IGNITION ENERGY AND IGNITION PROBABILITY OF METHANE-HYDROGEN –AIR MIXTURES

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ABSTRACT

The European Commission are funding an investigation of the feasibility of using existing natural gas infrastructures to transport and distribute hydrogen, as a mixture of natural gas and hydrogen, from the point of hydrogen production to the point of use. Since hydrogen has different chemical and physical properties to that of natural gas, and these will affect the integrity and durability of the pipeline network and the ignition and combustion behavior of released gas, it is necessary to assess the change in risk to the public that would result. The subject of this paper is an experimental study of the effect of the hydrogen content of the natural gas-hydrogen mixture on the minimum energy required for ignition, and the probability of achieving ignition given a particular level of energy discharge. It was possible to normalize the results for ignition energy such that, given information on the minimum ignition energy and the equivalence ratio at which the minimum ignition energy occurs, the lowest ignition energy for any other equivalence ratio can be predicted. The results also showed that the ignition process has a probabilistic element and that the probability of ignition is related to the equivalence ratio and the energy level of the source. It was observed that the probability of ignition increased with increasing energy of the source and that the rate of rise in probability was steepest for the equivalence ratios close to the equivalence ratio at which the minimum ignition energy occurs.

1.0 BACKGROUND

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 require 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, or 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.

The existing gas pipeline networks are designed, constructed and operated based on the premise that natural gas is the material to be conveyed. If this infrastructure is to be used to transport a different gas, such as a natural gas-hydrogen mixture, then the overall safety of the network and the risks presented to the public must be reassessed. Hydrogen has different chemical and physical properties and these will affect the integrity and durability of the pipeline network and the ignition and combustion behavior of released gas. The effect of hydrogen content of the released gas on the minimum energy required for ignition of a gas-air mixture, and the probability of achieving ignition given a particular energy discharge is the subject of this paper.

2.0 SCOPE OF THE WORK

The work described in this paper has been undertaken to provide information on the lowest levels of energy required to ignite methane-hydrogen-air mixtures and to identify how these energy levels vary as the fuel (methane-hydrogen mixture) varies and as the fuel-air ratio varies. Analysis of the results obtained also provides information, over the whole range of flammable methane-hydrogen-air mixtures, on the likelihood (probability) of a particular level of energy resulting in ignition.

3.0 EXPERIMENTAL ARRANGEMENT

3.1 The Test Facility

The work on the ignition energy and the likelihood of ignition of methane-hydrogen-air mixtures was carried out at Loughborough University using a purpose built test facility. The approach adopted followed the method initiated by Lewis and von Elbe (1987) [1]. The circuit used for generating and measuring the energy was based on the standards BS EN 13821: 2002 [2] and ASTM E582 1988 [3].

A diagram of the test facility is shown in Figure 1. The test facility comprised: a cylindrical combustion chamber; a gas supply, delivery and evacuation system; an ignition system; and a data acquisition system. The main components are described below.

