

VAPOUR CLOUD EXPLOSIONS FROM THE IGNITION OF METHANE/HYDROGEN/AIR MIXTURES IN A CONGESTED REGION

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ABSTRACT

To facilitate the transition to the hydrogen economy the EU project NATURALHY is studying the potential for the existing natural gas pipeline networks to transport hydrogen, together with natural gas, to end-users. Hydrogen may then be extracted for hydrogen fuel-cell applications, or the mixture used directly by consumers in existing gas-fired equipment, with the benefit of lower carbon emissions. The existing gas pipeline networks are designed, constructed and operated to safely transport natural gas, mostly methane. However, hydrogen has significantly different properties that may adversely affect both the integrity of the network, and thereby increase the likelihood of an accidental leak, and the consequences if the leak finds a source of ignition. Consequently, a major part of the NATURALHY project is focused on assessing how much hydrogen could be introduced into the network without adversely impacting on the safety of the network and the risk to the public. Hydrogen is more reactive than natural gas so the severity of an explosion following an accidental leak may be increased. This paper describes field-scale experiments conducted to measure the overpressures generated by ignition of methane/hydrogen/air mixtures in a congested but unconfined region. Such regions may be found in the gas handling and metering stations of the pipeline networks. The 3 m x 3 m x 2 m high congested region studied contained layers of pipes. The composition of the methane/hydrogen mixture used was varied from 0% hydrogen to 100% hydrogen. On the basis of the experiments performed, the maximum overpressures generated by methane/hydrogen mixtures with 25% (by volume) or less hydrogen content are not likely to be much more than those generated by methane alone. Greater percentages of hydrogen did significantly increase the explosion overpressure.

1.0 INTRODUCTION

Hydrogen is seen as an important energy carrier for the future which offers carbon free emissions at the point of use. However, transition to the hydrogen economy is likely to be lengthy and will take considerable investment with major changes to the technologies required for the manufacture, transport and use of hydrogen. In order to facilitate the transition to the hydrogen economy, the EC funded project NATURALHY is studying the potential for the existing natural gas pipeline networks to transport hydrogen from manufacturing sites to hydrogen users. The hydrogen, introduced into the pipeline network, would mix with the natural gas. This mixture could then be used directly by consumers as a fuel within existing gas powered equipment, with the benefit of lower carbon emissions. In addition, hydrogen could be extracted from the mixture for use in hydrogen powered engines or for hydrogen fuel cell applications. Using the existing pipeline network to convey hydrogen in this way, would enable hydrogen production and hydrogen fuelled applications to become established prior to the development of a dedicated hydrogen transportation system, which would require considerable capital investment and time for construction.

Part of the NATURALHY Safety Work Package was to perform explosion experiments on a large scale, paying particular attention to the potential for transition from deflagration to detonation. In order to assist in focusing the large-scale experiments on the hydrogen/methane/air mixtures of most

interest, preliminary field-scale experiments were performed to measure the explosion overpressures generated by methane-hydrogen-air mixtures in well-understood repeated pipe congestion.

This paper describes the experiments to measure the overpressures from ignition of methane, hydrogen and air in a 3 m by 3 m by 2 m high rig containing nine layers of vertical grids in the bottom half and seven layers of horizontal grids in the top half. The objective of the work was to perform experiments with each of methane-air, hydrogen-air and 25/75, 50/50 and 75/25 mixtures (by volume) of hydrogen and methane with air at nominal equivalence ratios of 1.1 for mixtures containing methane and 1.2 for 100% hydrogen with air. The aim was to determine the amount of hydrogen that can be added to methane without giving a very large increase in the overpressures generated on ignition.

2.0 TEST FACILITY AND SET-UP

2.1 Test facility

The test facility was situated at the Dalehead site at the Health and Safety Laboratory at Buxton. The test facility comprised a:

- Purpose-built concrete pad, measuring some 10 m x 10 m inset in a 24 m by 18 m tarmac pad;
- A confining wall, left in position from previous trials [1], to prevent exposure of the main laboratory to high overpressures;
- Two parallel 0.2 m wide x 0.2 m deep ducts spaced 1.0 m apart (capped with galvanised sheeting), one for fuel / air delivery and the other for instrumentation cables;
- Additional 0.1 m wide x 0.2 m duct running at 45° to the other duct for another line of instrumentation;
- Remote-controlled fuel, air and purge gas delivery system;
- Local (30 m from the firing pad) instrument cabin containing the signal conditioning units and data logging system; and
- Remote control room (300 m from the firing pad) with video displays of the trials area and the secure, radio frequency, control board.

