Minimum ignition energy and ignition probability for Methane, Hydrogen and their mixtures

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Minimum Ignition Energy and Ignition probability for Methane, Hydrogen and their mixtures

by

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A Doctoral Thesis Submitted in partial fulfillment

of the requirements for the award of

DOCTOR OF PHILOSOPHY

of Loughborough University

June, 2009

Department of Chemical Engineering

ABSTRACT

In the present hydrocarbon economy, energy is primarily derived from fossil fuels, like Coal, Oil and Gas. The petroleum oil obtained from mother earth is further, refined into gasoline, diesel, and natural gas. However, the burning of these hydrocarbon fuels causes the emission of greenhouse gases and other pollutants. Hydrogen the lightest of all gases and the most abundant element in the universe, is being considered for use as an energy carrier (for storing and transporting energy) for future generations. Emphasis on mitigating global climate change and reducing pollution, strengthens the case of hydrogen over other fuels. The environmentally benign nature of hydrogen coupled with the finite supply of fossil fuels supports the hydrogen economy.

A possible transition to the full hydrogen economy is envisaged which will take place through several phases. The current work is concerned with the transitional phase and involves an investigation into the possibility of using the existing natural gas infrastructure for transporting hydrogen as a natural gas-hydrogen mixture. Likely impacts on the natural gas infrastructure as a consequence of the introduction of hydrogen are being studied as part of a European Union funded research project called Naturalhy. The work that is the subject of this thesis forms part of the safety work package of the Naturalhy project. In turn the part of the safety work package with which the work of this thesis is concerned is the changes that handling a mixture of natural gas and hydrogen rather than natural gas will have on the risk that will be posed to the general public. In particular, it is concerned with the changes that might result to such parameters as the ease with which mixtures of hydrogen and natural gas might be ignited compared with natural gas and hence the change to the frequency with which such events as explosions within domestic properties might increase.

The work commenced with a review of the literature on the subjects of failure probability and ignition probability associated with natural gas infrastructure. The analysis and the outcome of this literature review suggested that the most sensitive area affected by the addition of hydrogen is accidental gas releases into confined enclosures such as domestic property. The presence of hydrogen is likely to increase the probability of fire and/or explosion due to the characteristic properties of hydrogen (wide flammability range, lower minimum ignition energy etc.).

The ignition characteristics for the gases (methane, hydrogen and methane-hydrogen mixtures) was studied using an experimental rig based on the principle of capacitive spark discharge. Consequently, the data obtained through experiments was used to calculate the Minimum Ignition Energy (MIE) of a particular gas and the Lowest Ignition Energy at various flammable gas concentrations for a particular gas. The results and observations were further analysed to provide information on the ignition probability associated with various ignition energy values for all the gases. The results for MIE are compared with the available data in the literature for methane and hydrogen gas.

Generalised correlations for predicting the ignition energy for pure gases and for two component (methane-hydrogen) gas mixtures were developed. Methane gas release incidents are compared with hydrogen to estimate increases in the probability of fire and/or explosion incidents using a few deterministic release rates for the two gases.

ACKNOWLEDGEMENT

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Nomenclature used

A   -   the area of the region being in contact with the gas cloud (m^2)

a   -   probability of Ignition source being active

b   -   constant

C   -   Concentration (%)

ΔC -   Error in parameter C (%)

CP   -   Capacitance (pF)

d   -   Pipe diameter / shell diameter (m)

d_h -   diameter of hole or puncture in pipeline (m)

d_o -   Outside diameter of shell (m)

d_w -   diameter of optical window (for the experimental vessel) (m)

E   -   Energy or Capacitive spark energy (mJ)

EF   -   Expansion Factor

ER   -   Equivalence Ratio

f   -   Allowable working stress (MPa)

I   -   Current magnitude (ampere)

IE   -   Ignition Energy (mJ)

IG   -   Number of successful Ignitions of the gas air mixture

IP   -   Ignition probability

LFL   -   Lower Flammability Limit

m   -   mass release rate of gas (kg/s)

MIE   -   Minimum Ignition Energy (mJ)

MW   -   Molecular Weight (gm)

N   -   Number of Ignition sources

NER   -   Normalised Equivalence Ratio

NI   -   number of Non – Ignitions for the gas air mixture

NP	-	Number of moles of all Products formed
NR	-	Number of moles of all reactants
P, p	-	Pressure (kPa)
ΔP	-	Error in parameter P (kPa)
ph	-	Hoop stress (MPa)
po	-	ignition potential (probability of ignition) of single source
qd	-	Quenching distance (cm)
Q	-	probability of Non Ignition
R	-	Ratio of moles of methane to moles of hydrogen
ΔR	-	Error in parameter R
SPH	-	Specific heat of gas (J/mol/K)
Su	-	Laminar burning velocity (cm/s)
T	-	Temperature (K)
Tf	-	Final temperature (adiabatic flame temperature) (K)
Ti	-	Initial temperature (room temperature) (K)
T_b	-	Burnt gas temperature (K)
T_u	-	Un-burnt gas temperature (room temperature) (K)
t	-	time duration (s)
t_a	-	time period for which the source is active (s)
t_i	-	time period between each activation of the source (s)
th_s	-	Shell thickness (for the experimental vessel) (mm)
th_w	-	Window thickness (for the experimental vessel) (mm)
UFL	-	Upper Flammability Limit
V	-	Voltage (volt)
ΔV	-	Error in parameter V (volt)

Greek symbols

μ -   ignition sources density per unit area of source ($1/m^2$)

λ -   frequency of activation of the source (1/s)

ρ -   molar density of gas (mol/m^3)

Ω -   resistance (ohms)

Subscripts

av	-	average value
gas	-	flammable gas under consideration
gas-air	-	flammable gas air mixture
MIE	-	at the MIE condition of the flammable gas
max	-	Maximum value
min	-	Minimum value
lower	-	Lower value
upper	-	Upper value
LFL	-	Lower Flammability Limit
UFL	-	Upper Flammability Limit
vac	-	vacuum condition
t	-	time

Introduction

1.1 Background

Life on Earth is driven by energy. Plants take it from solar radiation and are often referred to as the "initiator of life" on this planet. Energy captured slowly by photosynthesis is stored up often in a more denser and useable form. As denser reservoirs of energy were found over the course of Earth's history, we tended to use more energy by exploiting these denser resources. The ability to use energy outside the body, enables humans to use far more energy than any other living creature on this planet. The control of fire and the exploitation of fossil fuels (denser resources) have made it possible for us to release, vast amounts of energy, in a short time, that took centuries to accumulate within our mother earth, often referred to as natural resources.

Energy is critical to every aspect of our lives. All material things that are of any use to us – our food, clothes, houses, automobiles, – require energy to make and energy to use. All human activities like – working, managing, thinking, teaching, – require human energy. All such useful activities are often called "work." Thus, all work requires energy. In performing work, energy is always changed from more-concentrated to less-concentrated forms. In fact, this natural tendency gives energy its ability to perform work. Material things, such as food, wood, plastic and gasoline actually are concentrated forms of energy. Matter can be changed into energy, as when we eat food or burn gasoline. Usefulness can also be gained by changing the form of energy, as in using heat to make electricity and electricity to produce light. Whenever energy is used to perform work, it becomes more dispersed and disorganized.

Everything we do is connected to energy in one form or another. The amount of energy utilised may vary for each individual. Energy maintains our standard of living and economy. The developed world often takes it for granted that energy will be available whenever it is needed. Moreover industrial research and development is

aimed at bringing energy to the door step of each and every customer, hence the need for "Energy transportation".

Energy can be obtained through various sources provided by nature. The most common sources such as coal, oil, gas, wind, sun and nuclear fuels are often used for satisfying energy demands. During the early stages, the emphasis was on obtaining energy and less attention was paid towards its consequences. The impact on surrounding ecology, environment and finally on humans was felt with the increased utilisation of energy from various sources. In the present world most of this energy comes from fossil fuels (coal, oil, and gas) which are depleting at an ever increasing rate with the increase in population and our living standard. At current rates of consumption, known reserves of oil & gas will only last for a few decades and coal for a few centuries.

The technology for the commercial utilization of fossil fuels started in the early 18th century and hence this technology for harnessing the energy contained within resources such as coal, oil and gas is very well developed. The exploitation of these resources has resulted in making life easier through utilising the energy released to provide services such as transport, cooking, communication etc. Since the 1970's the adverse impact of combustion of these fossil fuels on the environment and the entire ecosystem has been noted. Consequently, research is aimed at reducing the environmental impact of fossil fuels by reducing the environmentally harmful emissions. In addition, to ensure the security of supply of energy sources, research is being undertaken to identify sustainable sources of energy. This will allow a transition to be made from the finite fossil fuels to the infinite sustainable sources of energy.

The energy sources are often categorised as renewable and non-renewable. Renewable energy sources are those which are continually being replaced such as energy from the sun (solar) and wind. If an energy resource is being used faster than it can be replaced (for example, coal takes millions of years to form) then it will eventually run out. Hence all the effort that is being expended to conserve these non-renewable energy sources.

These days, more emphasis is being focussed on the renewable sources which are believed to supply sustainable source of energy. Solar, Wind, Tidal, Wave, Biomass, Hydroelectric, Geothermal, etc. are often referred to as renewable energy sources but it is not guaranteed that they will produce energy all of the time. For example, renewable energy sources, like the sun, can not produce energy all the time. The sun does not always shine, so a device to store this energy and transport it from the location of generation to the location of utilization is required. Some experts believe that hydrogen can store this energy until it is needed. Then it can be transported to where it is needed, and used without producing harmful emissions. Hence, hydrogen has the capacity to form part of the energy infrastructure for future generations.

Indeed some experts think that **hydrogen** will form the basis of the energy infrastructure that will power future societies, replacing today's natural gas, oil and coal. They see a new "hydrogen economy" to replace our current "fossil fuel-based economy".

1.2 Motivation

Hydrogen, the lightest of all gases and the most abundant element in the universe, is being considered for use as an energy carrier (a means of storing and transporting energy obtained from sustainable sources) for the twenty first century because the recovery of energy stored in the form of hydrogen can be obtained in environmentally benign ways. The idea of using hydrogen as an energy carrier has been around at least since 1870, when Jules Verne incorporated the concept in his science-fiction classic, 20,000 Leagues under the Sea. There is a widespread assumption that the increased use of hydrogen as an energy carrier will bring significant benefits. Some emphasise the prospect of cleaner air in cities as vehicles stop emitting pollutants, others point towards mitigating global climate change.

The **hydrogen economy** is in its infancy. Research is required to increase production from sustainable sources. Research is needed on CO_2 sequestration, if H_2 is produced from a source containing carbon. Research is needed on the utilization of hydrogen such as fuel cells and combustion equipment.

While moves towards an increased use of hydrogen are starting to gather speed, this growth is restricted by a number of constraints at the political, commercial, technical and social levels. Safety concerns are still widespread in relation to the use of hydrogen in our daily routine life. The public perception of the dangers of hydrogen transportation and distribution need to be addressed if widespread hydrogen use is envisaged in the future. At a practical level, there are real issues in terms of how hydrogen is stored and transported. Hydrogen is a very light gas making it far more difficult to work with than other liquid / gaseous fuels.

In the meantime, in order to be ready to swing into action, when the above research bears fruit, work needs to be done on ways of transporting hydrogen from the point of production to the point of utilization. The most economical, safest and environmentally friendly method of transporting large quantities of liquids and gases on a regular basis is by pipeline. Hence the construction of the natural gas, and water pipeline networks and pipelines for gasoline, etc. A small number of purpose built hydrogen pipelines exist. However, the construction of a nation wide hydrogen network will only commence, if at all, once large quantities can be produced and a market exists for its use.

From an economic point of view, the costs of switching over to a full hydrogen based economy are high. For example, the capital investment required to convert existing gasoline stations to provide hydrogen to vehicle drivers will run into millions of pounds. The present phase can be termed as a transition phase for the hydrogen economy. A means of carrying hydrogen from its production to its utilization sites is required during the period of transition; from now until the full hydrogen economy is introduced. A system is required that can readily increase in volume and throughput as production and demand increases and can also manage the mismatch between production and demand. Such systems already exits for natural gas and an examination of the use that the natural gas infrastructure can play in the transport of hydrogen, in particular during the transition to the full hydrogen economy, must be undertaken.

The distance between the production and utilization points for hydrogen can be a few metres, to hundreds of kilometres. This requires a connection between the production and the end-use facilities. This link can be made by means of road, rail, ship, pipeline or air transport. As mentioned above, pipelines have been shown to be the most

economical, safest and environmental friendly method of transporting large quantities of material such as liquid or gas on a regular long term basis. This most viable option has resulted in the development of the extensive natural gas infrastructures within Europe, USA, Asia, etc. and is still expanding to other areas of the world.