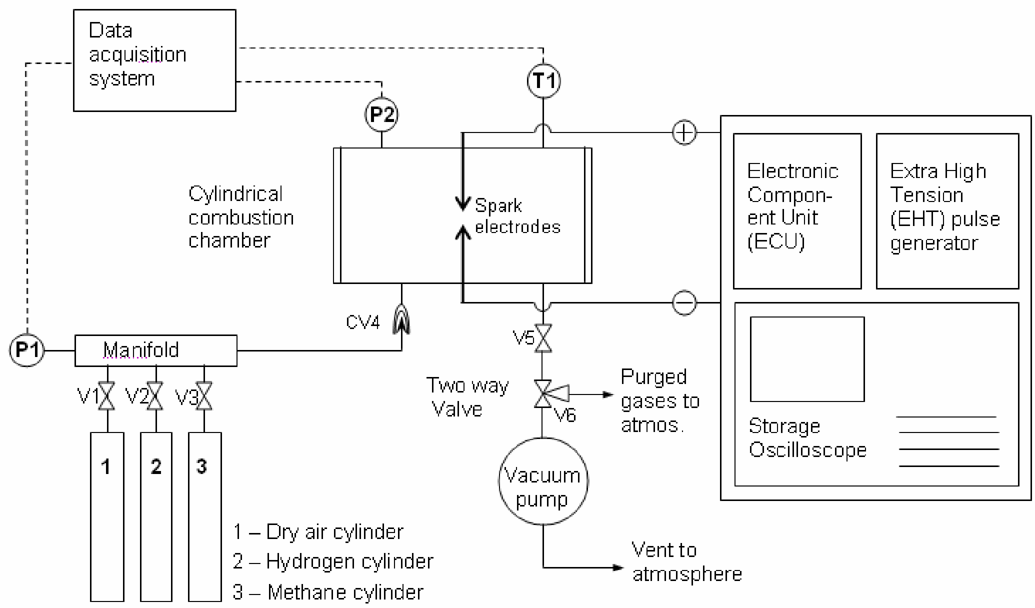

Figure 1: Diagram of Test Facility

3.2 Cylindrical Combustion Chamber

The combustion chamber was constructed from a 152 mm diameter, 232 mm long stainless steel pipe. Flanges were welded at each end to attach polycarbonate windows so that ignition of the fuel-air mixture formed inside the chamber could be observed. Access points were provided in the cylindrical wall of the chamber to allow gases to enter the chamber to form the fuel-air mixture and to extract the combustion products by means of a vacuum pump. A further two ports were used to enable the pressure, P2, and temperature, T1 (see Figure 1) to be measured during the combustion process to confirm that ignition of the fuel-air mixture had or had not been achieved. The final two access points were used for the two electrodes forming the spark gap in the centre of the chamber. A diagram of the combustion chamber is shown in Figure 2.

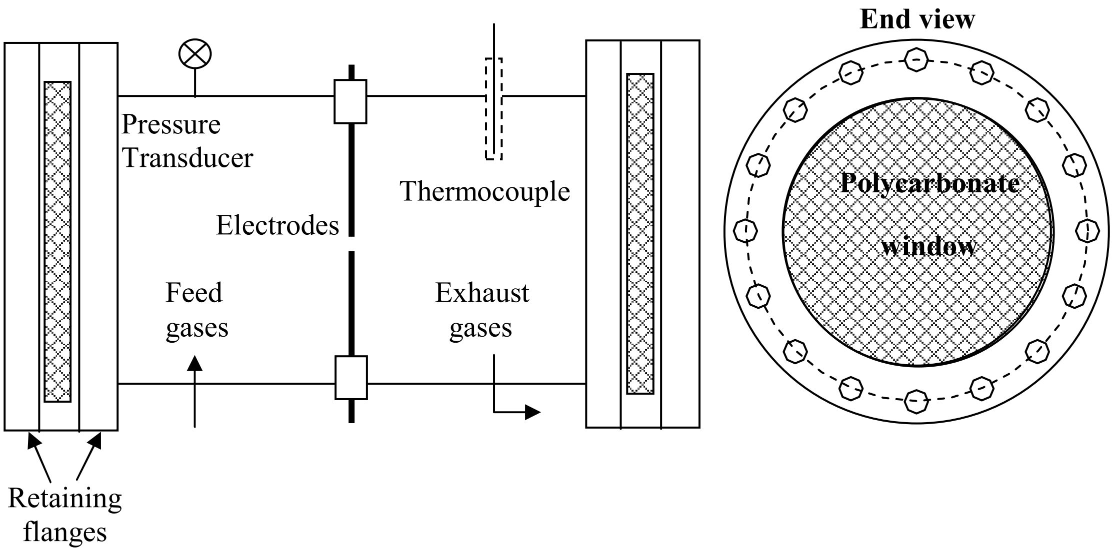

Figure 2: Diagram of Cylindrical Combustion Vessel

3.3 Gas Supply, Delivery and Evacuation System

The gases used to form the fuel-air mixtures in the experiments were methane, hydrogen and dry air supplied in gas cylinders at an initial gauge pressure of 200 bar. Each gas cylinders was connected to a manifold which in turn was connected by a filler pipe to the inlet port of the combustion chamber. A pressure transducer, P1 (see Figure 1) located on the manifold enabled the gradual build up of pressure inside the combustion chamber to be monitored. The first step in the formation of a fuel-air mixture was to evacuate the chamber. Then the fuel gases were admitted, one at a time, followed by the dry air. The desired mixture was obtained by the method of partial pressures with the total absolute pressure set to 1 bar. The dry air was admitted to the chamber with sufficient energy to ensure thorough mixing of the gases. However, prior to ignition, a short time was allowed to elapse to enable the temperature to stabilise at the ambient temperature within the laboratory and any motion to decay. This ensured that a quiescent uniform stable mixture was achieved. Once the fuel-air mixture had been formed within the chamber, and prior to attempting ignition, the delivery manifold was isolated from the chamber by removal of the filler pipe.

3.4 Ignition System

The electrical circuit used to generate a capacitive spark at the gap between the electrodes located at the centre of the combustion chamber and the equipment used to determine the energy discharged from the capacitor is shown in Figure 3. Characterising the spark energy as the stored energy in the capacitor followed the practice initiated by Lewis and von Elbe [1]. The circuit used for generating and measuring the energy was based on the standards BS EN 13821 : 2002 [2] and ASTM E582 1988 [3].