The previous trials [1] indicated that the confining wall does not influence the free field overpressures.

2.2 Congestion rig

The congestion rig comprises a 3 m (width) x 3 m (depth) x 2 m (height), metal framework, structured to consist of eighteen 1 m³ cubic units. The framework is capable of holding a range of metal grids. For the nominal 20 % area congestion, each grid comprises a number of 26 ± 1 mm diameter (nominal 1”) bars spaced 125 mm apart. The grids are inserted vertically into the lower layer of cells and horizontally into the upper layer of cells. In the lower layer, there are nine different lengths of grids. The grids are arranged within the rig to form concentric squares around the centre cube. For the experiments described in this paper, seven concentric squares of grids were used around the central 1 m cube, which had an additional two concentric squares of grids. The bars of each grid were in line with the bars on the other grids and the grids were spaced 0.15 m apart. The first vertical grid was 0.27 m from the ignition point and the last grid at 1.43 m. In the top layer of cells, the grids are placed horizontally and are all of the same dimensions. Each grid runs the full length of the frame (3 m) and is one cell wide (1 m). Hence, three grids are required to fill one complete layer within the upper cells. Seven layers of grids, with alternating layers running North-South and East-West (the bars of the lowest layer) were used for the experiments described in this report. The first horizontal grid was 0.57 m from the ignition point and the last grid 1.43 m. The volume of the steelwork in the rig was 0.793 m³ and hence the available gas volume was 17.207 m³. The volume blockage was 4.40%.

The outside of the metal frame and grid arrangements was covered with a thin (23 µm) plastic film, similar to cling-film. The purpose of the film was to produce a near-airtight cover to the rig to enable it to be filled with a flammable fuel-air mixture. The sides of the rig were wrapped using a single,

continuous length of film, wound in a spiral pattern around the edge of the frame. The roof was then covered with strips of film; the edges of the roof were over-lapped onto the sides to produce a gas-tight seal. The film was sticky, allowing one sheet to adhere to another. By over-lapping the film, the sticky nature was sufficient to seal the joins. The film was held to the frame using spray tack adhesive. The gap between the bottom of the frame and the concrete pad was filled with expanding polyurethane foam to minimise gas leakage. The congestion rig with the plastic film in position is illustrated in Figure 1.



Figure 1. Congestion rig with plastic film in position

2.3 Gas supply

The fuel gases for filling the rig and carbon dioxide for purging the cable ducts were supplied from standard cylinders. The gases were piped from the cylinders to the test facility via a pressure regulator and a pneumatically controlled flow valve. The valve actuators were controlled remotely using a radio frequency control system. The fuel supply line was split into four outlets positioned at the four vertical edges of the lower, innermost cube of the congestion rig. Each outlet was positioned ~ 150 mm above floor level and terminated with an air amplifier to entrain air and thereby aid mixing of the fuel and air in the rig. The air amplifiers were orientated such that the fuel-air mixture ejecting from each device was pointed towards the centre of the rig. Since, with this system, mixing only occurs while the fuel is being added; additional mixing was achieved by use of an additional supply of compressed air fed to the rig through a large air amplifier. Addition of fuel or air to the rig was controlled remotely from a control room ~ 300 m from the test facility. Whilst the addition of more air to the rig decreases the concentration of fuel slightly, the action of the air amplifier entraining the air in the rig adds to the mixing process. The decrease in the concentration of fuel is slow.

The gases used in the trials were purchased pre-mixed from BOC Ltd. For the mixtures of hydrogen and methane, the supplier indicates that the cylinders are volume filled to $\pm 5\%$ of the desired concentrations and the hydrogen or methane concentration then measured. The relative error of the percentage quoted on the test certificate was given as $\pm 5\%$ of the hydrogen concentration. Hence for the nominal 75% methane 25% hydrogen mixture, the quoted hydrogen concentration was 25.5% with an absolute error of $\pm 1.3\%$. The concentrations of the gases used are summarised in Table 1.