Europe's Natural Gas (NG) pipelines are some of the most advanced in the world and the infrastructures are evenly spread. These infrastructures are also connected to other continents such as Asia and Africa. It is prudent to investigate whether or not these existing NG infrastructures can be utilised to assist a transfer to the hydrogen economy.

A possible vision of the use of the NG infrastructures to assist a transition to the hydrogen economy is through various phases where increasing amounts of hydrogen are blended with the NG to produce a mixture. The financial burden during the transition period will be eased considerably by making use of the existing NG infrastructure with its inherent economic value. It will also help in achieving a seamless move to the hydrogen economy.

Several research and development activities have been initiated by the European commission (EC) to study the subject of hydrogen transportation, including by pipelines. These include the projects NATURALHY, HYSAFE, HyWays, Roads2HyCom etc. The NATURALHY project is focussed especially on studying the practicality of using the existing natural gas pipeline networks to assist in the transition to the hydrogen economy by transporting mixtures of natural gas and hydrogen.

1.3 The Naturalhy project

The NATURALHY project is a major "Integrated Project" which has been funded by the European Commission within the sixth framework programme. The purpose is to prepare the European natural gas industry for the introduction of hydrogen by assessing the capability of natural gas infrastructures to accept mixtures of hydrogen and natural gas.

The objective of the NATURALHY project, is to investigate the feasibility of using the natural gas infrastructures (pipeline networks) for transporting hydrogen-natural gas mixtures from the point of hydrogen production to the point of use where the hydrogen is extracted from the mixture. It is envisaged that this would aid the transition to a hydrogen economy, i.e. under an expected situation where the demand for hydrogen is increasing from a low base and the hydrogen supply is carried out in parallel with the supply of natural gas.

The hydrogen injected into the natural gas network could be extracted by techniques like membrane separation and Pressure Swing Adsorption (PSA) to utilise hydrogen in a pure form. However, whatever separation technique is adopted, not all the hydrogen can be extracted, and the gas supplied to users connected to the network is likely to contain some amount of hydrogen. As hydrogen combustion will not result in CO_2 formation there is some environmental benefit and this is often referred to as the "greening of gas". Hence the greening of gas is a secondary objective of the Naturalhy project and will result in Environmental benefits towards a reduction in CO_2 emissions. In 2004, the total primary energy consumption in the EU15 countries amounted to 6.54×10^4 kJ of which 24% was provided by natural gas. If 1% (equivalent to 3% by volume) of the energy content of the natural gas were replaced by hydrogen (produced through CO_2 free production technique), the total CO_2 emission of the EU15 would be reduced by about 7.4 Million-tons/year (in the case of oil 11 Million-tons/year and in the case of coal 13 Million-tons/year). This is about 0.2 % of the total annual CO_2 emission in the EU15 countries. Accordingly, the potential of hydrogen-natural gas mixtures for the reduction of CO_2 emissions could be significant. (Florisson, et. al., 2006). To summarise, the NATURALHY project objectives are two fold

- Means of transporting hydrogen from point of production to point of use by hydrogen fuelled applications

- 'Greening' of gas by replacing some NG by H_2 , resulting in lower emissions

NATURALHY has a total budget of 17.3 million euros and started in May 2004, for a duration of five years. The project partners comprise of gas companies, research institutes, academic institutes, consultants, government bodies and planning and implementing organisations, with Gasunie Research (N.V. Nederlandse Gasunie) assigned the role of project coordinator.

The NATURALHY project is subdivided into eight Work Packages (WP) each based on an area of research and development. The WP project title, WP lead organisation and a short description of the work being conducted are presented below :

Work Package	Project Title	Project Leader
WP1	Socio-economic and Life Cycle Analysis	Loughborough Univ., UK
WP2	**Safety**	**Loughborough Univ., UK**
WP3	Durability	Gaz de France, France
WP4	Integrity	Gas - und Umwelttechnik GmbH (DBI), Germany
WP5	End Use	University of Oxford, UK
WP6	Decision Support Tool	Instituto de Soldadura e Qualidade (ISQ), Portugal
WP7	Dissemination	Exergia, Greece
WP8	Project management	Gasunie Research, The Netherlands

WP1 Life Cycle and Socio-economic Assessment comprises a comparison of the existing natural resource requirements, environmental impacts, employment consequences and economic costs over the complete life cycle of current natural gas and related energy systems; proposed transitional natural gas/hydrogen systems; and future complete hydrogen systems from source to point of use.

WP2 Safety is discussed at the end of this section in detail.

WP3 Durability determines the effect of hydrogen on the durability of materials and components used in the natural gas infrastructure. For example, the permeability of pipeline materials to hydrogen is being assessed and any effect on "ageing". Hydrogen impact on pipeline materials is being evaluated through experimentation, the results processed to produce mathematical durability lifetime models. A durability assessment tool developed for assessing the ageing of pipeline materials and components, will help provide a practical way of evaluating the lifetime of pipeline systems under an increasing percentage of hydrogen.

WP4 Integrity assesses the effectiveness of NDE (Non Destructive Evaluation) tools and techniques to monitor the condition of pipelines (transmission and distribution) exposed to natural gas-hydrogen mixtures. The maintenance and repair procedures for such pipeline systems are evaluated. Critical defects leading to pipeline failures, under gaseous atmosphere are studied to arrive at the defect assessment criteria relevant for natural gas-hydrogen mixtures. The output of this package is to develop an integrity management tool for a pipeline system conveying a natural gas – hydrogen mixture.

WP5 End Use examines the implications of providing distributed pipeline natural gas-hydrogen mixtures to end-users and the effect on existing appliances. The impact of added hydrogen on the performance characteristics of domestic and industrial burners are studied. This will lead to greater understanding of the requirements for appliances to operate safely and efficiently with increasing levels of hydrogen. This work package is also studying the potential of using high efficiency membranes for separating hydrogen from the gas mixture for end-use applications as an alternative to PSA. Methods for controlling the gas quality within the network as hydrogen is added and removed in the network are also being studied.

WP6 Decision support tool brings together the results of WP1 to WP5 into a software based decision support tool, which can be used by pipeline operators to assess the possibility of adding hydrogen to their network or part of their network.

WP2 : Safety

The aim of Work Package 2 is to evaluate the change in risk presented to the public by the natural gas infrastructure, if it were used to carry a mixture of natural gas and hydrogen rather than simply natural gas. The existing gas pipeline infrastructure was designed, constructed and operated based on the premise that natural gas is the material to be conveyed. Since, hydrogen has different properties to natural gas, which may adversely affect the risk presented to the public, this must be re-evaluated for the mixture.

Risk is defined as a combination of likelihood of an undesired event (such as pipeline failure, gas release, gas ignition etc.) and the consequence of that event (such as the severity of heat load from fire or overpressure wave from explosion). Mathematically risk is expressed as :

$$RISK = \sum_{\text{All events}} \text{likelihood} \times \text{consequences}$$

For gas pipelines, the likelihood can be due to 'pipeline failure' leading to gas release. The likelihood of ignition of released gas often termed as 'ignition probability', can be also considered (along with pipeline failure) to arrive at the likelihood of a fire. Consequences depend on the properties of released gas (flammability limits, heat of combustion, specific heat, flame temperature, burning velocity etc.) and also the surrounding conditions (weather, confined/unconfined release etc.). Different scenarios/events can be considered to calculate risk through summation of all these events to arrive at the risk level for the gas pipeline system.

Adding hydrogen to the gas infrastructure may affect both the likelihood and consequence (severity) of undesired events and hence potentially change the risk level. It is important to quantify this effect in order to establish if the risk remains acceptable and to identify the maximum hydrogen concentration that can be added to the natural gas without this risk becoming unacceptably high.

To this end, the Safety Work Package is examining the fire and/or explosion hazard situations pertinent to the gas infrastructure, based on a natural gas-hydrogen mixture being involved. To assess the change in consequences small and large scale experiments are undertaken to provide data to aid model development and validation. Specifically these are:

- Laboratory experiments to assess the burning velocities of methane/hydrogen mixtures
- Large scale experiments of
    - o gas build up and explosions in a domestic environment

- o gas build up and explosions in an industrial enclosure/building

- o explosions in congested regions (Vapour Cloud Explosions)

- o high pressure Jet fires

- o fire hazards following failure of a transmission pipeline

Consequence models are being developed and validated using this experimental data, These models are then used to assess the impact of different levels of hydrogen addition on the severity of the hazards. Fire and explosion consequences, and impact of hydrogen on these consequences, are not part of this thesis, as the focus here is on the likelihood of the event.

The impact of hydrogen on the likelihood of an adverse event is studied through experiments in this work package and also by work within the work packages on Durability and Integrity. Data from WP3 and WP4, will help re-evaluate the failure frequencies of pipelines and associated equipment. Within WP2, Laboratory scale experiments are being conducted to help to re-assess ignition probability. Specifically, ignition probability and the minimum ignition energy of natural gas-hydrogen mixtures are being studied and this is the main focus of this thesis.

The above work on likelihood and consequences will then be combined to produce a risk assessment tool, which can be applied to gas transmission networks conveying methane-hydrogen mixtures. The risk to the public can be calculated for differing levels of hydrogen introduction and compared with the risk for a system conveying natural gas. The risk assessment tool will evaluate both individual and societal risk to the surrounding population. A risk assessment methodology for low pressure releases within a property, addressing the risk of an explosion will also be considered. Again the situation with natural gas–hydrogen mixtures will be compared to that with natural gas alone.

Natural gas properties are often considered as similar to pure methane, since it is the major constituent of the natural gas. A comparison of hydrogen and methane properties suggest changes (in likelihood and consequences parameters) as

presented in Table 1.1. Only qualitative changes are presented here, since quantification is the objective of study within the NATURALHY project.

Table 1.1 : Qualitative change due to hydrogen addition

Parameter	Potential qualitative change compared to natural gas
Leaks (pipeline failures)	Increase
Ruptures (pipeline failures)	Increase
Ignition energy for gas	Decrease
Ignition probability	Increase
Energy content per m^3	Decrease
Flammable range in open atmosphere	Increase
Burning velocity	Increase
Heat radiation from flames	Decrease
Explosion severity	Increase
Total Impact on risk	**Unknown**

As a first step towards quantifying the impact of hydrogen addition, on the likelihood of adverse events, the failure probability of high pressure pipelines and low pressure systems conveying natural gas are considered in section 1.4. The effect of hydrogen on this failure probability is considered in section 1.5.

1.4 Likelihood of failure

Despite the fact that pipelines are considered reliable for transmitting energy, there have been incidents of gas release resulting in loss of life and property damage. This has prompted the need to understand the various modes of failure of pipelines, and increasing effort is directed towards reducing the frequency of these accidents. The first step is to estimate the failure probability (frequency analysis) for existing pipelines operating in different parts of the world.

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Pipeline failure frequency estimation based on historical incident data has formed the basis for many risk assessment studies. (Acton 2002 and Willcocks et. al. 2000). The source of data considered in the present analysis are worldwide pipeline failure events. Pipeline failure databases from the UK, Europe and the US are considered. Low pressure failures pertaining to the UK, are also discussed.

1.4.1 The UKOPA pipeline fault database

Natural gas for consumption in the UK comes to shore from the North sea and the Irish sea. The gas, after processing, is transferred through high pressure transmission pipelines, normally up to one metre in diameter, with a pressure of around 70 bar. Compressor stations located at intervals of about 70 km along the transmission pipelines are used to boost the gas pressure. Finally, the local distribution of gas is carried out at low pressure through small distribution pipes from 20 mm to 180 mm in diameter. The gas is then passed to customers through a gas metering system (www.transco.uk.com). The United Kingdom Onshore Pipeline Operators Association (UKOPA) provides the view of the UK pipeline Operators on strategic issues related to safety of pipelines in the UK.

The UKOPA database presents pipeline and product loss incident data from onshore Major Accident Hazard Pipelines (MAHPs) operated within the UK by National Grid, Shell UK, BP, Huntsman and Powergen UK, covering operating experience of 654,732 km.yr. (for 21,727 km of pipeline by the end of 2004). The database is designed to reflect the ways in which the UKOPA operators design, build, operate, inspect and maintain their pipeline systems.

Table 1.2 : Failure Frequency and Causes for Pipeline Failures within UK

Product Loss Cause	Percentage
Girth Weld Defect	19
External Interference	22
Internal Corrosion	1
External Corrosion	18
Unknown	5
Other	23
• Internal cracking due to wet towns gas	17
• Pipe-Fitting Welds	2
• Leaking Clamps	1
• Lightning	0.6
• Soil stress	0.6
• Threaded Joint	0.6
• Electric Cable Arc Strike	0.6
Pipe Defect	8
Ground Movement	3
Seam Weld Defect	1
Total (based on 172 incidents)	**100**
Failure frequency	
Average failure frequency (1962 to 2004) per 1000 km.yr.	**0.263**
Failure frequency for the last 5 years (2000 – 2004) per 1000 km.yr.	**0.028**

The overall failure frequency over the period 1962 to 2004 is 0.263 incidents per 1000 km.yr.. The failure frequency over the last 5 years (2000 – 2004) is 0.028 incidents per 1000 km.yr., which is a better representation of the pipeline failure frequency for

new generation pipelines. The causes of the failures of the pipelines for the various incidents as observed are given in Table 1.2.