The electrodes were manufactured from 2 mm diameter copper rod. These were housed in 10 mm diameter Teflon sleeves extending to about 10 mm from the end of the spark gap end of the electrode. These Teflon sleeves provided structural support and insulated the electrodes from the combustion chamber. The gap between the electrodes across which the spark formed was set to 2 mm. At regular intervals during the experimental programme it was necessary to clean the tips of the electrodes to remove combustion deposits.

The energy level of the spark was varied by selecting different combinations of charging resistor and discharge capacitor. The combinations of resistor and capacitor used and the corresponding discharge energy are shown in Table 1.

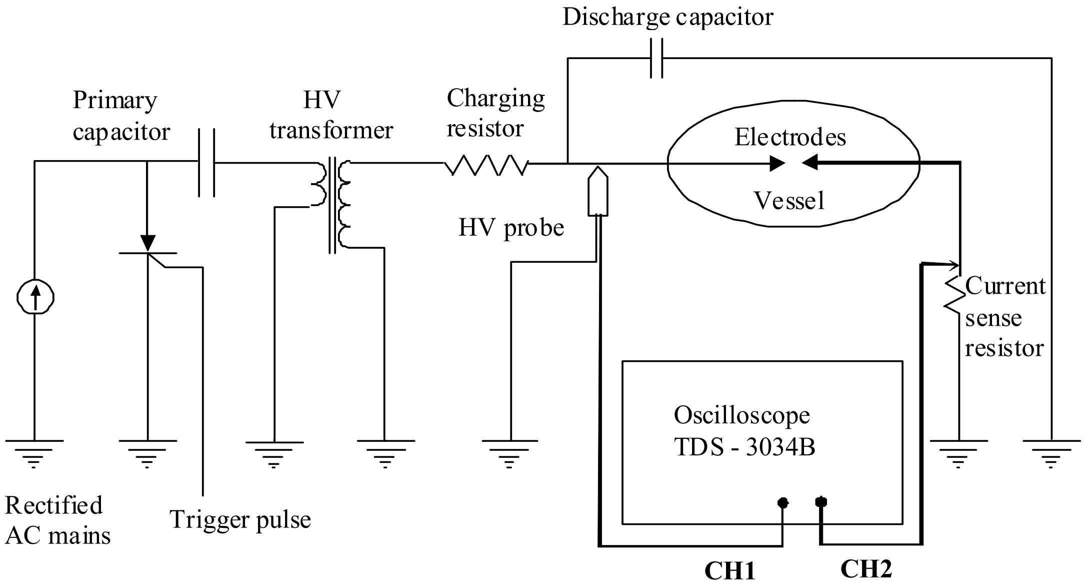

Figure 3: Electrical Circuit and Measurement Equipment

Discharge capacitor (pF)	Charging Resistor (kΩ)	Charging time (μs)	Nominal Breakdown Voltage(kV)	Nominal Energy (mJ)
10	1000	10	6.0	0.2
50	100	5	6.0	0.9
100	100	10	6.0	1.8
150	100	15	6.0	2.7
200	100	20	6.0	3.6
300	100	30	6.0	5.4

Table 1: Nominal Energy for Different Combinations of Discharge Capacitor and Charging Resistor

For each experiment, after filling the test rig as per Section 3.3, the appropriate combination of charging resistor and discharging capacitor were installed and the ignition system armed. The ignition system was then fired. On most occasions visual observation was sufficient to determine whether or not the fuel air mixture had been ignited. If there was doubt then ignition was confirmed, or otherwise by inspection of the pressure record obtained from pressure transducer P2.

If ignition was not achieved, then the spark energy was increased by selecting a different charging resistor, discharging capacitor combination.

This process was repeated a maximum of four times. If after four attempts ignition had not been achieved then the chamber was evacuated and a new experiment was started.

3.5 Data Acquisition System

The experiments were conducted using a semi-automatic procedure. The pressures measured by the pressure transducer, P1 located on the manifold (see Figure 1) indicating the partial pressures of the components of the mixture formed within the combustion chamber were recorded using a PC. The variation in pressure and temperature with time sensed by the transducers P2 and T1 (see Figure 1) following initiation of the spark were also recorded. The values of charging resistance (R) and discharging capacitance (*C*) were input manually to the PC as was the voltage (*V*) required to generate the spark across the gap in the electrodes as recorded on the oscilloscope. A marker was also recorded manually on the PC indicating whether or not the fuel-air mixture had ignited.

The records of pressure measured by transducer P1 were used to calculate the composition of the fuel and the fuel-air mixture. The voltage and capacitance were used to calculate the ignition energy.

3.6 Experimental Programme

An experimental programme to determine the minimum energy capable of achieving the ignition of a particular fuel (methane-hydrogen)-air mixture, and the probability of a particular level of energy achieving ignition was undertaken, covering as wide a range of flammable methane-hydrogen-air mixtures as could be achieved using the equipment available.