Table 1. Concentrations of gases used

Nominal gas mixture	Concentration of measured component
100% Methane	99.5 % Methane
75% Methane 25% Hydrogen	25.5% Hydrogen
50% Methane 50% Hydrogen	50.9% Hydrogen
25% Methane 75% Hydrogen	24.96% Methane
100% Hydrogen	99.995% Hydrogen

2.4 Ignition systems

The fuel/air mixture in the congestion rig was ignited using an ignition source located at a height of 500 mm and positioned in the centre of the lower, central cube. For hydrogen-air mixtures, an induction coil spark unit, activated using the remote control system, provided ignition. The ignition spark was detected and logged using a pick-up coil mounted on the HT cable supply to the spark plug. Typically, the type of spark used would have ignition energy of ca. 50 mJ. A larger, more energetic spark was used to ensure reliable ignition of the mixtures with methane. A high voltage power supply was used to charge a 20 nF capacitor to 15kV via a resistor. At the moment of firing, the power supply is first disconnected from the capacitor by means of a pneumatically actuated contactor. A fraction of a second later, a further pneumatic contactor connected the capacitor across a 6 mm spark gap located in the congestion rig; the energy stored in the capacitor is discharged in the form of a spark at the gap. The energy stored in the capacitor is 2.25 J although, due to cabling capacitance and losses at the contactor, not all this energy is available at the spark.

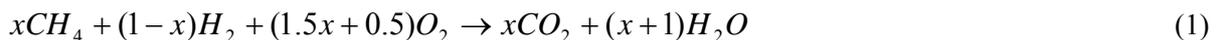
3.0 STOICHIOMETRIC RATIO AND MASSES OF FUEL GASES

3.1 Concentration, temperature and humidity sensors

In order to derive the stoichiometric ratio at the time of ignition, it was necessary to measure the fuel concentration, temperature and relative humidity. The concentration of fuel gas in the congestion rig was derived from measurements of the oxygen concentration within the rig. It was assumed that any decrease in the concentration of oxygen was caused by displacement of oxygen by fuel gas. The concentration of oxygen was measured using seven “AO2 International Technologies Automotive Oxygen” sensors. The sensors were distributed to give an indication of the degree of mixing within the rig. The sample standard deviation of the mean oxygen concentration was ≤ 0.35 %. The change in humidity is largely due to the injection of gas, whilst the change in temperature is due to the plastic-covered rig acting like a green house. The temperature was measured using a 3 mm stainless steel sheathed RTD mounted within the rig and the humidity was measured using a new (for each trial), calibrated (by the manufacturer) Honeywell HIH 3610 series sensor. The data was recorded to a computer at a frequency of 0.5 Hz.

3.2 Derivation of stoichiometric ratio

The equation for the complete combustion of a methane and hydrogen is as follows:



Where x is the certified % methane/100 (if the methane in the mixture is analysed) or x is $1 -$ the certified % hydrogen/100 (if the hydrogen in the mixture is analysed).

Partial pressures are equivalent to mole fractions for all real gases. Hence, the total pressure is given by:

$$1 = P_{\text{oxygen}} + P_{\text{nitrogen etc}} + P_{\text{water vapour}} + P_{\text{methane}} + P_{\text{hydrogen}} \quad (2)$$

The partial pressure ($p_{\text{nitrogen etc}}$) of nitrogen, argon and other inert atmospheric gases is derived from the ratio of oxygen to inert gases in dry air i.e. $p_{\text{nitrogen etc}} = 3.773 p_{\text{oxygen}}$. The partial pressure of water vapour ($p_{\text{water vapour}}$) is calculated (using a third order expression derived from 0 °C to 50 °C data [2]) from the saturated vapour pressure for the temperature (T , °C) measured inside the congestion rig and the measured relative humidity ($RH\%$). To simplify the calculations, the saturated vapour pressure of water in the air and fuel gas mixture is assumed to be the same as that for air alone.

$$P_{\text{watervapour}} = (0.0000007T^3 - 0.000002T^2 + 0.0006T + 0.006)RH\%/100 \quad (3)$$

