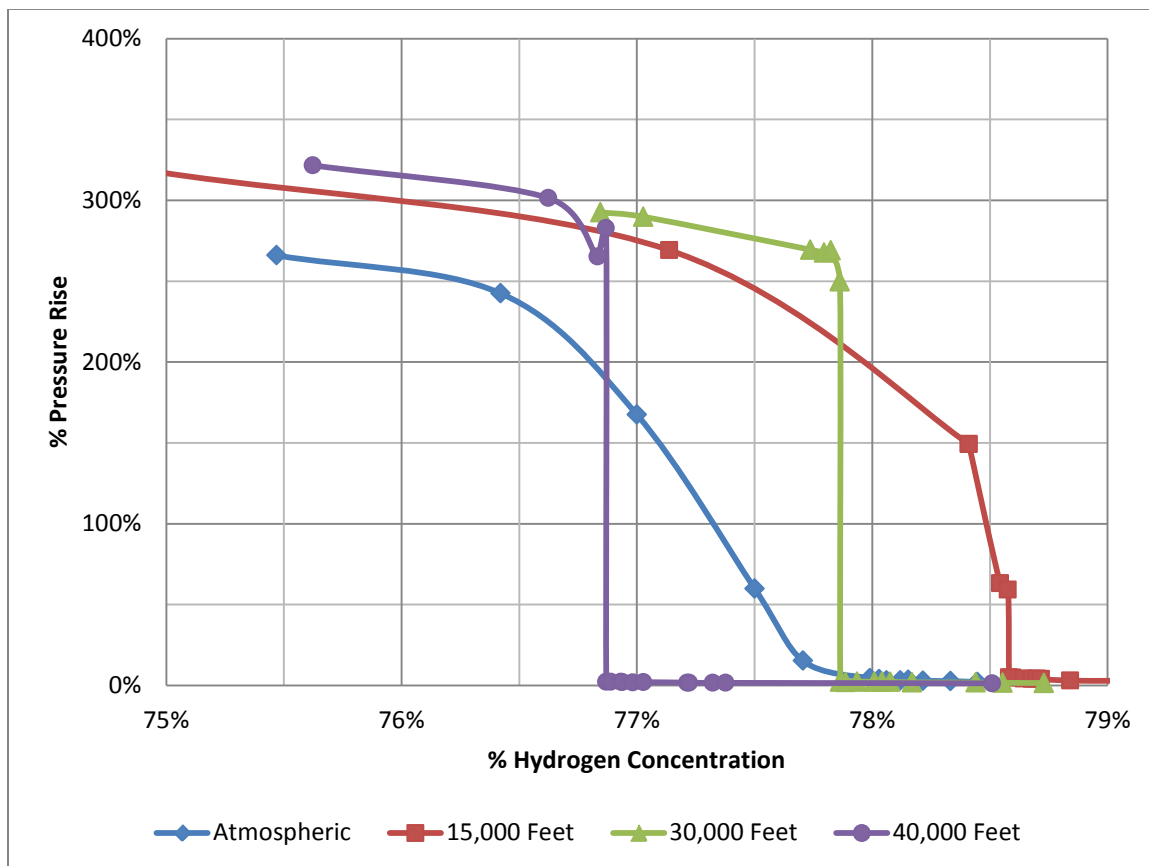


Figure 3.5: Explosion pressure data near the upper flammability limit at each altitude. Each point shows the peak pressure rise of a single test as a percentage of the initial pressure. Although it can't be seen in this graph because of the much greater pressure rise with small changes in H_2 concentration when compared to the LFL, a 3% pressure rise or more is still considered flammable.