The target test conditions for the five test series carried out (Series A to E) are shown in Table 2. The information presented in Table 2 consists of the target fuel compositions (methane-hydrogen mixtures) studied, the estimated upper and lower flammability limits (UFL and LFL, respectively) for each fuel composition, and the target concentrations of fuel in the fuel-air mixtures.

Test Series	$CH_4 - H_2$ Mixture (vol. %)	LFL (vol. %)	UFL (vol. %)	Target concentration of fuel in fuel-air mixture (vol.%)					
A	100 - 0	5.0	15.0	7	8	9	11		
B	75 - 25	4.7	21.1	8	12	16	20		
C	50 - 50	4.4	27.7	6	10	16	20	26	
D	25 - 75	4.2	40.5	8	14	21	28	38	
E	0 - 100	4.0	75.0	6	10	20	30	45	60

Table 2: Target Test Conditions for the Five Test Series A to E

4.0 RESULTS AND DATA PROCESSING

Approximately 2000 ignitions were attempted during the five test series shown in Table 2. For each attempted ignition the data recorded on the PC (see Section 3.5) was used to derive the following information:

- the actual methane-hydrogen mixture;
- the actual fuel-air mixture;
- the actual energy from the relationship: $E = \frac{C V^2}{2}$ (1)
- whether or not the fuel-air mixture was ignited.

4.1 Results

The detailed results obtained during Test Series B (75 vol. % methane – 25 vol. % hydrogen) are presented as an example of the results obtained during all the test series.

During Test Series B, experiments were performed using fuel-air mixtures over the target fuel-air mixture range 8 to 20 vol % as indicated in Table 2

For each of the target fuel-air mixtures, 8, 12, 16 and 20 vol.% fuel in the fuel-air mixture, twenty different fuel-air mixtures were prepared and ignition attempted up to four times for each.

Figure 4 shows each experiment plotted on an Energy-Equivalence Ratio diagram with the ignitions and non-ignitions identified. The overlap of ignitions and non-ignitions shows that that there is a probabilistic element to ignition, whereby ignition is related to both the equivalence ratio and the energy level of the ignition source.

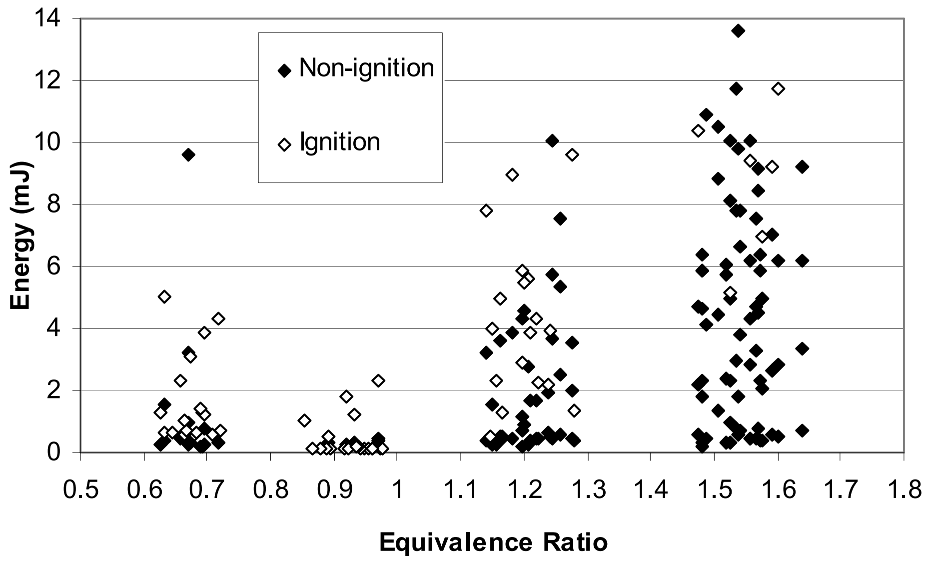

Figure 4: Results of Ignition Experiments for Test Series B (75 vol. % methane – 25 vol. % hydrogen)

Equivalence Ratio (*ER*) is the actual fuel-air ratio of a mixture, on a volume basis, compared with the stoichiometric fuel-air ratio. The stoichiometric fuel-air ratio is the ratio of the volume of fuel to the volume of air of a mixture containing just sufficient oxygen for complete combustion to be realised without resulting in excess fuel or excess oxygen in the combustion products.

The Equivalence Ratio (*ER*) is given by:

$$ER = \frac{\left(\dfrac{Volume\ of\ \ flammable\ gas\ in\ the\ actual\ mixture}{Volume\ of\ \ air\ in\ the\ actual\ mixture}\right)}{\left(\dfrac{Volume\ of\ \ flammable\ gas\ in\ a\ stoichiometric\ mixture}{Volume\ of\ \ air\ in\ a\ stoichiometric\ mixture}\right)}$$

A lean mixture, which is one with excess oxygen, will have an equivalence ratio less than 1. A rich mixture, which is one with excess fuel, will have an equivalence ratio greater than 1.