For the hydrogen-air trial, the oxygen partial pressure (p_{oxygen}) is taken from the measured (oxygen depletion) percentage concentration divided by 100. For the methane-air trial the partial pressure of methane was calculated from the depleted oxygen as for hydrogen (direct methane concentration measurements were also made but as there was very good agreement between methods, details are not given here). In the trials with mixtures of methane and hydrogen, the methane and hydrogen mixtures were purchased from BOC as made up volumetric mixtures. It was assumed that these mixtures did not separate out in the cylinder during storage and the oxygen depletion measurements again used to derive the stoichiometric ratio. From equation 1, the fuel/oxygen ratio for complete combustion is given by $1/(1.5x+0.5)$ and hence the equivalence ratio for each mixture is given by:

$$S = (1.5x + 0.5)p_{\text{fuel}} / p_{\text{oxygen}} \quad (4)$$

3.3 Masses of the fuels present

The mass (M_g , kg) of each fuel gas in the rig was calculated as follows:

$$M_g = p_{\text{gas}} \frac{V_{\text{free}}}{0.022414} m_{\text{wt}} \frac{P_{\text{atmospheric}}}{101.325} \frac{273.15}{(273.15 + T)} \quad (5)$$

Where: p_{gas} is partial pressure of methane xp_{fuel} or hydrogen $(1-x)p_{\text{fuel}}$, V_{free} is free volume inside the rig (17.207 m³), m_{wt} is the molecular weight of gas (methane 0.016043 kg, hydrogen 0.002016 kg), $P_{\text{atmospheric}}$ is the atmospheric pressure (kPa) and T is the gas temperature (°C).

4.0 OVERPRESSURE MEASUREMENT

Two types of overpressure sensors were deployed. Brüel & Kjær 8103 hydrophones were used to record ‘lower’ overpressures (up to 10 bar) and Kulite ETL-345F-375M Series 40 bara piezo-resistive transducers were used to measure ‘higher’ overpressures. The hydrophones have a nominally omnidirectional response in the radial XY plane (defined as a plane at 90° to the hydrophone axis) when unshielded and were orientated vertically such that they were facing upwards with the radial plane parallel to the ground. All the piezo-resistive sensors were mounted in specially made streamlined blocks. They were factory fitted with shields to protect the sensors against heat and flash light. The Kulite sensors had a plane response at their diaphragm and these were orientated vertically such that the diaphragm was facing upwards and parallel to the ground. As the Kulite gauges are being used at the lower end of their measurement range, evidence of a post event baseline shift is apparent in some of the overpressure records. This error (0.1% of full scale, 4 kPa) is a combination of hysteresis error and zero repeatability error caused by mechanical and electrical effects at the transducer diaphragm. Since the transducer signals are dc corrected by using the pre-trigger information, the maximum overpressure measurements are not affected by this error.

All the Kulite sensors were positioned at a height of 500 mm above the ground and were located as shown in Figure 2. Because of the topology, the hydrophone 15 (at 16 m) had to be mounted 1.2 m above the pad and hydrophone 16 (at 32 m) 4.4 m above the pad. The blocks containing the Kulite sensors were fixed into a short length of scaffolding, which was bolted into a standard floor fitting fixed to the ground.