1.4.2 The EGIG report on gas pipeline incidents

As per 2003 figures, 485 billion cubic meters of gas was supplied to Europe with 62% indigenous production and 38% imports. As per gas supply and consumption statistics Russia, Algeria, Norway and Netherlands are likely to remain as gas suppliers for the European countries (Stern, 2002 & Beckervordersandforth, 2004).

The unintentional release of gas from transmission pipeline systems is compiled to produce a comprehensive database for the European pipeline infrastructure. The initiative to gather data was started in the year 1982 by six system operators and by the year 2007 a total of fifteen companies were participating, comprising all of the major gas transmission system operators in Western Europe. This co-operation of gas companies is named EGIG (European Gas pipeline Incident data Group).

The EGIG report on gas pipeline incidents is based on the combined operating experience (3.15 million km. yr. from 1970 to 2007) of fifteen European gas transmission and system operators. The failure frequency and the distribution of the causes of the pipeline incidents for EU pipelines are reported in 7th EGIG report (2008) and is given in Table 1.3.

Table 1.3 : Failure frequency and Incident cause for gas pipelines from EGIG database

Incident cause	Percentage
External interference	49.6
Construction defect/material failure	16.5
Corrosion	15.4
Ground movement	7.3
Hot-tap made by error	4.6
Other	6.7
Total (based on 1123 incidents)	100
Failure frequency	
Overall failure frequency (1970 to 2004) per 1000 km.yr.	**0.37**
Failure frequency for the 5 years (2000 – 2004) per 1000 km.yr.	**0.17**

A total of 1172 incidents during this period gives the failure frequency of 0.37 incidents per 1000 km.yr. The failure frequency for the five years (2000 - 2004) is 0.17 incidents per 1000 km.yr. for a total exposure of 5.7 x 10^5 km. yr. during this period. The five year average failure frequency is less than half of the average frequency between 1970 - 2007, suggesting that improvement in the pipeline technology and better maintenance practices have had a favourable impact resulting in the reduction of failure frequency. However, these EGIG values are far greater than the UKOPA values.

1.4.3 The US Department of Transport (DOT) data

The U.S. natural gas transmission pipeline network consists of a total of 477,012 km. (298,133 miles) of pipelines consisting of 31,782 km. (19,864 miles) of gathering lines, plus 445,230 km. of transmission lines. These transmission pipelines feed over 2.96 million km. (1.85 million miles) of distribution lines (mains and service lines) operated by around 2,500 natural gas pipeline operators. The United States currently consumes

about 1.8 billion cubic meters (63 billion cubic feet) of natural gas daily (0.65 trillion cubic meters annually) (OPS, USA).

The office of pipeline safety (DOT, US) has recorded 2042 incidents from the natural gas transmission operators for the period 1 January 1986 to 31 December 2007. The data suggests an average of 102.1 incidents per calendar year. If we consider that the total transmission pipelines for the US is around 477,000 kms. (US DOT, 2007), then the failure frequency for these lines are approximately 0.214 incidents per 1000 km.yr. This value is comparable with the EGIG data. The causes of onshore incidents for natural gas transmission and gathering systems during the period 1985–2001 is presented in Table 1.4.

Table 1.4 : Failure frequency and various causes for gas transmission pipeline incidents within the USA

Cause	Percentage
External interference (Third-party damage)	28
External corrosion	17
Internal corrosion	11
Natural forces	10
Miscellaneous	2
Incorrect operation	3
Unknown	7
Other failure	4
Construction/installation	6
Manufacturer	6
Previously damaged pipe	4
Malfunction	1
Stress corrosion cracking	2
Vandalism	–
Total (based on 662 incidents)	**100**
Failure frequency	
Overall failure frequency (1986 to 2007) per 1000 km.yr.	**0.214**
Failure frequency for the last 5 years (469 incidents from 2000 – 2004 per 1000 km.yr.	**0.209**

(Source : Transportation research board, US)

The pipeline incidents databases considered in the above analysis show that these events have a very low frequency. However, they are important for risk assessment,

since they may have very large impact (consequences). The next section considers the low pressure side of the NG infrastructure.

1.4.4 Low pressure pipeline incidents

In risk analyst terminology, these incidents are typically high frequency, but relatively low consequences event. The incidents considered here are of gas releases from the distribution network, the meter area, carcass pipe-work (internal) and the home appliances.

The actual number of low pressure gas escapes are difficult to quantify since this number is based on 21.7 million gas customers in UK (year 2006) and public reporting of these gas escape incidents. Moreover a single gas escape is often reported by numerous customers and presently there is no provision to eliminate multiple calls for the same gas escape. The National Gird handled nearly 2.3 million emergency calls (in the year 2006 - 07) on the UK national gas emergency number on behalf of all the network operators (www.nationalgrid.co.uk).

To arrive at any gas release frequency the pipeline operators often only consider the mains and the service pipeline failure incidents, since maintenance of these pipelines are under the control of the gas distribution companies. The Gas Safety (Management) Regulations 1996 makes it mandatory for operators to report and investigate gas escapes that occur on their networks. The HSE's Safety Performance Indicator report (2005/06) suggest release frequencies for a period 1 April 2005 to 31 March 2006 as presented in Table 1.5.

Table 1.5 : Annual gas escapes requiring repair for year 2005/06 within UK

Incident	Annual number reported
Gas escapes requiring repair by pipeline operator	157,969
Gas In Building (GIB) cases	954
Major accidents (major injury or structural damage)	6
Fatalities	0

(source HSE, 2006)

Only serious incidents of gas escape get analysed as minor smelly leaks are often stopped as soon as the release is identified, either by the person reporting gas release or by the service engineer. Such releases are not given great importance and may not get recorded.

1.5 Impact of hydrogen

In the present work, hydrogen is considered to be injected into the existing natural gas network and the mixture (natural gas and hydrogen) is considered to be transported through the channels of transmission, distribution and finally to the end use appliance. If such a distribution is considered feasible, then it is imperative to consider the impact of hydrogen as presented in Table 1-1 earlier. Accordingly impact on pipeline failures is considered in the next section.

1.5.1 Impact on pipeline failures

Any gas coming into contact with a solid can be **ad**sorbed, i.e. it can adhere to the surface without penetrating the material, or it can also be **ab**sorbed, i.e. it can diffuse into the material and get stored in its bulk. Because of its small size, the hydrogen atom can rather easily diffuse and accumulate into most structural steels, where it can cause severe degradation, potentially yielding catastrophic failures.

Atomic hydrogen is believed to cause Hydrogen Induced Cracking (HIC) in steel, leading to blisters in the pipe wall surface, but the impact of hydrogen on the overall failure frequency needs to be quantified in order to provide meaningful results. Impact on plastic pipes (polyethylene, PVC, etc.) introduced into the gas distribution system are unknown. Even if, its believed that these pipelines are not affected by hydrogen, the smaller molecular size of hydrogen may lead to more leakages from the pipelines as compared to the steel pipes. The change in risk level because of the introduction of hydrogen needs to be quantified to help decision makers to evaluate the option of hydrogen introduction in the present infrastructure or to compare natural gas and hydrogen pipeline from a safety perspective.

Hydrogen is likely to increase the corrosion rate especially by hydrogen induced cracking and the fatigue crack growth rate because of the inherent properties of hydrogen gas. Hydrogen is known for its deteriorating effect on carbon steel, often referred as hydrogen embrittlement and includes surface cracking, slow crack growth, loss of ductility, and decrease in fracture resistance. This deterioration can lead to premature failure, possibly with little warning (Sofronis et. al. 2005). The quantification of this hydrogen effect, is being undertaken within the Work Packages on Durability and Integrity (WP3 and WP4) of the NATURALHY project.

Incidents involving the failures of the pipelines as per the UKOPA, EGIG and the DOT, US suggests following causes :

- External interference (or third party damage) is the single largest cause of failure contributing around 50 % for the pipelines

- Corrosion is second largest cause of pipeline failures contributing around 15 % to 19 %, which includes internal, external corrosion and stress corrosion cracking.

- Natural forces such as ground movement / lightning / earthquake etc. and human error contributes around 17 % for the failures

- Construction defect and material failure contributes around 16 % (EGIG data)

Examination of the above data suggests that the introduction of hydrogen into the pipeline is not likely to modify the causes for human error and natural forces (ground movement, lightening, earthquake etc.). These suggest that for around 17% of the causes of pipeline failure (for pipelines carrying natural gas and hydrogen mixture) are likely to remain the same. It is believed that hydrogen may have an impact on the other factors contributing to the failure of pipelines.

1.5.2 Impact on ignition probability

Hydrogen has some unique properties compared to other fuels. Some of these unique characteristics can make it safer than natural gas, while others make it more hazardous (www.humboldt.edu). Ignition energy, flammability range and leakage rate can be considered as key parameters influencing the ignition probability in accidental release conditions. Furthermore, the low viscosity and small molecular size of hydrogen may give it a greater propensity to leak than natural gas. For a given pressure and hole size, hydrogen will leak approximately 2.8 times faster than natural gas on a volumetric basis (if pressure is constant). However, the energy density of hydrogen is much lower than that of methane; therefore, the energy leakage rate for hydrogen would be less than that for methane for a given pressure and hole size. As shown in Table 1.6, the flammability range of hydrogen in air covers a much wider range of concentrations than for methane, propane, or gasoline vapour. Furthermore, it can be much more readily ignited under most circumstances due to its extremely low ignition energy. Hydrogen is also detonable over a very wide range of concentrations when confined.

Table 1.6 : Characteristics of hydrogen in comparison with other fuels

Properties	Hydrogen	Methane	Propane	Gasoline
LFL – UFL [a] (vol %)	4 - 75	5.3 – 15	2.1 - 9.5	1 - 7.8
Stoichiometric concentration with air (vol %)	29.5	9.5	4.1	1.8
Minimum ignition energy (mJ)	0.019	0.29	0.26	0.24
LDL – UDL [b] (vol %)	11/18 - 59	6.3 - 13.5	3.1 - 7	1.1 - 3.3

(source: Schatz research centre)

[a] LFL - Lower Flammability Limit   [b] LDL - Lower Detonability Limit
    UFL - Upper Flammability Limit   UDL - Upper Detonability Limit

The principal hazard foreseen for hydrogen systems is the uncontrolled combustion of accidentally released hydrogen. Various common physical processes (open flames, hot surfaces, friction, electrical spark and static discharge) can serve as sources of ignition for most gas releases. The much wider range of flammability and low Minimum Ignition Energy (MIE) may result in hydrogen having a higher ignition probability than other gases/vapours. Increasing dosage of hydrogen to the NG infrastructure is likely to lower the MIE for the gas mixture. This reduction in MIE for the gas mixture, could then increase the number of ignition sources with the potential to provide ignition to the released flammable gas - air mixture. The propensity of these ignition sources, in regions in which infrastructure handling hydrogen-natural gas mixtures exist, needs to be considered.

The above characteristics would tend to indicate that there is a greater risk of ignition for hydrogen-natural gas mixtures. The ignition risk can be quantified for the gas system through the concept of ignition probability. The substantially lower MIE of hydrogen (15 times lower than methane) suggests that the ignition probability for a hydrogen system will be greater than that for a natural gas system.

However, these comparisons cannot be so straight forward as they appear. In many accidental situations the lower flammable limit (LFL) is more important. The LFL for hydrogen (4%) is almost similar to that of methane (5%), and about twice that of propane. In addition, the minimum ignition energy for hydrogen at the LFL is also similar to that for methane.

The light nature of hydrogen along with its higher diffusivity, differentiate hydrogen from natural gas. Hydrogen is more diffusive and more buoyant than natural gas, (methane, ethane, and propane) and therefore tends to disperse more rapidly. For low-momentum, hydrogen leaks, buoyancy may affect the gas dispersion and mixing more significantly. For high-momentum leaks, which are more likely in high-pressure systems, buoyancy effects are less significant, and the direction of the release will determine the gas motion. Localized air currents due to wind or ventilation will also affect gas movement. At low concentrations the effect of buoyancy becomes less significant because the density of the flammable gas -air mixture will be similar to that of air.

