COMBUSTION CHARACTERISTICS OF HYDROGEN AS USED IN A FLAMMABLE TEST MIXTURE

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Abstract

Flammable test mixtures based on hydrogen are widely used in the aerospace industry to determine ignition thresholds of simulated lightning strikes and other ignition sources. Reasons include the facts that hydrogen has low minimum ignition energy; near the flammability limit the flame fronts propagate slowly and produce modest overpressures; and H₂O is the main combustion product. Most significantly for lightning testing, the low luminosity of hydrogen flames mean they can be used in conjunction with photographic test methods, unlike the hydrocarbon-based gas mixtures. Variability in ignition test results is often attributed to the intrinsic randomness of the threshold ignition process or variations in ignition source strength. However, we show that this may actually be due to the test mixture itself, particularly when using hydrogen in low concentrations. Some insight into the behavior of hydrogen combustion near the flammability limits is needed in interpreting the results of these tests and designing test programs. This paper presents the results of experimental studies on typical test mixtures near the lower limits of combustion. High-speed video schlieren and transient pressure measurements were taken to study ignition and flame propagation events in an 11.75-liter pressure vessel filled with hydrogen-oxygen-argon mixtures. A range of gas compositions was examined to bracket those of interest for lightning testing.

Introduction

In assessing the ignition threat to fuel tank vapor spaces due to lightning strikes on aircraft, the industry refers to the SAE Aerospace Recommended Practice 5416 Aircraft Lightning Test Methods (Ref 1), and the European equivalent ED 105 Lightning Testing Document. The recommended method for testing ignition sources is to use a flammable mixture consisting of 5% hydrogen-12% oxygen-83% argon by volume. This mixture has been selected to meet the requirement that the flammable mixture has a 90% or greater probability of ignition with a 200 μJ voltage spark source. The foundation of this work is published in the DOT/FAA/CT94/74 Aircraft Fuel System Lightning Protection Design and Qualification Test Procedures Development (Ref 2).

The mixture recommended in ARP 5416 is deliberately close to the lean flammability limit. Using mixtures so close to the lean flammability limit to determine incendivity creates a serious problem due to the difficulty of defining ignition limits in these situations. Britton (Ref 3) has discussed this issue in regards to standardized testing for determining flammability limits and the disparity between the results of various test methods. He points out the difficulty of defining a combustion event, even when pressure rise is measured, for near-limit cases for which only a narrow cone of the reactants is burned, producing a very small pressure rise. This is the same issue that we have identified in using mixtures with less than 6% hydrogen with the added complication of the unusual behavior of flames in lean hydrogen mixtures.

Flames in near-limit hydrogen-oxygen-diluent mixtures (Ref 4) are a special case. The high mass diffusivity of hydrogen molecules in the reactant mixture enables combustion to take place for extremely lean mixtures with very low flame temperatures as compared to hydrocarbon fuels near the flammability limit (Ref 5). The low temperature results in very low flame speeds and the flames are sensitive to fluid motion (e.g., turbulence), flame stretching due to motion associated with the buoyant rise of the hot combustion products (Ref 6,7), and radiation losses (Ref 8). As a consequence, the extent of combustion and resulting pressure rise are very sensitive to the experimental setup, as discussed by Cashdollar et al. in 2000 (Ref 9). This behavior has been known since the earliest studies (Ref 5) on hydrogen flammability and leads to the very large difference between "upward" (4% hydrogen) and "downward" (8% hydrogen) flammability limits for hydrogen-air mixtures. This has been extensively studied in the context of nuclear safety and the potential for hydrogen explosions following loss-of-coolant accidents.

Although many of the cited studies are concerned with finding the limits of flammability (Ref 5, 10) in terms of the critical mixture composition for a given (very strong) ignition source, the same considerations apply to using similar mixtures to determine the limiting strength of an ignition source. Ignition energy data are available for hydrogen-oxygen-diluent mixtures (e.g. Figure 165 in Lewis and von Elbe) (Ref 11) but there is no discussion of the difficulty of detecting ignition in engineering testing situations similar to those encountered when using the ARP 5416 guidelines. The specific issue of ignition by the growth of a flame from a hot gas volume has been
considered by a number of authors, for example Kusharin et al (2000) (Ref 8) for hydrogen-air and Mass and Warnatz (1988) (Ref 12) for hydrogen-oxygen. However, the determination of flammability or incendivity limits remains an empirical topic that relies primarily on experimental testing (Ref 10, 13).