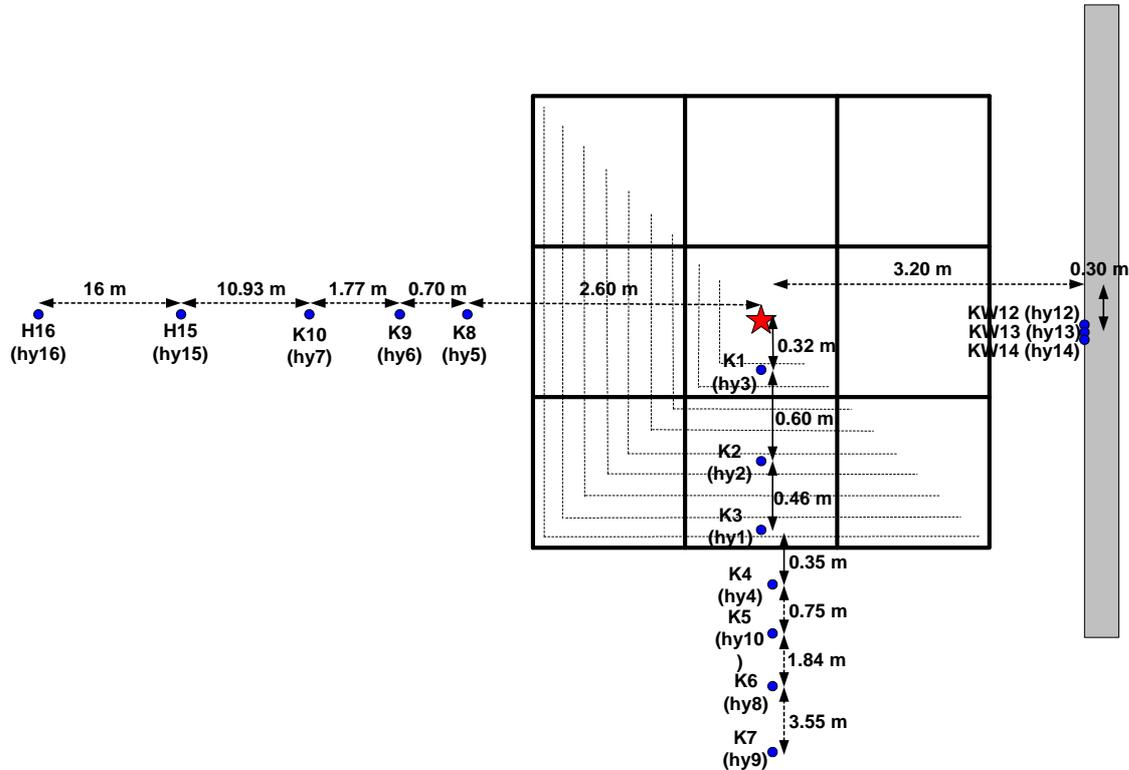


Figure 2. Pressure sensor positions

Overpressure measurements were logged to a computer and the data collected in burst mode at a frequency of 50 kHz. The data was processed on saving using the FAMOS software package.

5.0 VISUAL RECORDS AND METEOROLOGICAL MEASUREMENTS

Video cameras were used to monitor and record the trials. A Panasonic, F10 camera was located 20 m to the east of the congestion rig together with a Sony DV cam. A further Sony video camera was positioned about 150 m from the test rig. The images from the cameras were fed to video recorders and monitors in the control room. A high-speed cine camera (500 frames per second) was also deployed at a distance of 20 m to the east of the congestion rig.

The air temperature, relative humidity, wind speed and direction were measured at the instrument cabin 30 m from the pad using a Vector Instruments weather station. This comprised wind speed, wind direction, temperature and humidity sensors mounted 3.5 m above the ground. The instruments were connected to the data-logging equipment, allowing recording of the weather conditions to be made during the trials. As only the conditions (measured separately) inside the congestion rig were relevant to interpretation of the data, the data obtained is only indicative i.e. the instruments were not specifically calibrated for these trials. The barometric pressure was recorded using a Sensor Technics barometric pressure transducer located in the instrument cabin and connected to the data logger.

6.0 WORK PLAN AND OPERATING PROCEDURE

It was planned to perform the trials with 100%, 0% and 50% hydrogen initially and to then perform two further experiments at the concentrations of most interest. In the event, these were performed with 25% and 75% hydrogen. The experiment with hydrogen-air used a nominal stoichiometric ratio of 1.2 as this gave the highest overpressures in previous trials. In the trials with methane present, a nominal stoichiometric ratio of 1.1 was used as this gives the highest overpressures with hydrocarbons [3]. The stoichiometric ratio could only be controlled to ± 0.1 .

The following operating procedure was used.

- (a) 9 layers of vertical and 7 layers of horizontal grids were inserted into the congestion rig and the rig secured to the concrete pad using the bolt-down fixings.
- (b) All the sensors, the ignition system and the remote control valves were checked for correct operation.
- (c) The plastic sheeting was applied to the top and sides of the rig, held in place by spray tack applied to the uprights of the frame.
- (d) The fuel gas was used to charge the congestion rig to an initial concentration of flammable gas. An iterative process (involving monitoring of the gas temperature, humidity and concentration, calculating the stoichiometry and adding further fuel gas or air) was used until the required stoichiometry was achieved.
- (e) The instrument and gas supply ducts were flushed with carbon dioxide to provide a non-flammable atmosphere inside the ducts.
- (f) After making sure the exclusion zone is clear, the ignition system was activated.
- (g) The data from the explosion was recorded, backed up and stored.

7.0 CONDITIONS ON IGNITION AND OBSERVATIONS

The initial conditions for each trial are summarised in Table 2.