The Lowest Ignition Energy (E_L) is the lowest energy at which ignition was achieved for each methane-hydrogen-air mixture at each mean equivalence ratio

The data displayed in Figure 4 is summarised in Table 3. Similar data obtained from Test Series A and C to E are also shown in Table 3.

Test Series A: 100 vol. % CH_4 – 0 vol. % H_2							
Mean fuel concentration (vol. %)	6.69	7.96	8.95	10.63			
Mean equivalence ratio (*ER*)	0.68	0.82	0.94	1.13			
Lowest Ignition energy (E_L) (mJ)	1.82	0.24	0.22	0.83			
Test Series B: 75 vol. % CH_4 – 25 vol. % H_2							
Mean fuel concentration (vol. %)	8.01	10.67	13.49	16.70			
Mean equivalence ratio (*ER*)	0.67	0.93	1.20	1.54			
Lowest Ignition energy (E_L) (mJ)	0.58	0.10	0.52	5.19			
Test Series C: 50 vol. % CH_4 – 50 vol. % H_2							
Mean fuel concentration (vol. %)	6.14	10.03	13.48	16.16	20.10		
Mean equivalence ratio (*ER*)	0.39	0.66	0.93	1.15	1.50		
Lowest Ignition energy (E_L) (mJ)	2.72	0.16	0.09	0.10	4.11		
Test Series D: 25 vol. % CH_4 – 75 vol. % H_2							
Mean fuel concentration (vol. %)	8.43	13.64	17.90	21.43	27.22		
Mean equivalence ratio (ER)	0.40	0.67	0.92	1.16	1.56		
Lowest Ignition energy (E_L) (mJ)	2.06	0.10	0.07	0.11	1.24		
Test Series E: 0 vol. % CH_4 – 100 vol. % H_2							
Mean fuel concentration (vol. %)	6.32	10.72	20.97	25.54	30.34	45.74	62.08
Mean equivalence ratio (*ER*)	0.16	0.29	0.63	0.82	1.04	2.01	3.91
Lowest Ignition energy (E_L) (mJ)	0.76	0.10	0.04	0.03	0.02	0.06	0.22

Table 3: Lowest Ignition Energy for each Mean Equivalence Ratio

Figure 5 shows the lowest ignition energy given in Table 3 plotted against the mean equivalence ratio prepared for each methane-hydrogen mixture.

Also plotted on Figure 5 are data presented by Lewis & von Elbe [1] and Gexcon [4] for methane, and Lewis & von Elbe [1] and D.W.V [5] for hydrogen.

The lowest ignition energies observed for methane-air mixtures and hydrogen-air mixtures agree reasonably well with the previously reported data.

4.2 Normalised Lowest Ignition Energies

As can be seen on Figure 5, all the plots of lowest ignition energy against equivalence ratio for the different methane-hydrogen mixtures studied display a minimum at an equivalence ratio close to 1 (stoichiometric). The minimum value of the lowest ignition energy is called the Minimum Ignition Energy (E_M) and the Equivalence Ratio at which E_M occurs is given the symbol (ER_M).

Further inspection of the results shown on Figure 5 indicated the possibility of collapsing the data onto a single curve by normalising the data. First, the Minimum Ignition Energies (E_M) and the Equivalence Ratios at which the Minimum Ignition Energies occurred (ER_M) were identified for each fuel mixture as shown in Table 4.