The traditional viewpoint of combustion science (Ref 11) is that the ignition thresholds of fuels are characterized by the minimum ignition energy (MIE). The standard test methods (Ref 10, 13) for determining MIE rely on discharging a capacitor with a known energy through a specified gap. The pioneering work was done by Guest, Blanc, Lewis and von Elbe at the Bureau of Mines in the 1940s (Ref 11) and the data they obtained are still extensively cited by handbooks (Ref 10, 13). Improvements on this technique have been suggested by many authors, most recently by Ono et al at the University of Tokyo (Ref 14, 15).

In most instances, the ignition energy that is reported is the energy stored in the capacitor used in the discharge circuit

\[ E_{\text{stored}} = \frac{1}{2} CV^2 \]

(Ref 11) or the energy that is discharged into the spark gap (Ref 14, 15). However, not all of this energy is useful for creating a critical ignition kernel. The stored energy not only heats the gas in the spark channel, but also creates sound waves, electromagnetic radiation, visible light, IR emission, circuit losses, and some is left as residual charge in the capacitor. Only a fraction of the stored energy contributes to the thermal energy that heats up the gas and initiates combustion. How much of the stored energy is lost through each of these processes is dependent upon the particular circuit parameters so that the MIE depends on the test method itself. The MIE is considered to be a threshold value defined so that ignition consistently occurs above that level and does not occur below it (Ref 11). Extensive tabulations are available (Ref 10, 13) for MIE values determined in that fashion and the MIE is found to be a function of composition, electrical circuit, and spark gap size and construction. Such values are used throughout the electrical industry in designing explosion proof equipment (Ref 13). However, the combustion characterization used in the DOT/FAA/CT-94/74 takes a different approach that is more consistent with engineering test analysis. Ignition is treated as a statistical event and the outcome of a series of tests is used to define ignition probability as a function of some characteristic of the electrical circuit such as stored energy or peak current. It is reasonable to consider the outcome of engineering testing as having inherent variability and using statistical methods to analyze the results provides a more substantial basis for evaluating the risks associated with lightning strikes and similar potential threats to safety.

An example of this type of analysis is shown in Figure 1 for Jet A ignition tests performed by Lee and Shepherd at the California Institute of Technology using a capacitive spark discharge as the ignition source (Ref 16). Figure 1(a) shows the results of the 25 ignition tests plotted vs. spark energy, with a black diamond indicating ignition (“Go”) and a white diamond indicating no ignition (“No Go”). Figure 1(b) shows a probability distribution derived from the data using a logistic regression, with the original data points shown as well. The probability distribution has a mean value and a 95% confidence interval (indicated by the dotted lines), but no single threshold value like in the MIE view of ignition. This method of data analysis using a logistic probability distribution has been recently applied to hot surface ignition tests for automotive and aviation liquids (Ref 17). Introducing statistical methods of analysis introduces a key question. Is the statistical nature of the data due to an intrinsic probabilistic nature of ignition itself, or is it due to uncontrolled conditions in the experiments? Clearly, it is desirable that the experimental variability be minimized and quantified in order to have meaningful statistical results.

In ignition testing there are many uncontrolled sources of variability that can possibly contribute to inaccurate and unreliable test results. One major source of variability is the gas filling process, which can lead to variations in the flammable mixture composition. Another important cause of variability is the ignition source itself; the source must be reliable and repeatable, meaning every time it is initiated it produces an ignition source of known energy. A commonly used ignition source is a capacitive spark discharge, which then requires clear circuit characterization as well as attention to the spark gap size and condition of the electrodes. The degree of turbulence in the explosion vessel is a third possible source of variability, as the process of flame initiation and flame propagation can be affected by any turbulence in the gases. Finally, a fourth major source of variability in ignition testing is the method used to detect the ignition. There are many possible ways to detect a combustion event, such as pressure transducers, thermocouples, pressure relief vents covered with foil and visible light. However, if the detection method is unreliable or uncharacterized then the test results are also unreliable. The possible sources of variability are not limited to the four described above, but these are major issues that can substantially influence the accuracy of the outcomes from ignition tests and hence will be addressed in this work.