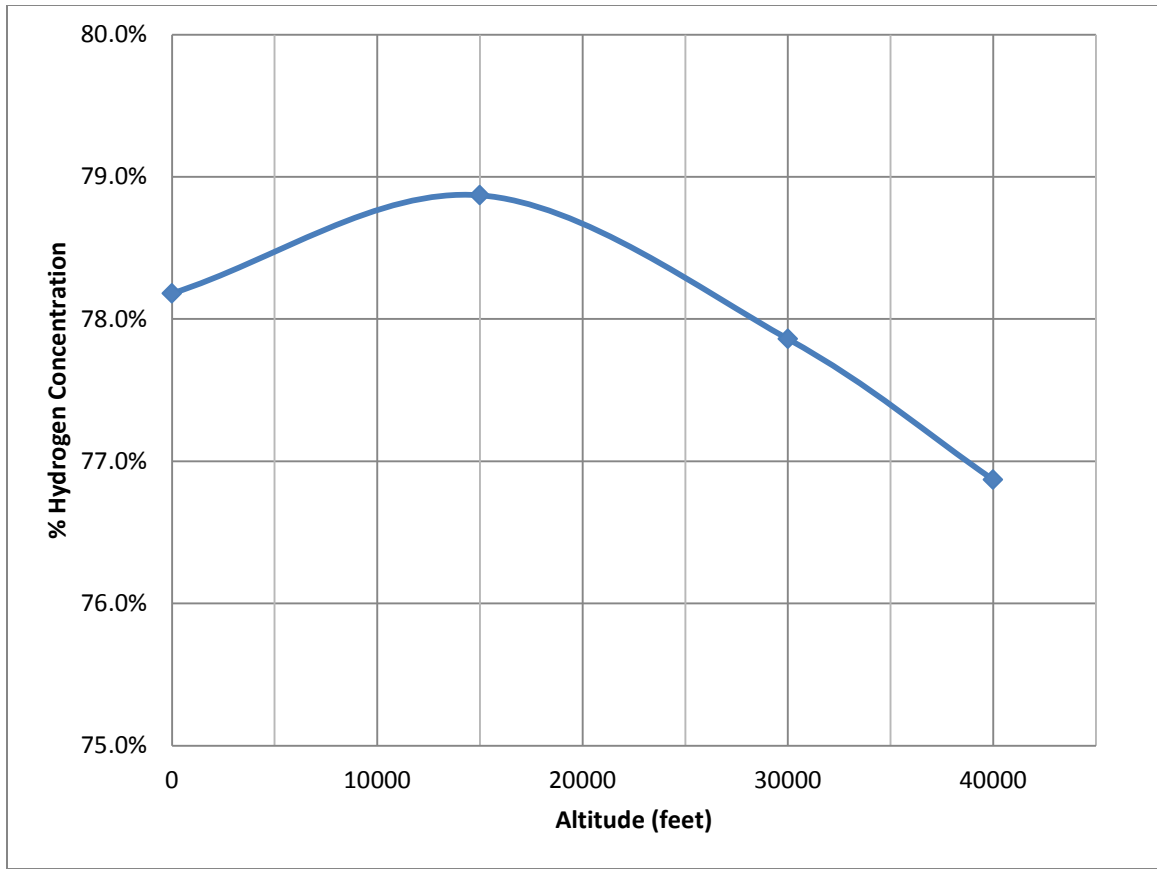


Figure 3.6: Hydrogen upper flammability limit in air as a function of altitude. The decrease in the UFL above 15,000 feet is due to not having enough spark energy.

Elevation (ft.)	Upper Limit
0	78.18%
15,000	78.87%
30,000	77.86%
40,000	76.87%

Table 3.2: Hydrogen upper flammability limit at each altitude using a 3% pressure rise criterion.

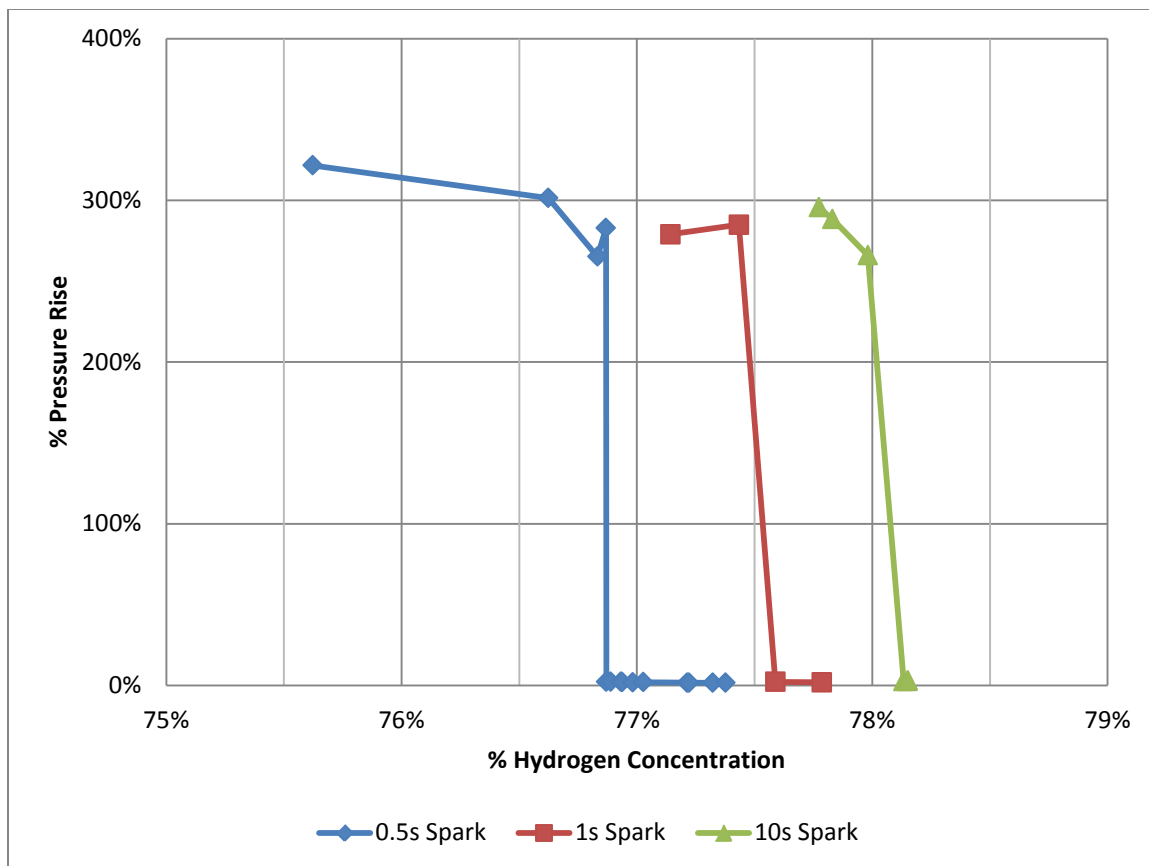


Figure 3.7: Upper flammability limit at 40,000 feet with different spark durations. The increased spark duration results in higher spark energy, raising the flammability limit at high altitudes. This shows that the UFL at high altitudes was limited by the spark energy.