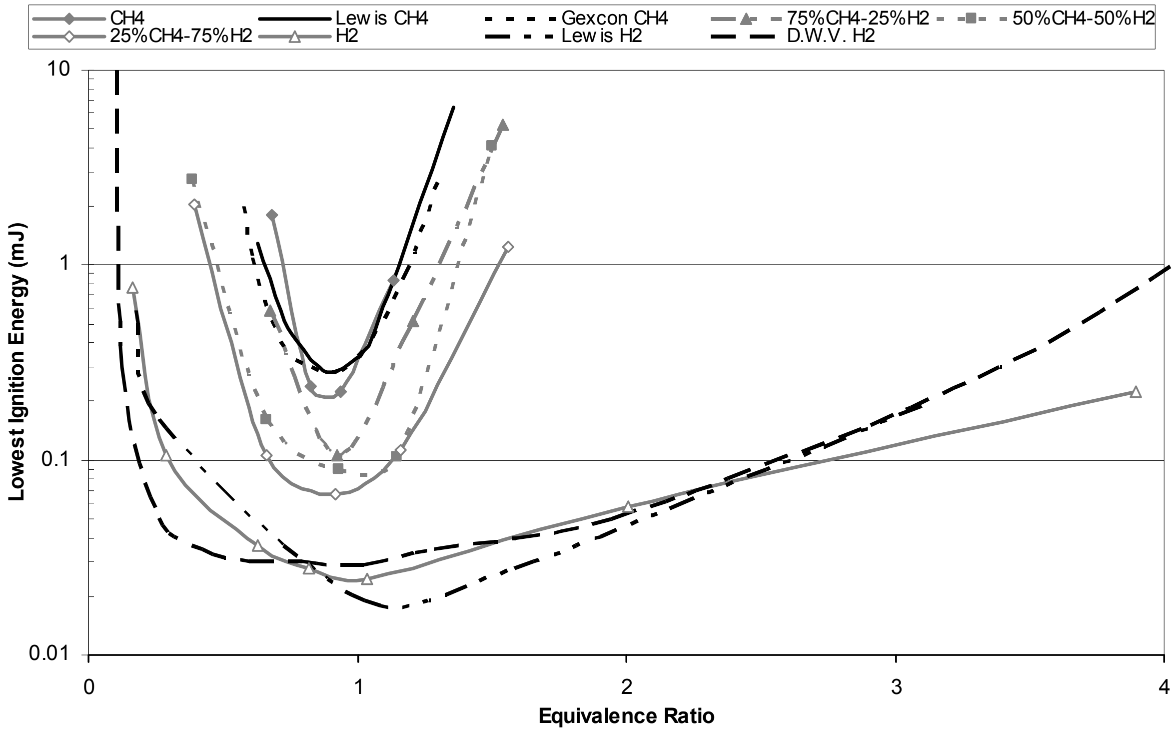

Figure 5: Lowest Ignition Energy Plotted against the Mean Actual Equivalence Ratio

Fuel Mixture	Minimum Ignition Energy (EM) (mJ)	Equivalence Ratio at which Minimum Ignition Energy Occurred (ERM)
100 % CH4 + 0 % H2	0.20	0.89
75 % CH4 + 25 % H2	0.10	0.95
50 % CH4 + 50 % H2	0.09	0.99
25 % CH4 + 75 % H2	0.07	1.00
0 % CH4 + 100 % H2	0.02	1.10

Table 4: Minimum Ignition Energy and Equivalence Ratio at which Minimum Ignition Energy Occurred for each Fuel Mixture

The actual equivalence ratios (*ER*) for which results were obtained for each fuel mixture were then normalised as follows:

If *ER* was greater than ER_M then the Normalised Equivalence Ratio (ER_N) was obtained as follows:

$$ER_N = \frac{ER - ER_M}{ER_{UFL} - ER_M} \quad (2)$$

If *ER* was less than ER_M then the Normalised Equivalence Ratio (ER_N) was obtained as follows:

$$ER_N = \frac{ER - ER_M}{ER_M - ER_{LFL}} \quad (3)$$

where ER_{UFL} and ER_{LFL} are the equivalence ratios corresponding to the Upper and Lower Flammability Limits, respectively, for each fuel air mixture.

The Lowest Ignition Energy (E_L) was normalised by dividing by the Minimum Ignition Energy (E_M). That is:

The Normalised Lowest Ignition Energy (E_{LN}) was obtained as follows:

$$E_{LN} = \frac{E_L}{E_M} \tag{4}$$

The normalised results as described above are shown in Figure 6 in which $\ln(E_{LN})$ is plotted against ER_N.