The goal of the present tests is to examine the characteristics and detection of lean hydrogen-oxygen-argon combustion near the flammability limit in a carefully controlled environment. In this test program we varied the hydrogen concentration at a fixed diluent to oxidizer ratio

\[ \text{Ar/O}_2 = 6.917 \]
to simulate typical mixtures used in lightning testing. In order to observe the detectable limits of the flammable mixture itself and not the limits of the ignition source, we use an extremely reliable ignition system with a spark energy well above the reported MIE of hydrogen, which removes uncertainty related to the ignition source. We also use the method of partial pressures and a carefully controlled plumbing system to fill the gases precisely and hence have accurate and repeatable mixture compositions. While the issue of turbulence and its effect on ignition is not addressed quantitatively in the present tests, there is a consistent period of time (90 seconds) between mixing and spark ignition so as to remove variability between the tests due to differing degrees of turbulence.

Although the recommended test method utilizes an open-box configuration where the effective volume is not constant, our tests are done using a closed-box constant volume chamber to eliminate the variability that is specific to each open-box configuration. A commonly used method of detecting combustion is to observe the pressure relief via an aperture covered by aluminum foil on the test vessel. For low hydrogen concentrations, however, the pressure rise can be very small and so a more sensitive detection method is needed to reliably detect ignition. Also, because low-concentration hydrogen combustion produces buoyant flames, the location of the aperture becomes important. To remove uncertainty relating to ignition detection, three sensitive and reliable detection methods are used in these tests. The most sensitive method is visualization of the flame using a schlieren system and high-speed video camera. The next most sensitive method is detecting the pressure rise using a pressure transducer. Finally, a thermocouple is also used to detect the temperature rise due to combustion. Having addressed all these sources of variability, a consistent and well-controlled experiment exists to examine combustion characteristics.

The experiments were conducted in a closed combustion vessel 11.75-liter in volume. It is constructed of steel slabs that form a rectangular chamber with internal dimensions of 7.48 in. x 8.0 in. x 12.0 in. Four walls are constructed of 1.25 in. thick steel plates. Two parallel steel vessel walls have 4.6 in. diameter, 1 in. thick glass windows for visualization. Each of the other two vessel walls holds an electrode for the ignition source. The vessel has a 1.5 in. thick steel base and lid. The lid has various holes for the pressure sensors, thermocouple, mixing fan, and plumbing.

The plumbing includes a 1.0 in. ball valve between the vessel and the vacuum line for evacuating the chamber. A gas feed line connected to a series of valves is used with the lab control system to fill various gases using the method of partial pressures. The static pressure is measured using a Heise model 901A digital optical manometer with a precise readout, allowing for filling of gases to within 0.01 kPa and therefore precise determination of mixture composition. The vacuum is also connected to the gas feed lines so that existing gases can be evacuated before filling with a new gas to eliminate errors in composition due to dead volume. The vessel and main features of the plumbing system are labeled in Figure 2.

**Figure 1.** (a) Ignition test data vs. spark energy for Jet A (Ref 7). (b) Probability distribution of ignition vs. spark energy obtained by performing a logistic regression on the Jet A ignition data in Figure 1(a). Dashed lines are 95% confidence intervals.