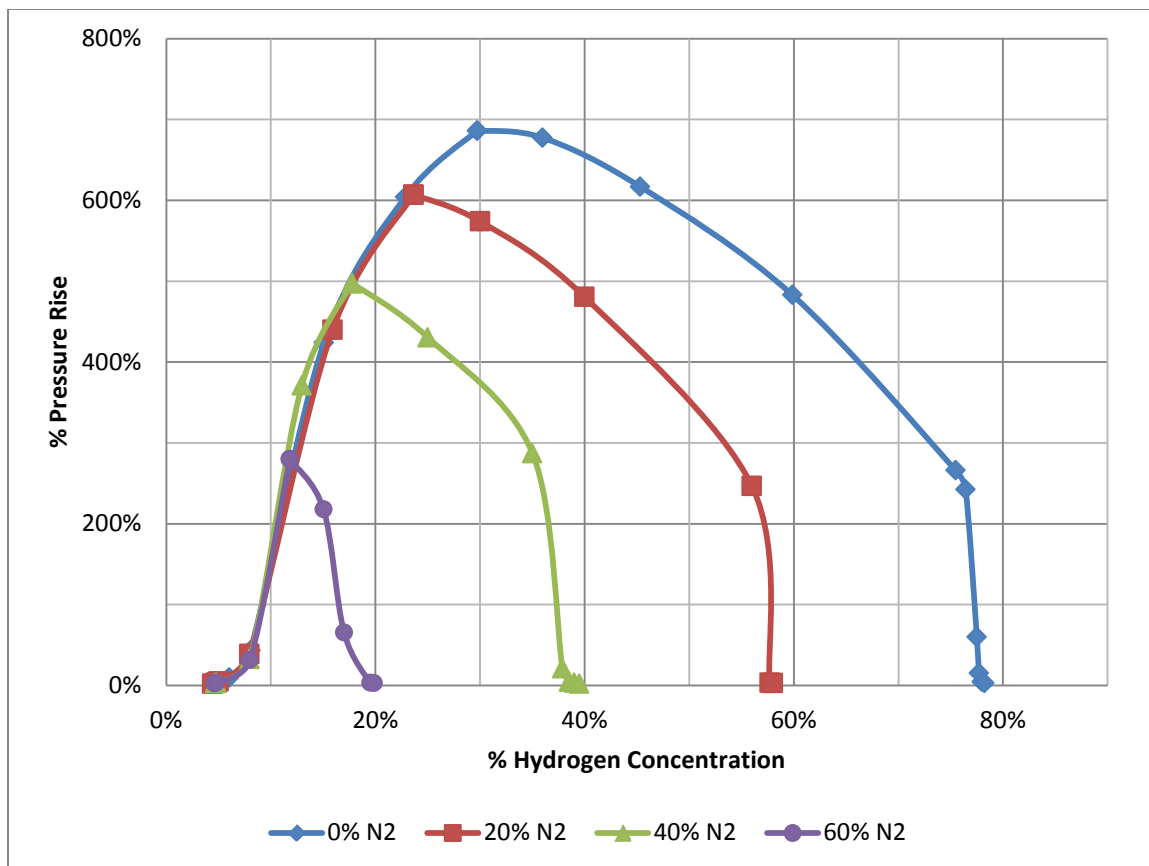


Figure 3.8: Atmospheric pressure hydrogen flammability in air with varying added nitrogen concentrations. Adding nitrogen depletes the air of oxygen resulting in lower UFLs and lower explosion pressures.