All the above considerations relate to hydrogen. However, there is no information available concerning methane–hydrogen mixture. The methane–hydrogen mixture will certainly behave differently than the individual gases, methane or hydrogen. Moreover the minimum ignition energy for the gas mixture is unknown and no correlation is available for determination of the ignition energy for various methane-hydrogen mixtures. Ignition probability, which is believed to be associated with the ignition energy, needs to be evaluated for the gas mixtures and also for the individual gases (methane and hydrogen). Hence in the work reported here, the impact of hydrogen on the ignition probability of methane–hydrogen gas mixtures is studied through experiments carried out at different ignition energy levels.

1.6 The Thesis structure

In this thesis the MIE, the lowest ignition energy at various concentrations, and the associated ignition probabilities are considered for the flammable gases under consideration. The impact of hydrogen on the ignition energy and associated probability are discussed based on experimental results from methane-hydrogen

mixtures. In particular the minimum ignition energy for various gases is identified. The thesis contents are outlined below.

Chapter 2 summaries the Literature review as carried out for the MIE, ignition energy, flammability limits and the ignition probability for the pure gases methane and hydrogen. The past data pertaining to the fire and/or explosion for the gas releases and the ignition probability models proposed for accidental gas releases are also discussed.

Chapter 3 summaries the experimental set-up designed and fabricated at Loughborough University for the measurement of the ignition energy for various gases. Detailed operating procedures and the relevant safeguards during the experimentation are also outlined.

Chapter 4 presents the results, observations and the analysis of the experimental data. The Minimum Ignition Energy (MIE) and ignition probability for all the gases studied are presented and compared with the available data in the literature.

Chapter 5 presents the conclusions of the experimental study. The experimental observations and the main findings of the study are presented for methane-air, hydrogen–air and methane–hydrogen-air mixtures. Ignition energy and the associated ignition probability for the flammable gases–air mixture are compared and discussed.

Chapter 6 utilises the experimental results to calculate ignition probability for methane and hydrogen releases from the deterministic analysis of gas releases. The analysis is based on the experimental studies carried out for natural gas releases (HSE, 1998).

Chapter 7 presents Issues for future work along with proposed modifications suggested for the experimental facility. Installation of instrument like high speed camera to the rig, will help determine flame speed, burning velocity, overpressure wave etc. giving greater insight to the burning process.

Literature Review

Ignition is a precursor to the fire or explosion process. It is of interest to researchers because of the complex nature of the ignition process and the uncertainties associated with it. These uncertainties (of ignition/non-ignition of a flammable gas–air mixture) have been observed in practice during the investigation of incidents involving gas release (fire and/or explosion event) and also during experimental attempts to ignite flammable gas–air mixtures. To determine the level of risk associated with the use of gas in domestic and industrial facilities requires consideration of the likelihood of ignition. Often historical data is analysed to arrive at the likelihood of ignition for a gas release. This section reviews available data and methods for assessing the probability of ignition of a gas-air mixture.

2.1 Ignition theory

Ignition of a combustible gas or vapour – air mixture may occur in two ways. First, the energy for ignition is supplied by a local source such as a spark or small flame at a point within the mixture as presented in Figure 2.1 (a). Second, the bulk gas mixture is heated up to its auto - ignition temperature as shown in Figure 2.1 (b).

Ignition source
(spark /
small flame)

Cloud heated to
auto-ignition
temperature

(a)   **(b)**

Figure 2.1 : Ignition of a flammable cloud (a) by spark (b) by auto-ignition

Combustion of a flammable gas-air mixture occurs, if the composition of the mixture (cloud shown above) lies in the flammable range and the conditions for ignition exist.

The flammable range is bounded by the Lower Flammability Limit (LFL) and the Upper Flammable Limit (UFL).

Ignition is a process whereby a material capable of reacting exothermally is brought to a state of rapid combustion with oxygen (usually air). At ambient temperature and pressure the mixture of flammable gas and air below auto-ignition temperature (between LFL and UFL concentrations) will not ignite unless a source of energy is provided. The source of energy could be heat, an electrical spark, another chemical reaction or just compression (pressure). The external energy breaks the molecular bonds of the fuel and oxygen, producing radicals. The fuel radical then combines with oxygen radicals releasing more energy. If this energy is sufficient to break further bonds then sustained combustion occurs. Although the minimum energy to break the bonds is known, predicting when ignition will occur under practical circumstances is difficult. Ignition has also been found to be sensitive to: temperature; fuel and oxygen concentration; volume of flammable mixture; pressure; area of contact with a heat source; time period of contact; geometry of mixture; and turbulence of the mixture.

Experiments with fuel-air mixtures give some indication of how easily a particular mixture in a particular situation will ignite. The relevant characteristics which are studied experimentally are:

- minimum volume

- minimum energy

- auto-ignition temperature

- ignition lag time

For a given mixture, at a particular temperature and pressure there will be minimum volume of flammable mixture required to sustain ignition. This is because there is a balance between the heat generated by the exothermic reaction of the specific volume of the material burnt and the heat lost which is related to the surface area of the flame. The minimum volume of the gas required to sustain ignition is where the heat generated matches the heat lost. The minimum ignition energy is that required to bring the minimum volume to a temperature that will allow combustion.

If the temperature of a flammable gas-air mixture is raised uniformly, it eventually reaches a value at which combustion occurs in the bulk gas. Across the range of flammable concentrations there is a mixture composition that has the lowest temperature at which the ignition will occur. This is called the Auto Ignition temperature as depicted in Figure 2.1(b). Even though, a flammable mixture may be raised to or above the temperature at which auto ignition occurs, ignition does not occur instantaneously; there is finite delay before ignition takes place. This time is called the ignition delay / lag time.

2.2 Historical data on ignition of gas releases

2.2.1 High pressure releases

Collated data is available from a number of sources relevant to gas escapes within the oil and gas industry. These are summarised in Table 2.1 below. Both EGIG and UKOPA data relate to high pressure transmission pipelines, whereas Offshore hydrocarbon releases, UK is a database relevant to offshore operations in the UK.

Table 2.1 : Observed ignition incidents of released gases

Data Source	Ignition incidents	Percentage of releases ignited	Reference source
UKOPA	9 out of 172 releases	5.2	Advantica, 2005 Report no. R 8099
Offshore hydrocarbon releases, UK	46 out of 1526 gas releases	3.0	Offshore hydrocarbon releases statistics and analysis, 2002 HSR 2002 002
EGIG	52 out of 1172 releases	4.4	Report of the European Gas pipeline Incident data Group, 2008 Doc.No.EGIG 08.R.0002

Accidental gas releases from high pressure pipelines (transmission lines) are ignited either because the flammable gas cloud formed by the release comes into contact with an ignition source, (sources near the location of release) or the release itself may cause the generation of an ignition source (such as metal and rocks impacting

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and generating sparks, Copper et. al., 2004). The tabulated values presented in Table 2.1, suggest that only a small proportion of high pressure gas releases get ignited (max. ~ 5%). A breakdown of these ignited incidents based on the release size, pinhole (0 to 2 cm), hole (more than 2 cm diameter hole) and rupture (< 16 inch and > 16 inch) was carried out as presented in Table 2.2. Ignition data, as analysed by other researchers, is also presented in the same table. This shows that larger releases are more likely to ignite.

Table 2.2 : Pipeline Ignition incidents based on release size

Data source	Percentage of releases ignited	
World - wide, Townsend & Fearnehough (1986)	leaks	10
	ruptures	50
US Gas, Jones(1986)	all sizes leaks	16
	ruptures	26
UKOPA (Advantica, report no. R 8099, 2005)	pinholes (0 - 2 cm)	4
	holes > 4 cm & ruptures	1.2
European Gas Pipeline Incident Data Group (2005)	pinholes / cracks	4
	holes	2
	ruptures < 16 in.	10
	ruptures > = 16 in.	33

The UKOPA data suggests the opposite observation (incidents involving pinhole releases result in more ignitions than incidents involving ruptures). This is probably due to the small number of cases reported in the database.

A review of world-wide pipeline failures carried out by Townsend & Fearnehough (1986) suggests that the ignition for natural gas leaks from pipelines is approximately 10% for 'leaks and 50% for 'ruptures'. The US data analysed for incidents of Natural gas transmission and gathering lines during the period 1970 to 1986 by Jones suggest ignitions for 'leaks' as 16% and for 'ruptures' as 26% (The data for offshore incidents shown in Table 2.1 have not been analysed for the effect of size as,

offshore releases are categorised differently to onshore pipeline releases. Moreover the sources of ignition for offshore platforms cannot be compared with the onshore ignition sources).

The variation in likelihood of ignition of pipeline releases arises as a result of the various data sources used in their derivation. The data comprises of onshore incidents from different pipe sizes, operating at different pressures, and with different leak sizes, different weather conditions, different terrain and, most importantly, variations in the distribution and type of ignition sources near the release at the time of failure.

2.2.2 Low pressure releases

On a gas distribution network, releases from low pressure pipelines may occur near buildings and result in gas ingress into the building. Similarly internal gas pipework could leak and cause a gas accumulation within a building. In both cases, ignition could lead to fire or explosion. The release rate is governed by the release size (the size of the hole) and the pipeline pressure above the ambient atmospheric pressure. Since the gas momentum is negligible (very small) the spreading and mixing are due to the nature of the gas and ambient weather conditions (and hence ventilation) prevailing at that time. A Gas In Building (GIB) event is defined (by HSE) as when the flammable gas concentration has exceeded 20% of the Lower Flammability Limit (LFL). Using data compiled by the HSE (HSE 2006) for the UK, the annual reported GIB cases and the number of fire or explosion incidents can be correlated to arrive at the percentage of GIB incidents which ignited.

Table 2.3 : Gas In Buildings events and ignition incidents for the UK

Duration	Number of Fire and Explosion (F&E) incidents	Reported GIB events	Percent GIB resulted in F&E
1990 – 1991	43	1317	3.3
1991 – 1992	50	1041	4.8
1992 – 1993	35	889	3.9
1993 – 1994	47	873	5.4
1994 – 1995	35	738	4.7
1995 – 1996	42	804	5.2
1996 – 1997	40	1165	3.4
1997 – 1998	45	973	4.6
1998 – 1999	37	923	4
1999 – 2000	56	616	9.1
2000 – 2001	38	597	6.4
2001 – 2002	43	451	9.5
2002 – 2003	33	453	7.3
2003 – 2004	34	971	3.5
2004 – 2005	37	936	4
2005 – 2006	31	954	3.2

(HSE, 2006)

The percentage of the cases which ignited is low, partly because only those escapes which produce flammable concentration can ignite, whereas the GIB data includes escapes which may produce accumulations with concentrations less than the LFL. It is also possible that no suitable ignition source was available in some cases or human intervention prevented further build-up of concentration/stopped the leak and/or increase the ventilation. This is considered in the following section.

2.3 Sources of Ignition

Stoichiometric fuel-air mixtures of gases, can be ignited with a very small amount of energy. The energy requirement is only a fraction of a milli-joule, so there are many potential sources of ignition (see section 2.3.2) for stoichiometric fuel–air mixtures of gases. Many hydrocarbons used as domestic fuels have minimum ignition energies of about 0.30 mJ and less (methane 0.29 mJ, propane 0.26 mJ, butane 0.25 mJ). These energies are extremely low when compared to sources of ignition from common activities in daily life. An ordinary spark plug has a discharge energy of 25 mJ. The human body, with a capacitance of 200 pF, can be charged with static electricity to a few kilovolts during normal routine operations, like walking across a rug, sliding across a car seat, or removing a jacket. A person feels minor discomfort with a "pin prick" sensation as a conductor is touched and the energy is discharged, but no harm is done. Such human body discharges are more than 10 mJ. (Crowl D.A and Louvar J.F. 1990). There are many other potential sources such as electrical, mechanical, frictional etc. that generate similar energies (> 10 mJ). Hence many sources routinely available are sufficiently energetic to act as an ignition source for many stoichiometric mixtures of flammable gases, sensitive dust clouds or aerosols and air (Eckhoff, 1997).

2.3.1 Common Ignition sources

There can be many potential sources for ignition of flammable gas clouds as discussed above. These can be divided into various types: heat; compression; chemical; and electrical. Ignition due to heat includes auto ignition, ignition by hot gases, hot surfaces, mechanical sparks and thermal radiation. Ignition by open flames is due to both chemistry and heat. Chemical sources include exothermic reactions and catalysts. Electrical ignition is due to electrical sparks or arcs and resistive heating of wires.

A non-exhaustive list of sources of ignition are presented below:

Naked flames, welding and cutting gear, flares, electrical sparks, electrostatic discharges, exothermic reactions, friction sparks (impact between two similar metals or dissimilar metals, such as aluminium on iron rust), lightning, hot surfaces, and internal combustion engines.