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**Experimental Setup**

**Combustion Vessel and Flammable Mixture**

The ignition source is a capacitive discharge through a transformer to create a spark between two electrodes.
Figure 3 is a schematic of the electrical circuit used to produce the capacitive discharge. The output of a voltage doubler circuit is used to charge a 5 μF capacitor through two resistors (33 and 30 kΩ) to 380 VDC, producing 361 mJ of stored energy in the circuit. An optocoupler is triggered using an external 12 V TTL signal, which then allows the capacitor to discharge through an SCR and a TR-2012 trigger transformer. The transformer steps the voltage up to about 25 kV, which causes electrical breakdown of the gap, a spark channel, and an initially cylindrical ignition kernel in the gas between the electrodes. The total energy stored in the capacitor is 361 mJ and the amount actually delivered to the gas in the form of thermal energy is less but unknown. However, this is a very large amount of energy and it far exceeds the upper limits of MIE data for lean hydrogen-oxygen-argon mixtures (Ref 11), so it is reasonable to expect that this is sufficient energy to ignite the mixtures of interest in this study. The capacitor is charged and the circuit functioned using voltage and TTL signals from the experiment control panel. The tungsten wire electrodes are 0.38 mm in diameter with a gap of approximately 4.75 mm. The spark gap is positioned approximately in the center of the combustion chamber.

Detection Methods

Three methods were used to detect both the onset of combustion as well as the magnitude of the event. A thermocouple, Omega K type with 24 AWG wires and a weld bead size of approximately 1.5 mm, was inserted through the top of the vessel to measure the gas temperature inside the chamber. An Omega model DP116 electronic temperature readout was used to convert the thermocouple output to temperature. A thermally-protected Endevco piezoelectric pressure transducer, model 8530B, was used to monitor the chamber pressure. The dynamic pressure and temperature signals were recorded by LabVIEW Data Acquisition Software running on a personal computer. The final pressure of the products was measured with the Heise transducer.

A schlieren system was used to observe the flame initiation and propagation through the windows on the combustion vessel. A schematic of the schlieren optical system is shown in Figure 4. A Phantom v5.0 video camera was used to record high-speed schlieren images at a rate of 1000 frames per second with a resolution of 1024 x 1024.
Results

Mixtures with hydrogen concentrations ranging from 3%, just below the flammability limit, to 13% were examined using the experiment and detection methods discussed above (Table 1). The tests were conducted to study ignition as a function of hydrogen concentration, detection method, and initial gas motion and turbulence.

Pressure Detection of Ignition and Ignition Sensitivity to Hydrogen Concentration

The sensitivity of ignition tests to hydrogen concentration can be assessed by observing the pressure rise generated by the combustion. The form of the experimental pressure trace is determined by the competition between the rate of thermal energy addition due to combustion and the loss of energy due to heat transfer to the chamber. Figure 5 shows the normalized pressures versus time traces measured for hydrogen concentrations varying from 3%, where no combustion occurs because the mixture is below the flammability limit, to 8%. It is clear from the pressure traces that a threshold exists at 6% hydrogen concentration, above which complete combustion occurs and the peak pressure exceeds 2.5 times the initial pressure. Below this threshold only partial combustion occurs and the peak pressures do not exceed 1.5 times the initial pressure. The pressure transducer used is sensitive enough to detect these small pressure rises near the flammability limit, but a less sensitive detection method may not detect the combustion event at all. The 6% hydrogen case also produces a pressure trace with multiple local peaks, whereas the other cases have the expected trend of rapid rise to a peak pressure, then decay due to heat loss to the vessel. Cases with hydrogen concentrations as high as 13% were tested, but as expected, mixtures with concentrations above 7% follow the same trend with increasing peak pressures.

The high-speed schlieren visualization was also used as an extremely reliable ignition detection method. This method provides visualization of the flame evolution due to the competition between the flame speed and inertia of the gases and buoyancy. Combustion with 5% hydrogen concentration (Figure 6) produces a slow, buoyant flame that propagates outward as well as upward. With hydrogen concentrations below the 6% threshold, the flame speeds are slow enough that buoyancy becomes dominant and the flame is forced to the top of the vessel where the flame is quenched. This quenching prevents complete combustion, with only a small cone of the fuel being consumed resulting in a very small pressure rise. Thus the pressure trace (Figure 5) has a longer time-to-peak and a much lower peak pressure than those for hydrogen concentrations above the 6% threshold. Alternative detection methods such as aluminum foil deformation or thermal flame front measurements may not be able to detect these partial combustion events due to insufficient overpressures or misplacement of the detection device relative to the flame motion.