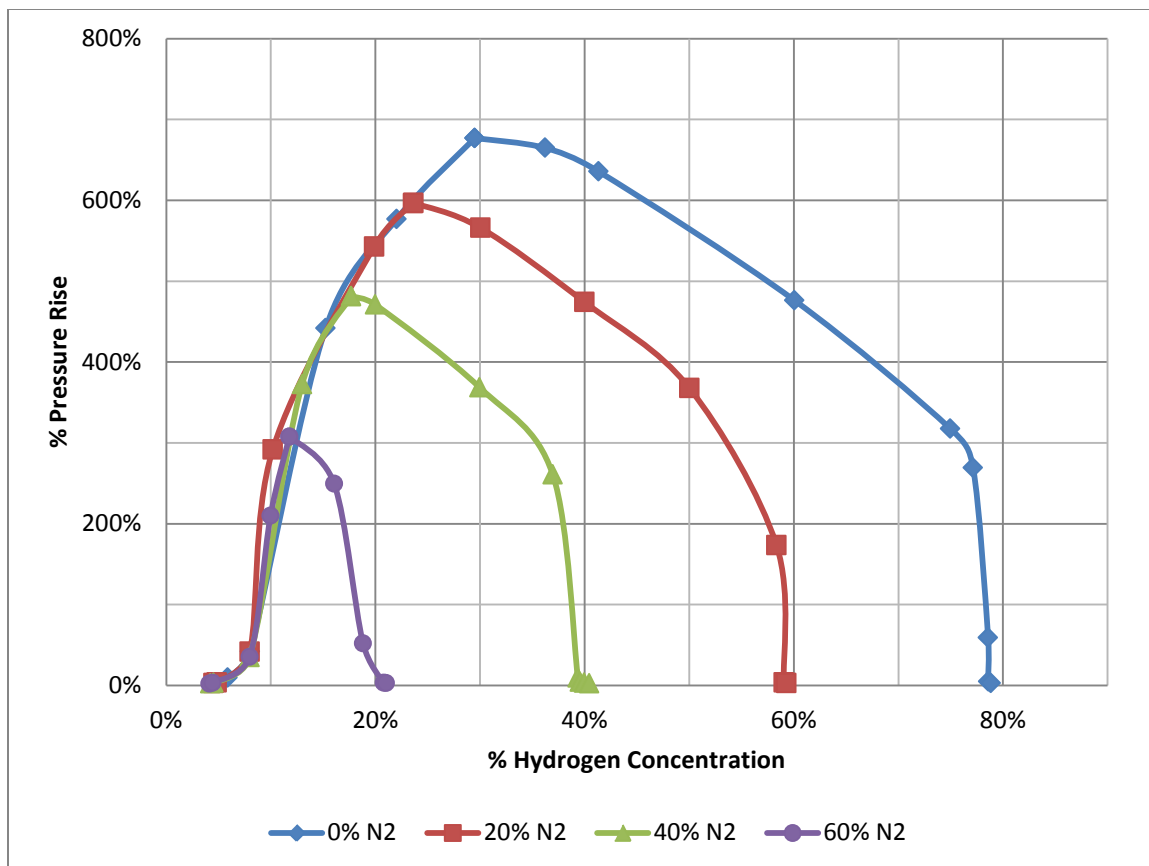


Figure 3.9: 15,000 feet hydrogen flammability in air with varying added nitrogen concentrations. Adding nitrogen depletes the air of oxygen resulting in lower UFLs and lower explosion pressures.

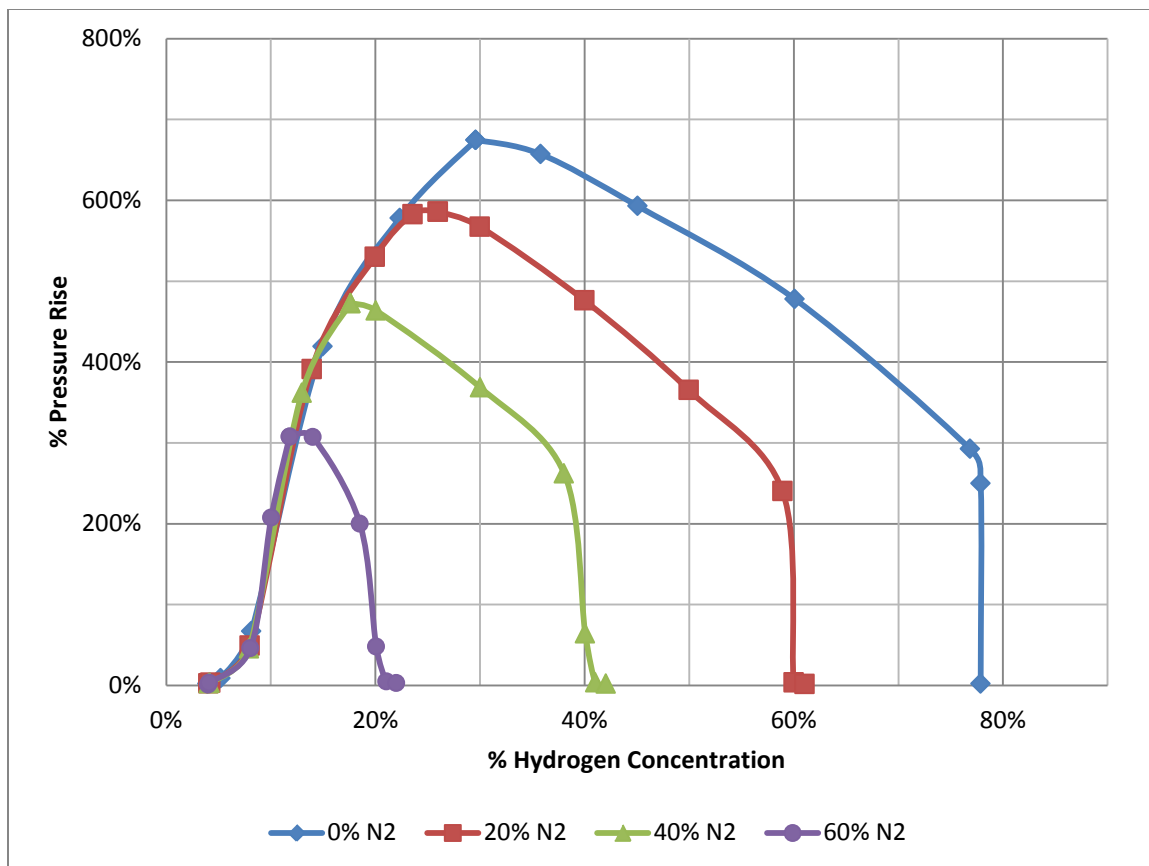


Figure 3.10: 30,000 feet hydrogen flammability in air with varying added nitrogen concentrations. Adding nitrogen depletes the air of oxygen resulting in lower UFLs and lower explosion pressures.