Crowl et. al (1990) have produced a study on ignition sources based on the analysis of a data bank of national incidents provided by the Health and Safety Executive. The study covers a one year period from April 1987 – March 1988 for process plants. These included a total of 968 incidents presented in Table 2.4, covering a range of fuels including solids and explosives and various offsite and onsite locations. These data of ignition sources, is for process plants fire (and/or explosion) accidents and not just for flammable gases.

Table 2.4 : Common Ignition sources for reported incidents in process plants

Ignition sources	Number of incidents	Percentage
Unknown	300	31
Flames	237	24.5
Hotwork	120	12.4
Electrical	70	7.2
Hot surfaces	48	4.9
Smoking	38	3.9
Friction	36	3.7
Spontaneous ignition	26	2.7
Autoignition	25	2.6
LPG fire equipments	24	2.5
Hot particles	20	2.1
Static electricity	19	2.0
Other	5	0.5
TOTAL	968	100

(Cox et al. 1990)

As seen from Table 2.4, the majority of the ignition sources are UNKNOWN (31%). Most of the time during accident investigation the source of ignition can not be positively identified, hence the large number in the Unknown category.

The past incidents of major fires in process plants as analysed by Crowl and Louvar (1990) for the known sources of ignition are presented in Table 2.5. The ignition sources are based on 25,000 fires recorded in the Accident Prevention Manual for Industrial Operations (1974). The sources of ignitions for past incidents are too numerous to be individuality identified, hence the list of sources considered by Crowl and Louvar are those with the greatest probability of causing ignitions.

Table 2.5 : Ignition sources for major fire incidents

Ignition sources	Percentage
Electrical (wiring of motors)	23
Smoking	18
Friction (bearings on broken parts)	10
Overheating of materials (abnormally high temperatures)	8
Hot surfaces (heat from boilers, lamps etc.)	7
Burner flames (improper use of torches)	7
Combustion sparks (sparks and embers)	5
Spontaneous ignition	4
Cutting and welding (arcs, heat etc.)	4
Exposure (fires jumping in new areas)	3
Arson	3
Mechanical sparks (grinding , crushing etc.)	2
Molten substances (hot spots)	2
Chemical reactions (runaway reactions)	1
Static sparks	1
Lightning	1
Miscellaneous	1

(Crowl and Louvar 1990)

The data in Table 2.5 is for process facility fire incidents and not specifically for flammable gases. The majority of these sources are relevant to flammable gas releases in open atmosphere, but the relative proportions of each may not be applicable.

2.3.2 Ignition sources for transmission pipeline release

In the event of gas releases from transmission pipelines, flammable gas may be present at ground level up to few metres from the release point. Ignition sources with a potential to cause ignition during such releases are discussed below. Many of the ignition sources depend on the local conditions and the extent of human activity nearby. Some ignition sources are closely associated with the cause of gas escape, for example, lightning may cause a failure and also ignite the release. (Advantica, R 8249, 2005)

- Road and rail vehicles

- Electrical sources

- Electrostatic sources

- Tools and machinery

- Lightning

- Flames and hot gases

- Impact generated sparks (rock-rock, rock-metal and metal-metal collision)

- Other sources (pyrophoric dusts, compression heating, electromagnetic radiation, catalyst, etc.)

Road and Rail vehicles

The combustion engine in the vehicles, is the most significant ignition source. Starting of the engine is an ignition hazard in any vehicle and may be a common source as a result of people escaping the gas release. Stalled petrol vehicles in a flammable gas atmosphere is a significant ignition hazard when the vehicle is restarted. In diesel engines the strongest ignition source is due to 'over-speeding' of the engine upon ingestion of the flammable gas-air mixture. Other sources include exhaust gases, hot surfaces, mechanical sparks, and vehicle electric sparks. Faulty vehicle wiring can also be an ignition source, if the faulty wiring generates a spark in

the flammable gas cloud atmosphere. Engines within excavators and heavy lifting machines are much larger than in a car and a much greater power is required to start the engine. The sparks generated from these machines are of greater energy and an engine of this kind may be in use at an excavation site on or near a pipeline (Copper et. al., 2004 and Langan, M.J. & Darby, S. 1998). Indeed the EGIG database suggest that about 50% of all transmission pipeline failures are due to third party interference, that is, by digging operations damaging pipelines. If the damage results in immediate pipeline failure, then the excavator can also provide an ignition source.

Rail transport produces similar hazards, as well as electric sparks from arcing. Moreover the mechanical sparks from the contact between the wheel and the track can also act as an ignition source. Sparks from a train was the ignition source of a large gas cloud formed by a leaking LPG pipeline near UFA, Russia in 1989 and resulted in a large number of fatalities (Mannan Sam, 2005).

Electrical sources

Street and rail electrical equipment produce sparks capable of gas ignition. These sparks have higher probability of ignition with electromechanical flashing traffic lights, when there are several bulbs (Cronin, P. and Smith, B.J., 2000). Overhead power cables can have ignition potential, if damaged by debris following a pipeline release. Security fences may also provide a high energy ignition source, if activated at the same time as a gas release. Damaged telecommunications cables in the ground near to the point of release may cause an ignition hazard (Copper et. al., 2004).

Electrostatic sources

Electrostatic discharges are capable of igniting gas-air mixtures. Discharges from clothing and the human body can act as an ignition source. It is possible that a static discharge from a person working near a pipeline may ignite a gas release. Debris (during an incident) impacting on an nearby electrically insulated object can build up static charge. Spark discharge may also occur to the pipeline or other metal objects, and so provide an ignition source for the released gas.

Tools and machinery

The ignition sources relating to tools and machinery are friction-generated sparks, hot spots, and hot surfaces. Mechanical sparks caused by friction or impact can have a high ignition potential depending on the materials involved in their production. Ignition can result from sparks (hot particles), or from high temperature (hot spots) at the point of contact. This ignition mechanism should be considered for incidents which occurred during work on an active pipeline – such as hot tapping or repairs (Copper et. al., 2004).

Lightning

Lightning is considered a likely cause of ignition, particularly if the gas release itself is resulted from a lightning strike. EGIG data suggest a 50% ignition probability from lightning, when the gas release was caused by lightning. The Ignition potential of lightning is extremely high for gas ignition but the probability of a lightning strike coinciding with a separate gas release is very small. Lightning strikes have also caused punctures (mostly pinholes) in pipelines leading to gas release without any ignition of the released material. Lightning is a potential ignition source for vent stacks on a transmission system, because of the continuous nature of the gas release.

Flames and hot gases

Open flames such as gas fired equipment, burn pits, furnaces and flares can ignite released gases with certainty. However the likelihood of such a flame being near to a gas release situation would need to be considered to calculate an ignition probability. The most common occurrence of open flames is in the pilot lights of balanced flue gas boilers, which draw in air directly from outdoors. Other, open flames include bonfires, arson and accidental fires (Spencer, et. al. 1998).

Incident generated sparks

The gas release itself can produce sparks with the potential to cause ignition, these are termed as release generated sources. Many transmission pipeline failures, (especially ruptures) have ignited despite the fact that the incidents occurred in remote rural locations, with no nearby sources of ignition (Copper et. al., 2004).

The tearing of the pipeline (during rupture) may cause sparks. Sparks can also be generated from debris impacting on nearby objects or on each other. Sparks with the potential to cause ignition can be generated from rocks impacting against other rocks or metals, and metal fragments impacting on each other. The nature of rock (Quartzitic), and the metal condition (rusted steel, aluminium, etc.) can affect the ignition potential. There are several other metals with similar sparking properties to aluminium (such as magnesium, titanium, etc.) which can result in spark generation. It can be considered that the presence of these other metals at the time of pipeline rupture is unlikely but cannot be discounted.

The high pressure in a pipeline can cause rock, soil and metal debris to be thrown from the rupture site at very high velocities. The rock and soil type, as well as the depth of cover is important in determining whether the high velocity debris has an ability to cause incendive sparks. The controlling factors in impact ignition are the materials involved, the temperature and the intensity of the sparks. These are discussed below.

For impacts of metal on metal, pyrophoric sparks are the most likely to cause ignition. Aluminium, magnesium, zirconium, titanium and cerium are the common pyrophoric metals. The incendiary nature of the pyrophoric metals arises from their reaction with atmospheric oxygen. The oxidation temperature generated in the reaction between the metal and the oxygen causes the spark temperature to rise resulting in ignition of the flammable gas.

Ignition by non-pyrophoric (carbon, tin, tungsten, zinc, platinum etc.) frictional sparks is highly unlikely because the temperatures produced by these sparks are considerably less than the pyrophoric sparks. (Langan, M.J. & Darby, S. 1998)

Aluminium based paints, when used to coat steel, and at temperatures above 150°C, easily caused ignition when struck lightly with steel. Alloys of magnesium and aluminium cause ignition on impact with steel, and this occurs most easily at methane-air equivalence ratios near 0.7. Several other materials will cause ignition with magnesium alloys, including lead oxide, silica and pyrites, and aluminium alloys will ignite on impact with lead and ammonium nitrates (Copper et. al., 2004).

An investigation of various past incidents of gas ignition relating to impact of debris (rock and metal) has been made by the HSE, 1969 (then the UK Safety in Mines Research Establishment). The HSE have studied these impacts further through laboratory scale experiments. Experiments with rock on metal, metal on metal and rock on rock, were carried out with rubbing, cutting and impact (low velocity, dropping and ballistic impact) within a flammable methane air atmosphere. The results of the investigations and experiments are presented in Table 2.6.

Table 2.6 : Summary of ignition between rocks and metal

Investigating material	Ignitions in methane air atmosphere
Sandstone with: -	
Sandstone	yes
carborundum (silicon carbide)	yes
corundum (aluminium oxide, alumina)	yes
Limestone	yes*
Shale	no
Ironstone	no
iron pyrites	yes*
Bronze	yes*
aluminium alloys	yes*
Brass	yes*
Steel with: -	
sandstone	yes*
iron pyrites	yes*
iron carbonate	yes*
carborundum (silicon carbide)	yes*
corundum (aluminium oxide, alumina)	yes*
Carborundum (silicon carbide) with: -	
carborundum (silicon carbide)	yes
Corundum (aluminium oxide) with:-	
corundum (aluminium oxide)	yes
High carbon / Tungsten steel with: -	
iron pyrites	yes*
Ironstone	yes*
quartzitic rocks	yes*
Sandstone	yes*
corundum (aluminium oxide, alumina)	yes*
Tungsten Carbide with: -	
Steel	yes
nickel and monel	yes
copper / nickel alloys	yes
Brass	no

(*Ignitions did not occur easily)

Experiments with rock - rock impact, suggested that ignition could easily occur from the rubbing of sandstone with sandstone, carborundum (silicon carbide) and corundum (aluminium oxide, alumina). Thus for a transmission pipeline passing through such soil type there can be a greater probability of a release of gas being ignited.

Other sources (pyrophoric dusts, compression heating, electromagnetic radiation, catalyst, etc.)

Pyrophoric iron sulphide scale /dusts can act as a potential ignition source for the released gas (at the point of release) if the gas stream is entrained with sulphide dust or if the dust occurs as debris in the gas. In warm, dry conditions the sulphide scale may glow red and act as a source of ignition. This situation can be of particular danger in relation to vent stack releases, where it is one of the few sources requiring consideration. As a safe practice, pyrophoric iron sulphide dust is damped down and then removed from the equipment. No attempt is made to scrape it before it has been damped. (Mannan Sam, 2005)

Adiabatic compression of flammable gas and air results in a rise in temperature. The resulting temperature rise may increase the mixture temperature to its auto-ignition range. Ignition is unlikely just because of compression heating alone, but frequent changes in pressure due to compression or shock waves can elevate the gas temperature considerably. The heated gas can then be brought to the auto-ignition temperature by other surrounding heat sources. (Mannan Sam, 2005)

Conductive materials (metal and machinery equipment pieces) in the radiation field of high frequency waves can act as aerials, generating electric currents in the material through electromagnetic induction. Sparks can be generated from these metal objects discharging to earth. Thus large military transmitters, strong radio transmitters or high frequency industrial generators can act as ignition sources. (Copper et. al. 2004)

Catalytic converters carry out the oxidation reactions (oxidation of CO and hydrocarbons) in a vehicle exhaust system. A catalytic converter can serve as an ignition source for a flammable gas, if it comes into contact with an exhaust system.

There are numerous other sources, which each have the potential to ignite a flammable gas/air mixture. The sources can be very specific to the particular situation, location or area to be addressed completely within this section. However the possibility of a gas release being ignited due to these sources cannot be ruled out. The industrial ignition database of process plant analysed by Crowl and Louvar, 1990 (presented in Table 2.4 and Table 2.5) includes a few of these other sources.

2.3.3 Ignition sources for Low pressure release

If the low pressure releases are outdoors, then many of the ignition sources discussed for transmission pipelines are still relevant. Low pressure releases from internal gas pipe work (carcass pipeline) needs consideration of indoor sources. Similar releases from distribution and service pipes may track through the ground and result in an accumulation within a building.