The case of 6% hydrogen concentration (Figure 7) is a marginal case where the effect of buoyancy is nearly balanced by flame front propagation. The flame is slow enough that buoyancy has a large effect and the flame propagates upwards and the upper surface flame is quenched at the top of the vessel. However, unlike the 5%-hydrogen case, the gases have enough inertia and the flame speed is high enough that the flame can continue to propagate downwards, and with assistance from convection induced by the flame, nearly complete combustion occurs. This leads to the two-peak pressure trace (Figure 5) that exhibits a higher overall peak pressure and a smaller time-to-peak than the cases with hydrogen concentration below the 6% threshold.

At a 7% hydrogen concentration (Figure 8) the flame speed is high enough to counteract the buoyancy effects. Therefore the combustion is characterized by a quasi-spherical flame front that propagates outward with a small amount upward motion of the flame ball due to buoyancy. The flame is highly unstable under these conditions and a cellular or folded structure is observed. Relatively complete combustion is achieved and the pressure rise is approximately 80% of the adiabatic value. These characteristics are manifested in the very short time-to-peak of about 230 ms and the significantly higher peak pressure of 3.89 bar (Figure 5). Because mixtures with hydrogen concentrations greater than the threshold 6% have significantly higher peak pressures, less sensitive detection methods can be used; however, since a mere 1% error in hydrogen concentration can lead to such drastically different combustion
characteristics, mixture composition accuracy becomes extremely important.

Figure 5: Normalized pressure traces for hydrogen concentrations of 3 to 8%.

Table 1: List of test data, including actual gas compositions achieved, peak pressure, and verification of ignition/no-ignition determined from the schlieren visualization.

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Figure 6: Images from schlieren video of combustion with 5% hydrogen.

Figure 7: Images from schlieren video of combustion with 6% hydrogen.

Figure 8: Images from schlieren video of combustion with 7% hydrogen.
The normalized peak pressures versus hydrogen concentrations for all tests that we carried out are plotted in Figure 9, along with the theoretical curve given by constant volume, adiabatic, equilibrium calculations performed using Cantera, a software package for problems involving chemically reacting flows (Ref 18). As expected, the peak pressures for hydrogen concentrations above 6% follow the same trend as the theoretical pressures, but the experimental values are on average 0.67 bar lower than the theoretical values since the calculations do not account for heat loss. For these ignition tests, there is a threshold at a 6% hydrogen concentration where downward flame propagation and complete combustion occurs. This threshold is strongly dependent on the vertical location of the ignition source within the combustion vessel. In our tests, the ignition source was located approximately in the center of the vessel, however, other work has found that the threshold concentration for a downward-propagating flame increases as the ignition source location nears the top of the combustion vessel (Ref 19). The case of 6% hydrogen concentration is a marginal case where the effects of buoyancy nearly counteract the flame speed and the inertia of the gases. The competition among these forces leads to a combustion event on the order of 1 second in length, with irregular flame front motion and a longer time-to-peak and lower peak pressure than cases above the threshold concentration. Mixtures with hydrogen concentrations below this threshold do not undergo complete combustion and the resulting peak pressures are small even when measured under constant volume conditions. These peak pressures are only about 30% of the theoretical pressures calculated assuming complete combustion.

**Figure 9:** Normalized peak pressure vs. percent hydrogen, experimental data points and theoretical curve for constant volume, adiabatic, complete combustion.

**Thermal Detection of Ignition**

Using a fairly common thermocouple configuration, as described in the Experimental Setup, temperature versus time traces were obtained for hydrogen concentrations ranging from 3% to 8% (Figure 10). Because of its positioning at the top of the vessel, the thermocouple measured both the rise in temperature of the unburned gas due to compression by the advancing flame and also the hot combustion products. The thermocouple used is neither the most sensitive nor the fastest for commercially available temperature measurement methods. Therefore the measurement is not intended to be numerically accurate but rather it is to be used to assess the viability of using temperature as a ignition detection method for these lean hydrogen mixtures. In spite of the limitations of the thermocouple, in these ignition tests the combustion was clearly evident in the temperature traces (Figure 10) and so thermal ignition detection was successful for the full range of hydrogen concentrations. However, as with the pressure detection method, placement of the thermocouple is vitally important; if the thermocouple had not been at the top of the vessel, it may not have detected the ignition for cases below the 6% hydrogen threshold.