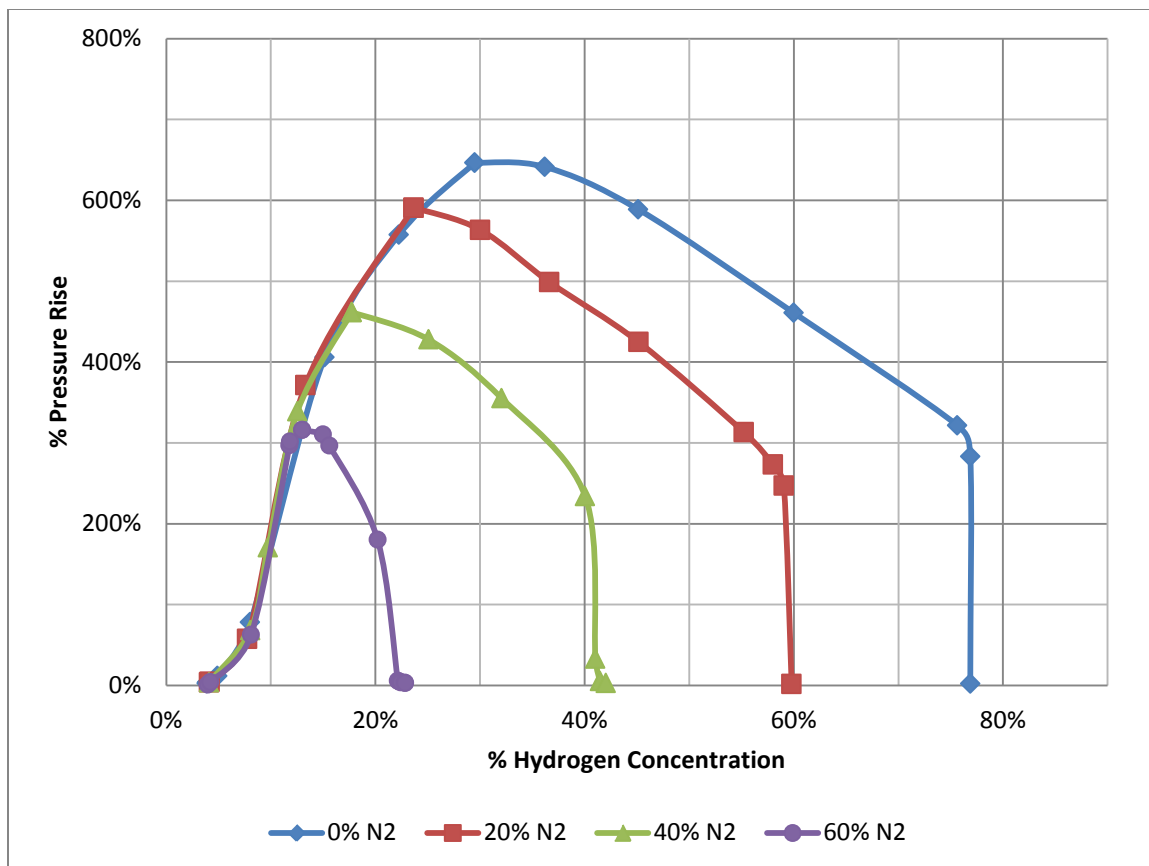


Figure 3.11: 40,000 feet hydrogen flammability in air with varying added nitrogen concentrations. Adding nitrogen depletes the air of oxygen resulting in lower UFLs and lower explosion pressures.