Table 2. Trial conditions

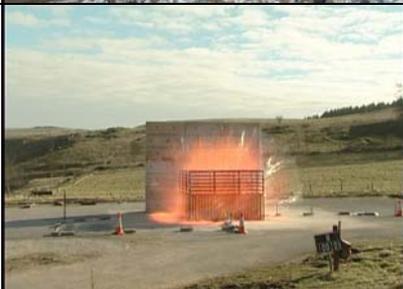
Measurement	NatHy_ 01	NatHy_ 02	NatHy_ 03	NatHy_ 04	NatHy_ 05
Methane (vol. %) in fuel gas mixture	0.0	100.0	49.1	74.5	25.0
Hydrogen (vol. %) in fuel gas mixture	100.0	0.0	50.9	25.5	75.0
Number of layers	9	9	9	9	9
Free volume (m ³)	17.207	17.207	17.207	17.207	17.207
Gas mixture temperature (°C)	11.0	4.8	14.5	14.6	18.3
Relative humidity (%)	30.7	85.1	42.5	58.2	35.4
Atmospheric pressure (kPa)	97.72	97.71	97.33	94.67	96.69
Mean oxygen concentration (%)	13.59	18.71	17.63	18.18	16.54
Calculated methane concentration (%)	0.00	9.94	7.45	9.14	5.08
Partial oxygen pressure	0.1359	0.1871	0.1763	0.1818	0.1654
Partial nitrogen etc pressure	0.5127	0.7059	0.6651	0.6859	0.6241
Partial water vapour pressure	0.0041	0.0076	0.0070	0.0096	0.0073
Partial fuel gas pressure	0.3474	0.0994	0.1517	0.1227	0.2033
Partial methane pressure	0.0000	0.0994	0.0745	0.0914	0.0508
Partial hydrogen pressure	0.3474	0.0000	0.0772	0.0313	0.1524
Ratio of methane + hydrogen to oxygen	2.556	0.531	0.861	0.675	1.229

Measurement	NatHy_01	NatHy_02	NatHy_03	NatHy_04	NatHy_05
Stoichiometric fuel/oxygen ratio 1/(1.5x+0.5)	2.000	0.500	0.809	0.618	1.143
Equivalence ratio of mixture on ignition	1.28	1.06	1.06	1.09	1.08
Mass of hydrogen (kg)	0.498	0.000	0.109	0.043	0.211
Mass of methane (kg)	0.000	1.160	0.837	0.998	0.560

Note that in these trials the humidity of the flammable cloud was uncontrolled and varied. It is known that this will have a minor affect the resultant explosion overpressures but an insignificant effect on the observed trends.

The images immediately after ignition, an approximation (by counting 40 ms frames) to the fireball duration and the condition of the plastics wrapping are summarised in Table 3.

Table 3. Summary of observations

Trial No. and amount of hydrogen present in fuel gas	Frame immediately after ignition	Approximate fireball duration and condition of wrapping
NatHy_02 0% hydrogen		720 ms Very large pieces
NatHy_04 25% hydrogen		480 ms Fairly large pieces
NatHy_03 51% hydrogen		480 ms Fairly large pieces

Trial No. and amount of hydrogen present in fuel gas	Frame immediately after ignition	Approximate fireball duration and condition of wrapping
NatHy_05 75% hydrogen		360 ms Fairly large pieces
NatHy_01 100% hydrogen		320 ms Finely shredded

8.0 RESULTS

The maximum overpressures generated inside and near the rig (parallel to wall data) for the different concentrations of methane and hydrogen are compared in Figure 3.