![Figure 10: Temperature traces for hydrogen concentrations of 3 to 8%.](image)

**Effect of Gas Motion and Turbulence**

Many studies have been conducted to assess the effects of turbulence on flammability, including extensive studies of hydrogen combustion under turbulent conditions in large scale testing (Ref 9, 19). Turbulent motion of the gas in the vicinity of the spark discharge influences both the ignition and flame propagation processes (Ref 19). Higher flow velocities and turbulence intensities may increase the MIE (Ref 20).
However, once a flame is initiated, flame front folding by turbulence can significantly increase the effective flame speed compared to values observed in quiescent systems (Ref 19). The effect of having some initial gas motion versus a quiescent mixture was briefly examined in the present tests for a 6% hydrogen mixture. Gas motion was introduced by operating a mixing fan at the top of the vessel, and the spark was initiated immediately after the fan was stopped, leaving some initial gas motion at the time of ignition. From comparison of pressure traces from both the quiescent and non-quiescent cases (Figure 11), it is clear that the initial gas motion increases the initial energy release leading to a higher flame speed. Thus more of the fuel is burned earlier in the event, and the pressure increases faster initially than in the quiescent case, consistent with observations in hydrogen-air testing (Ref 9, 19). Differences in the flame front evolution can also be clearly seen in the schlieren video (Figure 12), with increased downward propagation of the flame initially in the non-quiescent case. While the gas motion and turbulence are not quantified in this study, it has been shown qualitatively that it is another aspect of the ignition experiment that must be controlled to reduce variability.

Figure 11: Normalized pressure traces for 6% hydrogen concentration with little gas motion (solid line) and with a higher degree of motion (dashed line).

Figure 12: Schlieren images of combustion with 6% hydrogen with little initial gas motion (a) and with a higher degree of motion (b).

Conclusions

The present tests were performed using an experimental design that minimized variability due to the ignition source, spark gap, mixture composition, and gas motion. This allowed for an in-depth characterization of the ignition and flame propagation in lean hydrogen-oxygen-argon mixtures near the flammability limit. For mixtures between 3 and 6% hydrogen (including the ARP 5416 recommended 5% hydrogen case), the flame motion is dominated by buoyancy, only a small portion of the volume burns, and relatively low pressure rises are observed. For hydrogen concentrations greater 6%, the combustion is relatively fast, the entire volume burns, and the overpressures are sufficiently high that even the crudest methods will detect ignition.

In mixtures with hydrogen concentrations lower than 6%, ignition and flame propagation are highly sensitive to igniter location, gas flow and turbulence intensity, and the precise value of the hydrogen concentration. As a consequence, ignition tests conducted with less than 6% hydrogen and an insensitive detection method may give a false no-ignition result. These ignition events were successfully detected by the three methods used in our tests, but the overpressures generated by the buoyant flames may not be sufficient for a less sensitive method, such as observing the deformation of a thin film covering an aperture, as often used in lightning testing.

The present results represent a highly controlled situation. However, more typical engineering tests often use open combustion chambers and geometries which introduce additional variability. Using large-volume, vented chambers with low hydrogen concentrations may result in small pressure rises and localized thermal signatures, requiring sensitive ignition detection methods.

The tests performed in this work show that the combustion process is repeatable for the ARP 5416 mixture but very sensitive to the test conditions. If conditions are not carefully controlled the test results
may appear to be random in nature and require a large sample size to achieve a given confidence interval. The number of tests required can be reduced by eliminating uncontrolled parameters. Particular attention should be given to controlling the gas composition when using mixtures with less than 6% hydrogen.

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References