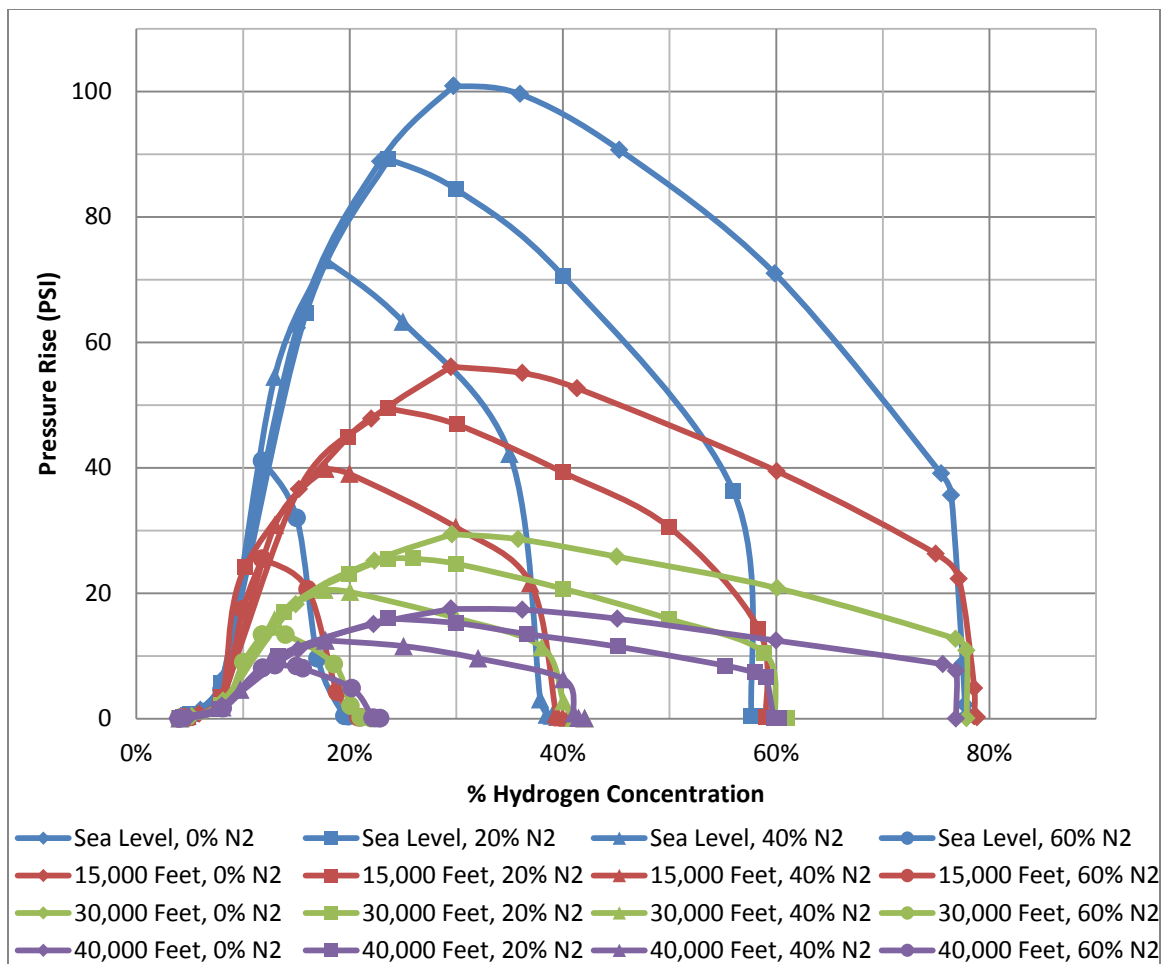


Figure 3.12: Peak explosion over-pressure at varying altitudes and nitrogen concentrations. The flammability limits at each nitrogen concentration widen as the altitude increases, but the peak explosion pressures at each altitude are lower because of the reduced mass of hydrogen and oxygen present.