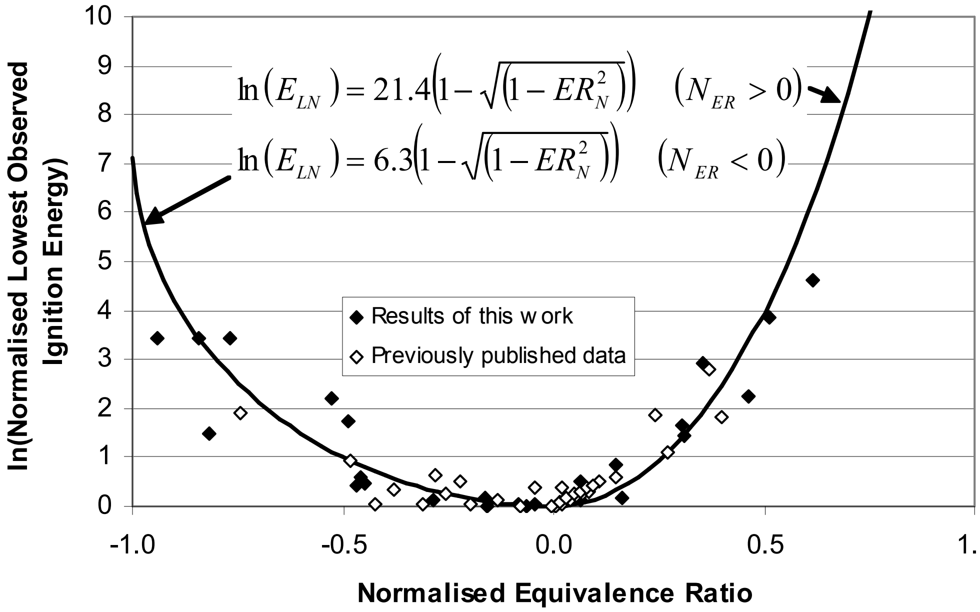

Figure 6: Natural Logarithm of the Normalised Lowest Ignition Energy against Normalised Equivalence Ratio

Two elliptical curves are shown on Figure 6. One curve is fitted to the data relating to negative values of ER_N and the other is fitted to the data relating to positive values of ER_N. Although other functions could have been selected to fit to the data, an elliptical function was selected because it displayed the necessary attributes, that is, it could be configured to be tangential to the X-axis at $ER_N = 0$, and tangential to vertical lines representing constant values of ER_N of -1 and 1. In addition, it provided a good fit to the data. Also shown on Figure 6 are the previously published data as detailed earlier on Figure 5.

4.3 Ignition Probability

As for the results described in Section 4.1, an analysis of the results obtained during Test Series B (fuel composition 75 vol.% CH_4 and 25 vol. % H_2) are presented as an example of the analysis undertaken for all the test series relating to ignition probability.

The data displayed in Figure 4 shows that, in general, there were more successful ignitions at the higher levels of ignition energy than at the lower levels. Therefore further analysis was carried out to determine the probability of achieving ignition for a particular fuel concentration at a particular energy level.

The entire range of ignition energies was sub divided into *m* bands each 2 mJ high. In each band there were a number of ignitions and non-ignitions. In addition it was assumed that if a fuel-air mixture had been successfully ignited at an energy level within a lower band, then this mixture would also have been ignited had the energy level been greater. Therefore, in calculating the probability of ignition associated with a given energy band, successful ignitions from all lower energy bands were also included.

Consequently the Ignition Probability (IP_m) associated with a particular ignition energy level (m) is given by:

$$IP_m = \frac{\sum_{j=1}^{m} SI_j}{NI_m + \sum_{j=1}^{m} SI_j} \qquad (5)$$

where SI_j = number of successful ignitions in band j

NI_m = number of non-ignitions in band m

The data shown in Figure 4 for Test Series B was analysed using Equation 5, as described above, to give the ignition probability associated with each ignition energy band for each actual equivalence ratio. This information is presented in Table 5. Similar information for the other Test Series A and C to E are also presented in Table 5.

The data presented in Table 5 for Test Series B is displayed in Figure 7.

As can be seen from Table 5 and Figure 7, the probability of ignition increases with increasing ignition energy. The rate of rise in probability is steepest for equivalence ratios close to the equivalence ratio at which the minimum ignition energy occurs. For example, for 75 vol.% CH_4 and 25 vol. % H_2, this equivalence ratio is 0.95, and it can be seen on Figure 9 that the probability of ignition increases most quickly when the equivalence ratio is 0.93.