Indoor sources are also too numerous to be considered completely in the present analysis, but the most common are discussed. Ignition sources inside residential houses, includes pilot lights, cookers, doorbells, clothes dryers, light switches, electrical tools, hair dryers, toasters, boilers (central and water heating) and other electrical appliances. The sources can be continuous or intermittent and may require human intervention (smoking, phones, light switching etc.). or automatic (refrigerator, freezer, thermostats etc.) The sources are grouped under the following subsections and discussed below:

- Pilot light / lamp

- Smoking

- Electrical appliances

- Gas boilers

- Static electricity

- Telephones / mobiles

- Cables

Pilot light / lamp

A pilot light/lamp is certain to cause ignition, if the gas release produces an accumulation, which can come into contact with the flame (Spencer et. al., 1998). Some appliances may contain a continuous pilot light whilst others are intermittent. Gas fired room heating is considered in this category. A cooker having an open flame is also considered in this category. Cooking equipment was the number one ignition source for home fires (during the years 1995 to 2001) as per the Ontario office of the Fire Marshal. (www.ofm.gov.on.ca). A leaking gas ingress inside a room/house may increase the gas concentration to a flammable level inside a confined space. A person subjected to the smell of gas for a prolonged period (especially an elderly person) can become desensitised to the odorant and be unaware of the presence of the gas accumulation. Operation of a cooker or other open fire sources under such circumstances may lead to major fire and/or explosion in domestic properties.

Smoking

Smoking materials are one of the most common potential ignition sources. There are two areas where smoking can pose an ignition threat, these being during the lighting of the material, and the material when lit. Smouldering tobacco in any form is an ignition source with a very low potential for a gas release. Tests by Jeffreys et al (1982) demonstrated that the ignition source related to smoking is, in almost all instances, due only to lighters or matches, and that burning cigarettes produce a negligible risk of ignition. Jeffrey's studies suggested that a lit cigarette cannot ignite flammable gas, even during inhalation. Matches and lighters are open flames for the duration of lighting the cigarette and at that time, have an ignition potential of unity. The probability of a person deliberately lighting a match in a flammable gas atmosphere is very slim, but cannot be totally ruled out as discussed in the paragraph above. An electrical cigarette lighter (similar to a car cigarette lighter) is not an ignition source.

Electrical appliances

Ignition can be caused by electrical appliances being switched on or off by human intervention or the switching can be automatic (Swaffield F., 1999). It is the transmission of charge through a circuit which produces the spark. The possibility of

gas ignition from the operation of electrical equipment depends upon the type of circuit (resistive, inductive and capacitive) involved and the electrical power input.

- For a resistive load, the rate of energy generation is $E = I^2R$ joules/s (where I is the current in ampere and R is the resistance in ohm). This energy will heat up the conducting material and the ambient atmosphere and thus ignition can be caused by the hot surface. Ignition would require a long time period of the hot wire in the presence of a flammable gas mixture, and the quantities which determine the likelihood of ignition from this source are the total heating time, the total energy flow (defining the current strength) and the core heating temperature.

- For an inductive load, a low voltage inductive break spark energy is $E = 0.5LI^2$, joules (where L is the inductance of the circuit in henry and I is the current in ampere). The energy can be compared with the minimum ignition energy of the flammable gas, to result in ignition.

- For a capacitive load, the spark energy is $E = 0.5CP^*V^2$ joules, and discussed in detail in Chapter 3, since the experimental apparatus used in the experimental work described in this thesis is based on this principle.

Sparks may be caused by faults in a damaged electrical circuit, or occur in normally working circuits containing components such as switches or relays. Sparks are likely to occur with mains switches, whereas arcing will occur with dirty contacts or damaged circuits.

Testing was undertaken by Advantica (Swaffield F., 1999 and Johnson D.M. & Wright S.J., 1989), on various kinds of electrical equipment to determine their ability to ignite a natural gas-air mixture. With some pieces of equipment, the electrical circuits within them were broken down, so that the individual components most likely to cause an incendive spark were tested. This was done at the higher end of the range of voltage/current ratings used in that equipment.

Theoretically, both mains operated and battery operated equipment can produce sparks of sufficient energy to be intrinsically hazardous, but testing suggested most

devices were unable to cause ignition of a flammable gas air mixture (within the flammability limits). The reason is, the spark locations are normally sealed from the atmosphere preventing any ingress of the flammable gas-air mixture. Even if the gas penetrates the seal, to the ignition source, there may not be a route out through which the flame could propagate to the whole cloud.

Devices such as vacuum cleaners, electrical drills, doorbells, automatic door entry system and most motors (inductive loads) are capable of ignition since the spark energy is greater than the minimum ignition energy of gas. Ignition of a gas cloud is plausible, if the casing, or the circuitry itself, is damaged, although no records have been found for testing of these items.

Automatic operation devices, (where mains power is being switched on and off, without human intervention) like circulation pumps, thermostats, timer switches, Video recorders, refrigerators etc. have sufficient energy to act as ignition sources. Inside buildings, previous incident ignitions have been attributed to thermostats, timers and refrigerators or freezers with automatic switches.

Experiments with a refrigerator (Swaffield F., 1999) resulted in ignition within a small number of cycles, and confirmed the findings of several explosion incident investigations that found that the only likely electrical source of ignition was a refrigerator. Newer designs are less likely to be a source, as the electrical elements are encapsulated, whereas older designs may have open elements.

Burglar alarms and smoke alarms produce a very weak spark and will not result in ignition because of the small current (Swaffield F., 1999). Standard non rechargeable battery systems do not result in ignition even with a worst-case direct short-circuit across the terminals on the highest common operating voltage.

Static electricity

Static electricity is the build up of electrical charge that is unable to flow to earth. Static electricity has been attributed as the cause for many explosions where no

other cause was apparent. Static electricity discharges commonly occur from the body and from clothing. The discharges are generally very weak, and do not cause discomfort to human beings. Any form of rubbing activity or friction may charge the human body or the tools and equipments used for pipe work. A charged body/object has the potential to cause a spark discharge resulting in ignition (Copper et. al., 2004). The energy of a normal human body spark is much stronger than the minimum energy level of 0.29 mJ required for the ignition of a methane gas-air mixture. Typical examples of household activities that result in a build-up of a static charge are walking across a rug, placing different materials in a tumble dryer, removing a sweater and combing hair. Clinging fabric, and sometimes audible sparks are the result of the build-up of a static charge. Fabrics are also capable of producing spark discharges with a likely potential for ignition under conditions of low humidity. At higher humidity the discharge is unlikely since clothing will usually contain enough moisture to be sufficiently conducting so as to prevent static accumulation.

Gas boiler

A boiler containing a strong ignition system is used for central/water heating in most households with mains gas. Gas boilers use ambient air, and if there is ingress of a gas - air mixture into the appliance, ignition is certain. Depending on the path of the burning flame and the presence of a flame arrestor, etc., boilers can act as an ignition source for the flammable releases (Spencer, et. al. 2004).

Telephones

Telephones, both mobile and fixed, are one of the most widespread pieces of electrical equipment, and in the case of a gas leaks (smell), are often used by people calling for help. Use of mobile telephones is forbidden at petrol/gas stations across the world, because its widely believed that these can be a source of ignition. However testing has shown that ignition from telephones in any kind of operation is highly unlikely. Experiments with mobile phones, in the worst-case of direct short circuit across the battery terminals have never resulted in ignition. (Johnson D.M. and Wright S.J., 1989).

Cables

Damaged or faulty wiring can cause ignition of released gas. Increased electrical load on a cable can also heat up the cable resulting in damage to the cable and hence provide a potential source of ignition. Short-circuiting could also be an ignition source for a gas release (Copper et. al., 2004).

Ignition sources discussed above were further analysed for ranking, based on experimental studies, observations and engineering judgement. Ranking can be done using the characterisation of ignition sources described in the next section.

2.3.4 Analysis of Ignition sources

Ignition sources can be analysed further for the certainty with which the ignition of a flammable gas is possible by that source. Britton (1992) provides an example of the ranking of ignition sources based on the consideration of their available energy in relation to the minimum ignition energy required for various flammable gas or dust clouds. During ignition all likely ignition sources are initially considered, which can deliver energies greater than the minimum required for the ignition of a flammable gas. A list of energies of various types of sources and the kind of flammable gas/ dust air mixture which could be ignited at that energy level is shown in Table 2.7.

Table 2.7 : Energies for various ignition sources

Source	Energy (mJ)	Typical substance which can be ignited
flames, chemical sources, large hot spots	100 – 1000	Methylene chloride, Ammonia
human body spark, bulking brush limit (discharge during powder filling - silo filling)	10 – 100	Lycopodium
brush discharge (single electrode discharge with no sharp tip - in a strong electric field)	1 – 10	Acetone
mechanical sparks, stray current sparks, small hot spots	0.1 – 1	Methane, methanol
discharges from textiles, weak inductive coupling, weak radio-frequency pick –up	0.01 – 0.1	Ethylene, hydrogen

(Britton 1992)

A semi – qualitative approach was used by Jeffreys et. al (1982) to rank the sources in terms of ignition potential. The ignition potential of strong, medium and weak were defined as described below in Table 2.8. The ignition potential is based on the experiments carried out for a 7% methane-air mixture. The ignition sources, such as pilot lights, gas fired heaters, open flames etc. warrants a further category of 'certain', with an ignition potential of unity. Experimental studies can be further used to define a category for those items with 'negligible' ignition potential like, radio frequency waves. A few sources along with their ignition potential are presented in Table 2.8.

Table 2.8 : Ignition potential for various sources

Source	Ignition potential
Pilot lights / gas fired heaters	1
Cigarette lighter (open flame)	1
Switches (electrical)	S
Electrical tools	S
Hair dryers	M
Toasters	M
Televisions	W
Electrical Appliances (washing m/c, iron, bulbs etc)	W
Smouldering /lit cigarette	0
Domestic and mobile phones	0

S = strong ignition potential with probability of ignition greater than 0.5

M = Medium ignition potential with probability of ignition between 0.05 and 0.5

W = Weak ignition potential with probability of ignition less than 0.05

2.4 Ignition model for gas releases

The strength of the ignition source (its delivered energy), spatial distribution of the sources and the operation (continuous or intermittent) of the source have major impact on the likelihood of ignition of the released gas. Accordingly, ignition models for gas release developed by various researchers consider these parameters. Ignition likelihood of the released gas, often referred to as ignition probability in the literature is now described in the subsequent sections.

Expert judgement over the years has produced estimates for gas ignitions (ignition probability) based on a single value or a few bands as presented in Table 2.9.

Table 2.9 : Estimates for Ignition probability by various researchers

Source	Type of release	Leak quantity / size of release	Location with/ Area	Ignition Probability
Browning (1969)	LPG	massive	'no' sources of ignition	0.1
Kletz (1977)	LPG	10 ton or more	Onsite area	0.5
	Polyethylene VCE	Small	plant area	1 in 10,000
	Hydrogen and hydrocarbons mix (hot, @ 250bar)	small to major	plant area	1 in 30
HSE (1981)	LNG vapour clouds	general	'no' sources of ignition	0.1
			'very few' sources of ignition	0.2
			'few' sources of ignition	0.5
			'many' sources of ignition	0.9
Blything & Reeves (1988)	LPG	Large (gas dispersion distance to LFL conc. is 60 m)	Onsite area	0.7

There is wide variation in the values of ignition probabilities given by the various researchers tabulated above. Many of the values are case specific and may not be directly applicable to general studies. A review was carried out for the proposed ignition likelihood/ignition probability models in the open literature. The correlations/models and ignition probability estimates discussed in subsequent sections are

- Cox, Lees and Ang model

- PIPESAFE - Ignition probability model for transmission pipeline

- WS Atkins model

- Ignition probability estimates for low pressure releases

2.4.1 Cox, Lees and Ang model

This model (Cox A.W. Lees F.P. and Ang. M.L. 1990) is based on the historical data for leak frequencies and fire and explosion frequencies. The approach is to make an estimate of the frequency of leaks, with the standard plant equipment inventory. The model has a simple relationship between the ignition probability and the release rate. Cox et. al. (1990) reviewed a number of estimates of the probability of ignition of various flammable releases, based on incident data as presented in Table 2.10.