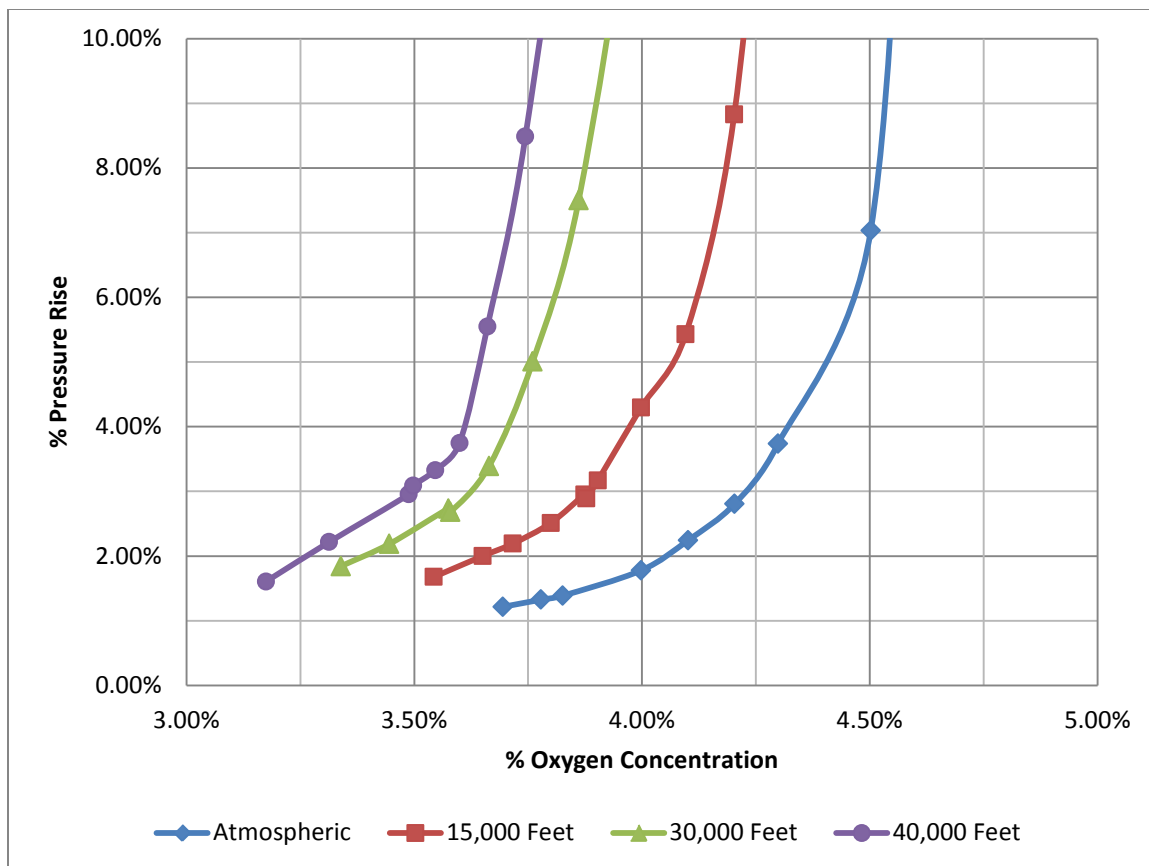


Figure 3.13: Explosion pressure data near the limiting oxygen concentration of hydrogen. Each point shows the peak pressure rise of a single test as a percentage of the initial pressure. A 3% pressure rise or more is considered flammable.

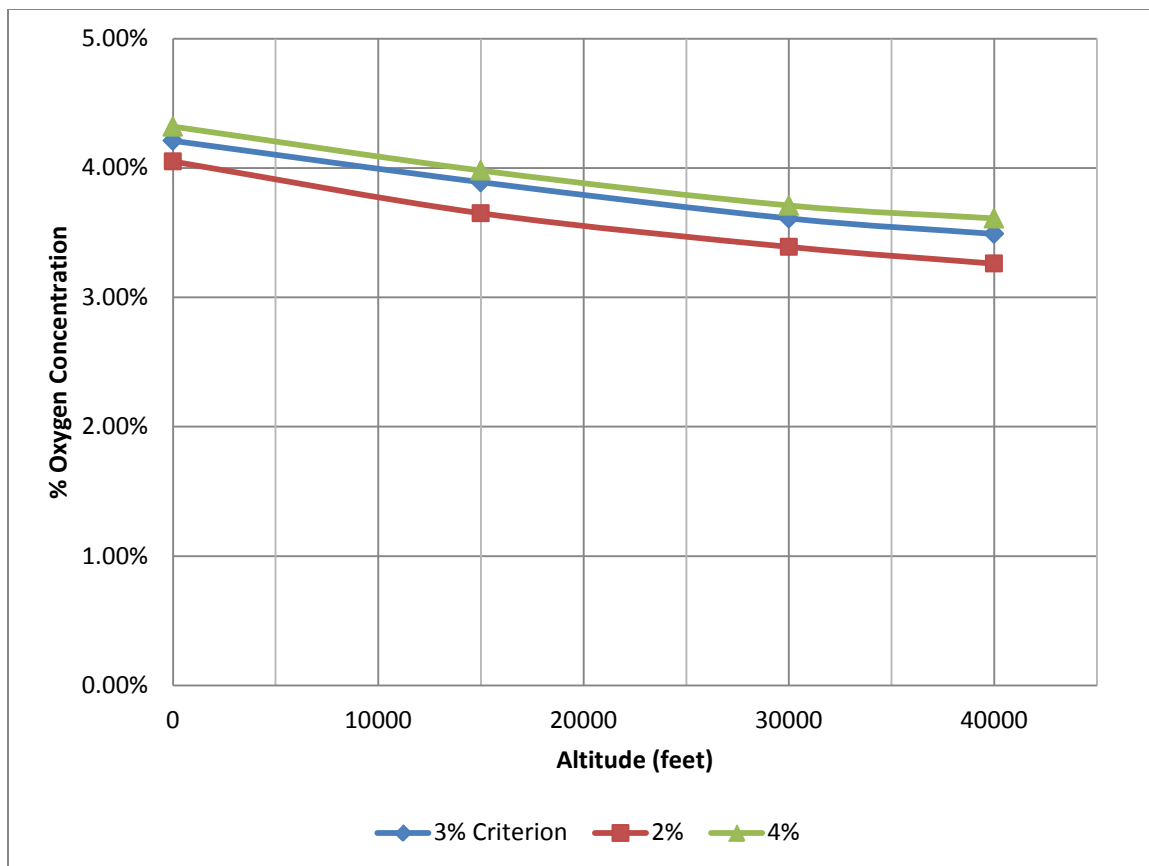


Figure 3.14: Limiting oxygen concentration of hydrogen as a function of altitude using three different flammability criteria. Also shown is a table for the exact values using a 3% pressure rise criterion.

3% Criterion	
Elevation (ft.)	Limiting O ₂ %
0	4.21%
15,000	3.89%
30,000	3.61%
40,000	3.49%

Table 3.3: Hydrogen limiting oxygen concentration at each altitude using a 3% pressure rise criterion.