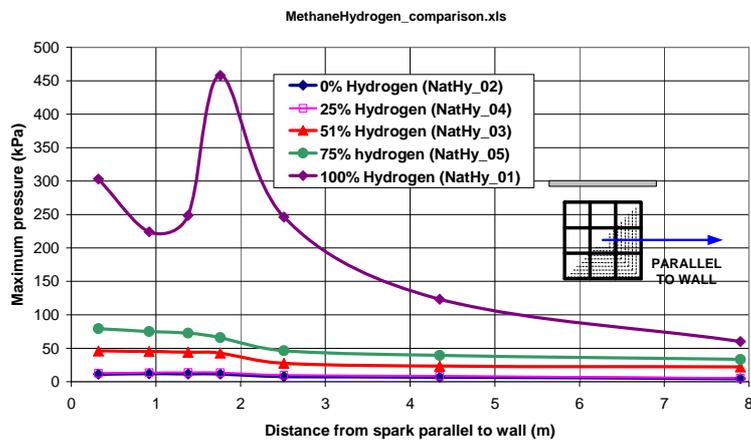


Figure 3. Maximum overpressures inside and near rig for different concentrations

Inside the rig, there was very little increase in overpressure for mixtures up to 25% hydrogen, some increase with the 51% and 75% hydrogen mixtures and a large increase with 100% hydrogen. A comparison of the further a field data (away from wall) is illustrated in Figure 4.

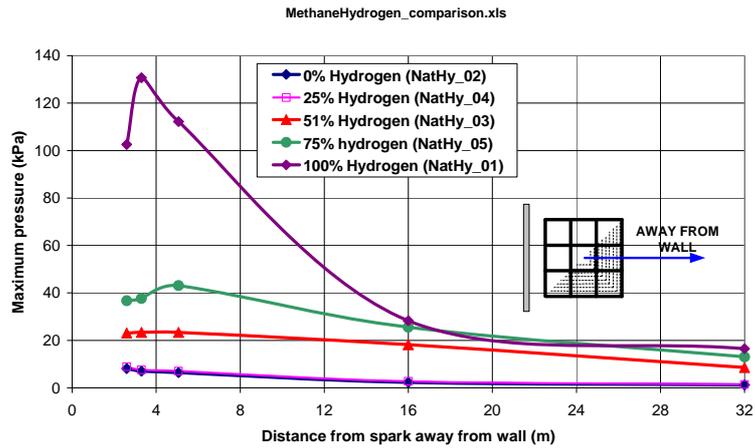


Figure 4. Maximum overpressures further a field for different concentrations

0% hydrogen and 25% hydrogen gave very similar maximum overpressures, 51% hydrogen gave maximum overpressures about four times higher and 75% hydrogen about 8 times higher and 100% at least 20 times higher than methane alone.

A plot of the overpressure traces at 16 m is given in Figure 5.

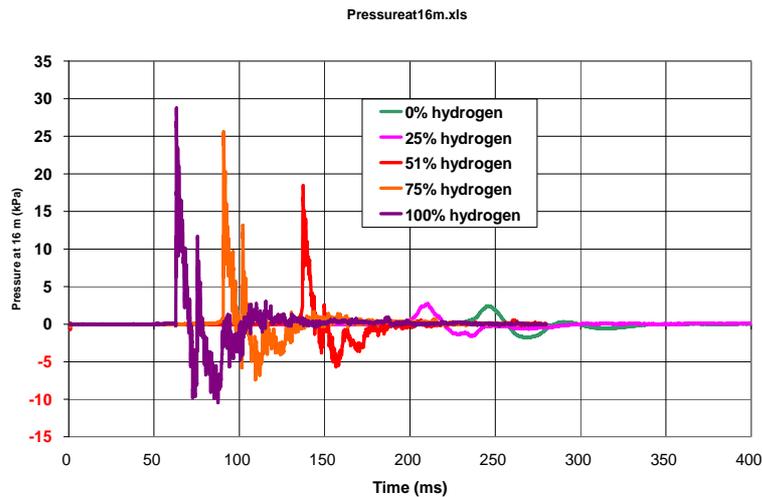


Figure 5. Overpressures traces at 16 m

The arrival time at 16 m is earlier the higher the hydrogen concentration. The maximum overpressures 1.5 m up the wall (KW12), inside the rig, just outside the rig (K4) and at 32 m (H16) are summarised in Table 3 for each hydrogen concentration.

Table 3. Summary of maximum overpressures

Hydrogen concentration (%)	Maximum overpressure on wall (kPa)	Maximum overpressure just outside rig (kPa)	Maximum overpressure inside the rig (kPa)	Maximum overpressure at 32 m (kPa)
0	14.8	11.4	11.8	1.2
25.5	19.3	13.7	13.7	1.4
50.9	98.0	42.8	44.0	8.6
75.0	171.3	66.1	79.3	13.0
100	614.5	457.7	303.2	16.5

The best representation of the data was a second order polynomial fit against the mass of hydrogen. The maximum overpressures from each trial are plotted against the mass of hydrogen in the gas mixture in Figure 6 with the Excel trend line fit.