Table 2.10 : Ignition probability estimates for flammable releases

Flammable gas release rate	Ignition Probability
minor (< 1 kg/s)	0.01
major (1 – 50 kg/s)	0.07
massive (> 50 kg/s)	0.3

Cox further assumed that the probability of ignition is proportional to a power of the released gas mass flow rate. The correlation for the probability of ignition (IP) based on the flammable releases analysed by Cox et al (1990) is

$$IP = a\ m^b \qquad\qquad ...(2.1)$$

Where   m is the mass flow rate of leaking gas in (kg/s)

            a = 0.017 and b = 0.74

Cox also proposed different coefficients for equation (2.1) above, when there is some control on ignition sources (a = 0.006, b = 0.77), no control on ignition sources (a = 0.074, b = 0.57) and also for self ignition (a = 0.003, b = 0.28).

The model is not intended for very high mass release rates, where it gives a probability of greater than 1. The constant of proportionality and the power are estimated from the observed historical data of ignition probability and the released mass of the flammable gas under consideration. As seen the model is very general

and does not consider individual ignition source characteristics contributing to the ignition process.

2.4.2 PIPESAFE - Ignition probability model for transmission pipeline

Advantica have produced an ignition probability model that is incorporated into their PIPESAFE package (Robinson, C. & Ronnie, S., 2005 and Advantica R 8224, 2005). The model is intended for transmission pipelines only and is based on the historical experience of pipelines operations. A list of 68 transmission pipeline incidents has been analysed by Advantica. Amongst the analysed incidents, where cause of failure was reasonably ascertained; 15 were caused by external interference, 18 by a defect in construction or repair, and 13 were due to corrosion, ground movement or other natural causes. A further analysis based on the ignition delay time (time between the gas release incident and the actual ignition event), suggested that the majority of the releases ignited immediately (within one minute). The maximum time from the start of the gas release to ignition recorded in this data was 90 minutes.

The model proposes a relationship of ignition probability values based on the pipeline diameter and pressure. Considering the complexity of the processes involved in pipeline release incidents, there are numerous other parameters affecting the ignition probability, but the data analysed for rupture incidents by Advantica suggest that ignition probability increases with line pressure and pipe diameter. Accordingly the equation utilised for calculation of ignition probability for ruptures in PIPESAFE package is

$$IP \text{ (ruptures)} = pd^2 \qquad \qquad \dots(2.2)$$

Where p is the pipeline operating gauge pressure (bar)

     d is the pipeline diameter (metre)

The relationship incorporated in the PIPESAFE package is used up to a pd^2 value of 48.2 bar m^2; above this value a limiting ignition probability of 0.8 is used.

The same approach is used for puncture releases, except that the hole size is used instead of pipeline diameter. The pd^2 value is halved since there is single source for a puncture release. Unlike the two sources contributing to a gas release from a rupture event. The equation utilised for puncture releases in the package is

IP (punctures) = $0.5*pd_h^2$ ...(2.3)

Where p is the pipeline operating gauge pressure (bar)

       d_h is the puncture hole diameter (metre)

2.4.3 WS Atkins model

The WS Atkins model for determination of ignition probability for a drifting gas and/or growing flammable gas cloud was developed by Spencer H and Rew P.J. 1997 on behalf of the Health and Safety Executive. (HSE CRR 146 / 1997).

The HSE report reviews ignition data and models for use in onshore risk assessment. The model takes account of the likely ignition sources in the area and the duration that the gas/vapour cloud, within flammable limits, interacts with these sources. Each source is characterised with an ignition potential defined as the certainty with which the source can ignite the flammable gas in an open atmosphere. Further, the number of each type of ignition source per unit area, and its rate of activity (continuous or intermittent) is also considered in the model.

The parameters that are used to define each ignition source in the model are summarised below:

- Different ignition sources were identified and the density per unit area (μ), of each source is determined

- The probability of ignition or ignition potential (po), is the basic probability that ignition will occur when the source is active and in contact with the gas, i.e. source is surrounded by a flammable gas cloud. The ignition potential (po) value has to be defined for each ignition source

- Ignition sources as per their operation, intermittent or continuous, are then considered in the analysis. The period for which the source is 'active', t_a, and the period between each activation, t_i, is specified. From these t_a, and t_i, values the frequency, λ, at which the source is able to ignite the gas, and the probability of the source being active, a, is estimated

$$a = \frac{t_a}{t_a + t_i} \ , \quad \lambda = \frac{1}{t_a + t_i}$$

- Intermittent sources are considered as a special type of the generalised source with $t_a = 0$, and thus $a = 0$. Continuous sources are a special case with $t_i = 0$, and thus $a = 1$ and $\lambda = 0$

Table 2.11 : Source parameters for the WS Atkins model

Source parameter	Intermittent sources	Continuous sources
Time period for which the source is active (t_a,) min	$t_a = 0$	t_a = continuous
Time period between each activation of source (t_i,) min	t_i = finite	$t_i = 0$
Probability of the source being active (a)	$a = 0$	$a = 1$
Frequency of activation of the source (able to ignite the gas) λ per min	λ = finite	$\lambda = 0$

In reality the sources can never be completely Continuous or Intermittent, hence all real World sources have some finite t_a, t_i, a and λ values. The overall ignition probability after time 't' is calculated based on the following summation of the probability

$$IP(t) = 1 - Q(t) \qquad \qquad ...(2.4)$$

and

$$Q(t) = \sum_{j=1}^{J} \exp\left\{A * \mu_j \left[(1 - a_j po_j) e^{-\lambda_j \, po_j \, t} - 1\right]\right\} \qquad ...(2.5)$$

Where:

IP(t)   overall probability of ignition from various sources at time t

$Q(t)$ probability of non-ignition at time t

A the area of the region being in contact with the gas cloud

j   represents the 'j' th. ignition source (when 'j = 1, 2, 3, ... to J' nos. of sources)

μ_j ignition sources density per unit area of source 'j'

a_j probability of the source 'j' being active

λ_j frequency of activation of the source 'j' (per min.)

po_j ignition potential (probability of ignition) of single source 'j' in consideration

t   the time duration the source has been in contact with the gas cloud (min.)

Simple correlations for the probability of ignition in terms of the cloud area for different conditions (day, night, industrial, urban, rural etc.) can be developed using the model.

The WS Atkins model presents an area based method for the ignition of flammable gas clouds for offsite areas. The method does not include a model for the dispersion of the gas. Separate applications of the method are needed for each vapour cloud scenario/ wind direction etc. The ignition source type, potential and its distribution is difficult to estimate because of numerous ignition sources.

2.4.4 Ignition probability estimate for low pressure releases

Although originally developed to study large outdoor vapour clouds, the WS Atkins model was utilised for estimating the ignition probability within a residential area

(house/room). Low pressure releases could occur from the internal pipe work (carcass pipe), gas appliances or any external gas ingress into the building. It is assumed that the gas leak has caused enough gas accumulation to exceed the LFL concentration inside the building. Thus the probability of ignition is computed given that an ignition source is in contact with the flammable gas cloud within the building. A typical list of ten ignition sources with their source characteristics (as described in the Atkins model) are presented in Table 2.12. The ignition sources include pilot light (gas fire, candle, cooker etc.), gas lighter, electrical switch, fridge/freezer, gas boiler, Microwave, Washing m/c, TV set, and Room heater. Some of the sources are human operated and others are automatic as per the instrument settings. The values of ignition potential (certainty with which the source can ignite the flammable gas) is assumed based on engineering judgement.

Table 2.12 : Typical Ignition sources within homes

Sources	No.s	Source characteristic parameters				
		t_a	$t_{i,}$	λ x 1000	a	po
		min	min	per min	-	-
Pilot light (candle, cooker, etc.)	1	5	595	1.67	0.01	1
gas lighter / cigarette lighter	1	1	479	2.08	0.002	1
gas boiler	1	10	110	8.33	0.08	1
Electrical switch	5	0	180	5.56	0	0.5
Electric room heater	1	30	570	1.67	0.09	0.2
Toaster	1	3	717	1.39	0.004	0.2
Fridge - freezer	1	20	100	8.33	0.13	0.05
Microwave	1	6	294	3.33	0.02	0.05
Washing m/c	1	60	1380	0.69	0.04	0.05
TV set	1	180	600	1.28	0.23	0.05

The probability of ignition based on the activation of sources as presented above is computed from Equation 2.5 presented by the WS Atkins model. The only modification to the equation is instead of area (A) and the source density (μ) the total number of similar ignition sources (N_s) is used as follows.

$$N_s = A * \mu$$

$$IP(t) = 1 - \sum_{j=1}^{J} \exp\left\{ N_s^j \left[(1 - a_j po_j) e^{-\lambda_j po_j t} - 1 \right] \right\} \qquad \text{...(2.6)}$$

Where:

$IP(t)$ Overall probability of ignition from various sources at time t

N_s^j Nos. of similar 'j' th. sources (i.e. electrical switches = 5 nos.)

a_j probability of the source 'j' being active

λ_j frequency of activation of the source 'j' (per min.)

po_j ignition potential (probability of ignition) of single source 'j' in consideration

t the time duration the source is in contact with gas cloud (min.)

The ignition probability under the activation of ignition sources was estimated at various time durations (t) and the variation as a function of time is presented in Figure 2.2.

Figure 2.2 : Ignition probability variation with time for gas leak inside house

As observed the ignition probability increases with time and attends a value of unity after 300 min (5 hrs.) i.e. Ignition is almost certain after 5 hrs. The computed ignition probability is based on activation of the 10 ignition sources and at the rates assumed in Table 2.12.

2.4.5 Model limitation

The ignition energy of a flammable gas (methane, propane, butane, hydrogen, etc.) and air mixture varies with its concentration. Therefore it is expected that the ignition potential (as defined in the WS Atkins model) should also vary with concentration. There is very limited information on the individual gas - air mixture ignition energies at various concentrations covering the entire flammability limits. Most of the gas ignition information gives the minimum ignition energy (without associated probability) at stoichiometric or near stoichiometric concentrations. Also, due to local fluctuations in concentration within a flammable gas cloud or plume, an ignition source within a cloud may experience a range of concentrations, hence the ignition potential (po) of each source will vary with position within the flammable region. The ignition potential of individual sources is the key for ignition of the gas cloud. The ignition probability thus will vary greatly with variation in ignition potential of each

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source. Hence experiments to determine the ignition energy (and associated probability values) were planned within this project not only for the stoichiometric air-gas mixture but also over the entire flammable ranges of methane (5% to 15%), hydrogen (4% to 75%) and methane-hydrogen mixtures. The ignition of flammable gas-air mixtures is a function of the minimum ignition energy of the gas. Hence the issues considered in the next section is the MIE values for the gases methane and hydrogen.

2.5 The Minimum Ignition Energy of gases

For a flammable gas (within its flammability limits) to be ignited the most important criteria which must be met is the requirement of the minimum energy to start and sustain the combustion of the gas. It therefore is reasonable to describe the ignitability of the mixture in terms of the limiting value of the energy, which just ignites the mixture. The energy liberated by the source must be sufficiently large to stimulate a propagating flame reaction into the gas volume, in spite of the energy losses to its surroundings (unburnt gases).

The energy requirements are different for different gases and it is also different for the same gas if the gas concentration is different. For a given gas, if the composition of the gas-air mixture is varied, the curve of the limiting energy necessary for ignition exhibits a distinct minimum. This minimum value is designated the Minimum Ignition Energy (MIE). Gases often exhibit their MIE very close to the stiochometric concentration. Hence MIE for most of the gases are reported at or near the stiochometric concentration. The energy required for ignition of gases is progressively larger on either side of the concentration at which the ignition energy is a minimum until the Lower and Upper Flammability Limits are reached. (Lewis & Von Elbe 1987 and Esseghir M. & Polymeropoulos C.E., 1988). The requirement of ignition energy becomes infinitely large at the limiting concentration (LFL or UFL) of gases.

The energy delivered by a source and the energy received by the gas mixture during the ignition process can be different quantities. Hence the MIE is defined as the minimum value of the energy (stored energy delivered by the source), which on discharge, just ignites the mixture of quiescent gas (gas at rest) in the most ignitable

composition. MIE is an extremely important property for safety standards as well as for the fundamental understanding of the ignition process of combustible mixtures. Data on MIE's are necessary for fuel ignition systems and safety standards in relation to possible explosion hazards in fuel handling and in industrial situations.

MIE's for most of the flammable hydrocarbon gases are very small (0.3 mJ and less). The measurement of such a small magnitude of energy is difficult. Spark, Laser and energy delivered by hot wire (by passing electric current through wire) are a few of the techniques used to provide such a low magnitude of energy (Martin Hattwing, 2004 and Lewis 1987). A few experiments of igniting flammable gases (methane, hydrogen etc.) in air have been carried out using frictional sparks from light alloys (Komai et. al., 1994). The aim of these experiments was to examine the incendivity of the sparks and to understand the safety aspect of handling flammable and explosive gases. Spark ignition through electrical discharge has been the preferred and standard method for determining the MIE, ASTM E 582 1988.