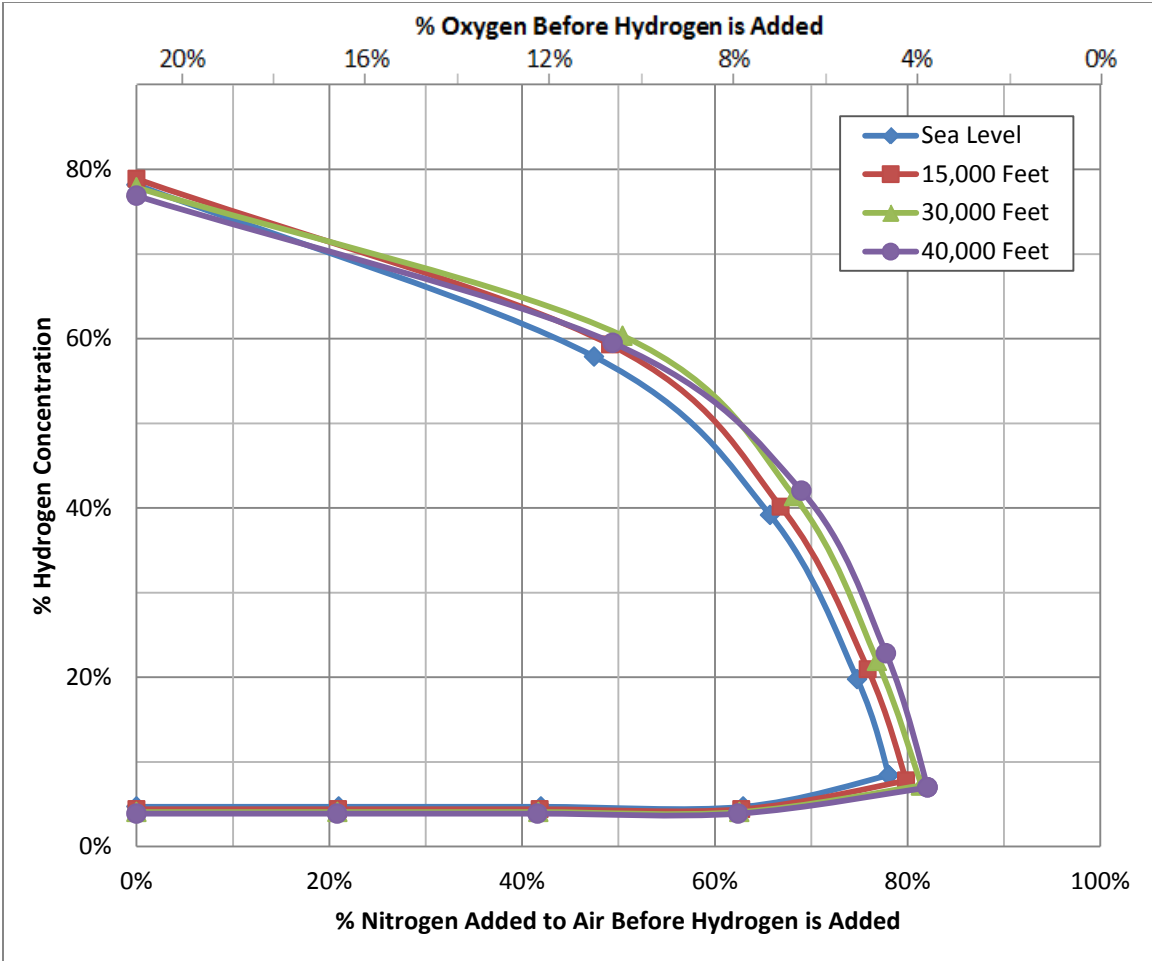


Figure 3.15: Flammability limits of all possible combinations of hydrogen, oxygen, and nitrogen at each altitude. Every combination inside of the curves is considered flammable. The amount of nitrogen and oxygen shown would be the amount needed before the hydrogen is added. The added hydrogen would then displace part of that mixture.

Chapter 4: Conclusion

The flammability of hydrogen was tested in a 20 L vessel at air pressures simulating sea level up to 40,000 feet of altitude using a flammability criterion of a 3% pressure rise over the initial pressure. The lower flammability limit was 4.70% hydrogen at atmospheric pressure and decreased with increasing altitude, reducing to 3.89% hydrogen at 40,000 feet. The upper flammability limit was 78.18% hydrogen at sea level, increased to 78.87% at 15,000 feet, then decreased to 76.87% at 40,000 feet. This data showed good agreement with previous experiments and followed the trend of widening flammability limits as the altitude increased except for the UFL at 30,000 and 40,000 feet. Those two tests were limited by the spark energy instead of the rich limit of hydrogen. This limited spark energy did not affect the other tests however.

With the use of added nitrogen, the peak pressure increase across the full range of hydrogen and oxygen concentrations was found as well. At high altitudes, the gas mixtures remained flammable across a wider range of hydrogen concentrations, but the lower altitude tests produced much higher peak pressures due to the increased mass of hydrogen and oxygen present. The highest pressure reached out all of the tests performed was the stoichiometric hydrogen and air test at sea level. The pressure rose 100.8 psi over the initial pressure 12 ms after the spark was triggered. This corresponded to a 686% pressure rise.

The limiting oxygen concentration at each altitude was found to decrease as the altitude increased, following the same trend as the other flammability limits that widened with increased altitude. The LOC was 4.21% oxygen at atmospheric pressure and decreased to 3.49% oxygen at 40,000 feet.

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