Test Series A: 100 vol. % CH_4 – 0 vol. % H_2							
Mean ER	Ignition probability for energy bands						
	0-2 mJ	2-4 mJ	4-6 mJ	6-8 mJ	8-10mJ	10-12mJ	
0.68	0.11	0.09	0.44	0.27	0.65	0.79	
0.82	0.29	0.82	0.95	0.95	1.00	0.95	
0.94	0.49	1.00	0.90	1.00	1.00	1.00	
1.13	0.15	0.47	0.75	0.68	0.88	0.94	
Test Series B: 75 vol. % CH_4 – 25 vol. % H_2							
Mean ER	Ignition probability for energy bands						
	0-2 mJ	2-4 mJ	4-6 mJ	6-8 mJ	8-10mJ	10-12mJ	
0.67	0.52	0.94	1.00	1.00	0.95	1.00	
0.93	0.73	1.00	1.00	1.00	1.00	1.00	
1.20	0.10	0.56	0.79	0.94	1.00	0.95	
1.54	0.0	0.0	0.07	0.15	0.40	0.55	
Test Series C: 50 vol. % CH_4 – 50 vol. % H_2							
Mean ER	Ignition probability for energy bands						
	0-2 mJ	2-4 mJ	4-6 mJ	6-8 mJ	8-10 mJ	10-12mJ	12-14mJ
0.39	0.00	0.11	0.29	0.43	0.54	0.71	0.8
0.66	0.67	1.00	1.00	1.00	1.00	1.00	-
0.93	0.72	1.00	1.00	1.00	1.00	1.00	-
1.15	0.65	1.00	1.00	1.00	1.00	1.00	-
1.50	0.00	0.00	0.11	0.18	0.38	0.50	-
Test Series D: 25 vol. % CH_4 – 75 vol. % H_2							
Mean ER	Ignition probability for energy bands						
	0-2 mJ	2-4 mJ	4-6 mJ	6-8 mJ	8-10mJ	10-12mJ	
0.40	0.00	0.08	0.38	0.63	0.69	0.75	
0.67	0.69	1.00	1.00	1.00	1.00	1.00	
0.92	0.74	1.00	1.00	1.00	1.00	1.00	
1.16	0.65	1.00	1.00	1.00	1.00	1.00	
1.56	0.05	0.29	0.55	0.75	0.85	0.88	
Test Series E: 0 vol. % CH_4 – 100 vol. % H_2							
Mean ER	Ignition probability for energy bands						
	0-4 mJ	4-8 mJ	8-12 mJ				
0.16	0.03	0.10	0.15				
	Ignition probability for energy bands						
	0 - 0.5 mJ	0.5 – 1 mJ					
0.63	0.72	1.00					
0.82	0.77	1.00					
1.04	0.77	1.00					
2.01	0.61	1.00					
	Ignition probability for energy bands						
	0 – 1 mJ	1 – 2 mJ	2 – 3 mJ	3 – 4 mJ	4 - 5 mJ		
0.29	0.22	0.62	0.79	0.89	0.95		
3.91	0.17	0.42	0.77	0.84	0.91		

Table 5: Ignition Probability for each Ignition Energy Band for each Equivalence Ratio

5.0 DISCUSSION

The data, displayed in Figure 5 shows that, as expected, the lowest ignition energy decreased from the value for methane down to the value for hydrogen as the proportion of hydrogen in the fuel mixture increased. Also, again as expected, the range of equivalence ratios over which ignition occurred increased. The minimum ignition energy for each fuel (that is, the lowest value of all the lowest ignition energies across the flammable range) occurred at a fuel-air ratio close to stoichiometric.

The data was collapsed onto a single curve by normalising the lowest ignition energy for a particular fuel-air mixture and the equivalence ratio. The results of this normalisation and the collapse of the data are shown in Figure 6. Using the relationships shown on Figure 6 and information on the minimum ignition energy and the equivalence ratio at which the minimum ignition energy occurs, it is possible to predict the lowest ignition energy for any methane-hydrogen-air mixture across the flammable range.

The results also showed that the ignition process has a probabilistic element and that the probability of ignition is related to the equivalence ratio and the energy level of the source. It was observed that the probability of ignition increased with increasing energy of the source and that the rate of rise in probability was steepest for the equivalence ratios close to the equivalence ratio at which the minimum ignition energy occurs.

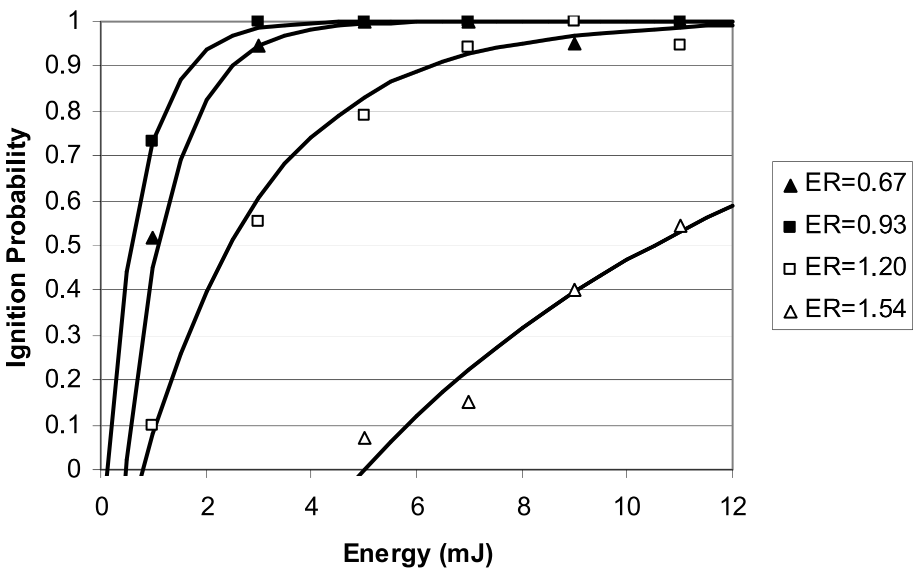

Figure 7: Ignition Probability against Energy for each Equivalence Ratio for Test Series B

6.0 ACKNOWLEDGEMENTS

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