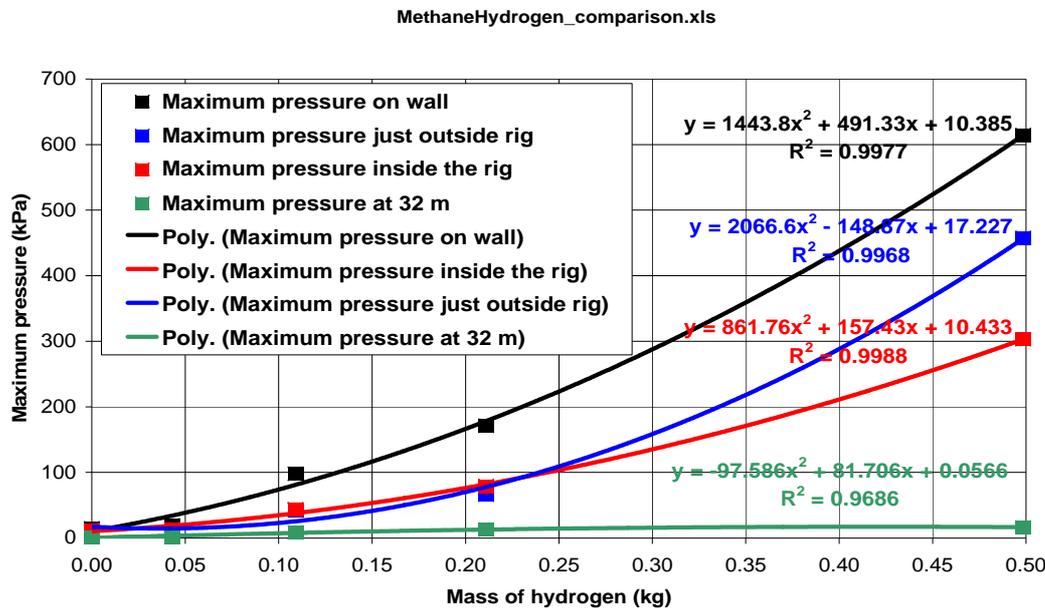


Figure 6. Maximum overpressures versus mass of hydrogen

The results suggest the explosion effects from the mixtures are reasonably correlated with the mass of hydrogen in the mixture if the same congestion and volume is used. On the basis of the trials performed, the maximum overpressures generated by methane/hydrogen mixtures with 25% (by volume) or less hydrogen content are not likely to be much more than those generated by methane alone.

In the trial with 100% hydrogen, the plastic wrap was shredded into very small strips (see Figure 7). 24 pieces were selected at random and their mean width determined to be 20 ± 7 mm. Groethe et al. [4], in trials with volume blockage 9.1% (cf. 4.4%), indicates that one characteristic of detonation is the shredding of the plastic wrapping into similar very small strips; the width of the strips being equal to the detonation cell width. Although no diagonal overpressure measurements were made in these trials, it is known from previous trials that the extra congestion moving towards the corners generates higher overpressures. The shredding in the 100% hydrogen trial and the very bright flash (see Table 3), indicates that as the flame propagated towards the corners of the rig, transition from deflagration to

detonation occurred, even though the sharp pressure spike characteristic of detonation was not recorded.



Figure 7. Shredding wrapping from hydrogen-air trial

9.0 CONCLUSIONS

The main conclusions are as follows:

- (a) For 100% hydrogen, the shredding of the plastic film into very narrow strips indicates that transition to detonation occurred at the corners of the rig.
- (b) The overall maximum overpressures close to the edge of the rig were 0.12, 0.14, 0.44, 0.73 and 4.57 bar for hydrogen concentrations of 0, 25.5, 50.9, 75 and 100%, respectively. Further a field (32 m from ignition), the corresponding overpressures were 12, 14, 86, 130 and 165 mbar.
- (c) The results suggest the explosion effects from the mixtures are reasonably correlated with the mass of hydrogen in the mixture if the same congestion and volume is used.
- (d) On the basis of the trials performed, the maximum overpressures generated in large scale trials by methane/hydrogen mixtures with 25% (by volume) or less hydrogen content may not be much more than those generated by methane alone.

10.0 ACKNOWLEDGEMENTS

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