2.5.1 MIE determination

The electrical discharge used during an experiment can be measured to estimate the quantum of energy delivered. Since the amount of energy delivered by these sources is often very small, measurement of these energies is only possible with advances in electrical measurement devices (Huang et. al., 2007 and Ono et. al 2007). The electrical devices either measure the voltage or the amount of total charge transferred during the discharge to estimate the energy delivered by the specified spark process.

Electrical spark discharges can be deliberately generated in a flammable mixture and the various parameters measured to arrive at the energy discharged and thus enable the MIE to be estimated. The standard way to characterize capacitive discharge sparks is in terms of the quantity of stored electrical energy, measured in (mJ). The actual amount of energy that is deposited in the gas by the discharge is lower and is some fraction of the stored electrical energy measured through electrical devices.

Beginning with the work of the US Bureau of Mines in the 1940s and continued by Blanc et al. (1947), an extensive series of spark tests (Lewis and von Elbe, 1961) were carried out to determine the ignition energy in hydrocarbon-air vapours. Extensive experiments were carried using methane, the major constituent of natural gas. Ignition energy was found to be a function of the concentration of methane in the methane–air mixture. The ignition energy was found to be a U-shaped function of concentration with the vertical portions of the "U" occurring at the flammability limits and the bottom of the "U" at some intermediate composition. The ignition energy for other hydrocarbon fuels also follows a similar trend to that of methane gas. The U.S. Bureau of Mines tests were aimed at understanding the ignition characteristics of methane and its energy. The objective was to prevent any chance of methane explosion occurring due to sparks generated during the cutting of coal with mining equipment.

The experimental apparatus for the determination of the MIE essentially consisted of electrodes with a spark gap, a charging capacitor, a charging resistor and a high voltage source. The spark was produced in a vessel filled with a known concentration of gas. Once the gas mixture was prepared inside the vessel, the voltage was slowly raised across the electrode gap through the charging resistor. At the breakdown of the dielectric field strength of the gases between the electrode tips, a spark is observed. Theoretically, a spark discharge in a gas occurs when the free electrons that are present in gases are accelerated in an electric field to such a high kinetic energy that the gas molecule is ionised in the collision. (Martin Glor / Martin Hattwing, 2004). After this ionisation an avalanche of ions or electrons is created, which then depending on the field strength and, in accordance with the laws of physics, leads to a gas discharge. The dielectric field strength depends on the type of gas and the gas pressure. The dielectric field strength increases proportionally with the gas pressure, starting from the initial value of about 3 kV/mm at normal ambient pressure.

Lewis and von Elbe 1987, (including Blanc, 1947 and the US Bureau of Mines, 1940) have used similar experimental setups for spark ignition studies. The apparatus was designed to produce sparks with a capacitance of 100 pF or more. The gases were admitted to a test bomb which was made of stainless steel and had an inside diameter of 5 inches. The spark electrodes were mounted in the centre of the bomb. The electrodes were adjusted through a micrometer to precisely measure the spark

gap distance. The electrodes were connected to a system of fixed and variable condensers and a variable range calibrated voltmeter. The electrical circuit was designed to minimise the electrical resistance. The aggregate capacitance was varied between about 100 pF to 5000 pF. The exact value of the capacitance was measured through a Wein bridge. A high voltage was supplied via a resistor to the electrode – capacitor system. A rotary charger with a small metal sphere mounted on either end of hard rubber alternately touched the terminals of the power source and the spark electrode transferring charge during each contact of the metal spheres to the electrodes. Thus the spark circuit was effectively isolated from the power source. The voltage at which the spark occurred was observed and the actual capacitance in the circuit was noted. If the mixture did not ignite, the capacitance was increased and the experiment repeated. The value of voltage and capacitance at the ignition threshold was used to calculate the ignition energy as $E = 0.5*CP*V^2$ (where capacitance, CP (F) and Voltage, V (V)). The lowest ignition energy value obtained through this setup at the most ignitable composition was reported as the MIE. Similar setups have been used by various researchers to arrive at the MIE of various gases.

2.5.2 MIE and quenching distance

Lewis and von Elbe 1987 have carried out experiments with glass flanged electrodes. The spark electrodes during the experiments were tipped metal rods, and in addition, were also flanged by glass plates. Glass flanges were used so that the spark occurs at the centre (between metal rods). Several series of experiments were carried out in which the composition, pressure and temperature of the flammable gases were held constant and the length of the spark gap was varied with a micrometer assembly. Lewis observed that the minimum ignition energy with glass flanged electrodes takes a sharp vertical turn (increases very rapidly) when the distance between electrodes is reduced below a certain critical value. The glass flanges have the effect of suppressing the ignition (initiated at the centre by metal electrodes) altogether, when the electrodes were within a critical distance. This critical distance was named the quenching distance. The quenching distance was observed to be independent of the mode of ignition. The same or nearly the same value of quenching distance was observed from experiments in which the flammable gas was enclosed in a rectangular channel bounded by two plane parallel plates, and ignited at one point by a pilot flame. The materials used for the walls also did not have any effect on the

quenching distance. Glass and metals were equally effective as heat sinks, even though the heat conductivity of metals far exceeds that of glass. The reason being the heat conductivity of solids exceeds heat conductivity of gases by an order of magnitude. The use of flanges made from an electric non-conductor (glass) assured that the sparks remained centred between the electrodes. A plot of minimum ignition energy and the electrode (flanged electrodes) distance, showed a distinct minimum before a very steep rise in minimum ignition energy at very small electrode distances. At electrode distances larger than the quenching distance the size and shape of the electrodes did not affect the value of the minimum ignition energy. (Lewis and von Elbe 1987). At the bottom of this minimum ignition energy versus electrode distance curve, the energy values depend primarily on the mixture variables, notably the composition and the pressure. The quenching distance at the stoichiometric concentration along with the MIE values of methane and hydrogen is presented in Table 2.13.

Table 2.13 : MIE and quenching distance for flammable gases

Gases	Gas concentration (% by vol.)	MIE (mJ)	Quenching distance (mm)
Methane	9.5%	0.33	2.16
Hydrogen	30%	0.019	0.64

The above values suggest that there may be a relationship between the minimum ignition energy and the quenching distance. Kondo et. al 2003 has suggested two theoretical expressions for calculating the minimum ignition energy of gases from the quenching distance; one is based on the amount of energy that the minimal incipient flame contains, and the other is based on the heat loss from the surface of the minimal flame.

In the first equation Kondo et. al 2003 assumed a certain minimum energy at a spot in time is required to ignite a flammable gas mixture. The amount of energy was equated with that required to increase a certain minimal sphere of the mixture to the flame temperature. The diameter of the sphere was approximated to be the quenching distance and the following equation was obtained.

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$$\text{MIE} = \frac{1}{6}\pi \, qd^3 \, \rho_b \, \text{SPH}_{av}(T_b - T_u) \qquad \qquad \ldots(2.7)$$

where,

qd quenching distance (cm)

ρ_b molar density (mol/cm^3) of the burnt gas at the temperature T_b

T_b flame temperature (K)

T_u unburnt gas temperature (room temperature).

SPH_{av} represents the molar heat capacity (J/mol/K) at constant pressure averaged for the temperature range T_u to T_b

$\text{SPH}_{av}(T_b - T_u)$ represents an amount of enthalpy required to heat up one mole of gas from T_u to T_b

In the second equation, Kondo et. al 2003 assumed, the outward growth of flame depended on the balance between the heat of reaction and the heat loss from the surface of the sphere. The minimum ignition energy was estimated from the heat losses from the surface of the sphere, the average laminar burning velocity, and the temperatures T_u and T_b as follows

$$\text{MIE} = \pi \, qd^2 \frac{\lambda_{av}(T_b - T_u)}{S_{av}} \qquad \qquad \ldots(2.8)$$

where,

qd quenching distance (cm)

λ_{av} heat conductivity (W/cm/K) averaged for the temperature range T_u to T_b

T_b flame temperature (K)

T_u unburnt gas temperature (room temperature).

S_{av} laminar burning velocity (cm/s) averaged for temperature range T_u to T_b

Since there is a large temperature difference between both sides of the flame front T_u and T_b, the value of heat conductivity (λ_{av}) was averaged by Kondo et. al, for the temperature range. On the other hand, laminar burning velocity (S_{av}) was used to derive the time for the gas to pass through the width of flame front to complete the combustion reaction. The averaged value for the temperature range between T_u to T_b is also used here. The data for the quenching distance and burning velocity at

room temperature used for the calculation as well as the data of minimum ignition energy was taken from NACA (now NASA) report 1957. The flame temperature was calculated using the adiabatic flame temperatures for gases. The heat capacity and coefficient of heat conductivity were from the thermo-physical properties of gases and used to obtain equations to estimate approximate values at higher temperatures.

MIE as estimated by Kondo et. al 2003 through the proposed equations are presented in Table 2.14, against the actual values of the MIE as observed through experiments.

Table 2.14 : Theoretical estimates of MIE for gases

Gases	Gas conc. (% by vol.)	MIE observed (experiments) mJ	MIE calculated (eq. 2.7) mJ	MIE calculated (eq. 2.8) mJ
Methane	9.5%	0.33	3.33	2.18
Hydrogen	30 %	0.019	0.04	0.04

The expressions (equations 2.7 and 2.8) of minimum ignition energy calculation by Kondo et. al, 2003 from the quenching distance do not compare well with the actual MIE values as observed through experiments especially for methane. This suggests that the burning process is so complex that simple theories for the prediction of ignition energy can not quantify the MIE. Hence researchers have relied on experimental values of MIE for flammable gases.

2.5.3 Historical MIE data

A literature search has been undertaken on the reported minimum ignition energies (MIE) of the flammable gases under consideration, that is, methane and hydrogen. Since the majority of accidental gas releases and subsequent ignition happen in the open atmosphere, the literature search was restricted to MIE values reported under ambient conditions of temperature and pressure and for quiescent mixtures.

Table 2.15, lists the MIE data as found in reports, standards and as given by various researchers that focused on the subject of fire and explosion. The reported values are based on the ignitions carried out using electrical sparks generated across an electrode gap charged to a high voltage. The stored capacitor energy at the time of the spark is considered as the MIE for the gas-air mixture. The experimentally observed values are at the stoichiometric fuel-air ratio, which can be, practically, considered as the minimum value of ignition energy.

Table 2.15: Minimum spark ignition energy data from various sources

Reference source	MIE (CH_4) mJ	MIE (H_2) mJ
Toriyama, Y. and Saito, S., 1942	0.67 – 0.58	-
Calcote et. al., 1952	0.47	0.028
NACA report, 1957 (now NASA)	0.33	0.02
US Bureau of Mines explosive research report, 1966	0.30	0.017
Sayers et. al., 1970	0.55	-
Moorhouse, J., 1974	0.63 *	-
Lewis and von Elbe, 1987	0.29	0.02
Lees F.P., 1996	0.29	0.019
Pratt T.H., 1997	0.21	0.016

* Ignition probability of 80 % is assigned to this energy

The historical MIE data, were determined using measured values of Capacitance and Voltage at the time of the spark. Also, the technology of electrical measurement has vastly improved over the period, hence the reduction in the MIE values for more recent studies as shown in Table 2.15. The criteria of MIE by Moorhouse, 1974 is for the 80% ignition probability of methane, hence it is likely to be greater than the MIE reported by other researchers/sources.

2.5.4 *Limitations of MIE apparatus*

MIE determination (historically and to date) has been carried out through the capacitive spark discharge. The charge built up was with a gradual transfer of high voltage to the electrode–capacitor system. A rotary charger with small metal spheres mounted on either end of hard rubber is used to slowly increase the charge by alternately touching the terminals of the power source and the spark electrode, transferring charge during each contact of the metal spheres to the electrodes.

This mechanism allows high voltage to be present on the electrodes for a considerable longer duration of time prior to the discharge. The long duration of high voltage permits the condition of a corona discharge. The high voltage prior to the spark can also ionise the gas, disturbing the actual dielectric strength of the gas mixture. This in turn affects the voltage at which a spark occurs and has a direct impact on the calculation of the energy. The spark inside the gas chamber occurred after a certain time lag, which was observed by Lewis to be considerably long. Radium capsules of various strengths were placed in the bomb to reduce the time lag (Lewis et. al 1987). The effect of radium capsules on the MIE values are unknown. In later work the apparatus was modified to replace the rotary charger with a resistor rod, of the order of 10^{11} Ω which served the same purpose of slowly transferring charge from the power unit to the spark electrodes. The resistor rod was moistened with glycerine or conversely (by rubbing the rod with a dry cloth), to accelerate or retard the rate of charge transfer (leakage). These activities helped in adjusting the charging process but required a lot of trial and error. Hence this method cannot be termed as a standard method for the determination of MIE.

2.6 Ignition energy and ignition probability for gases

Historically when the MIE's for gases were evaluated the primary goal was in relation to safety standards applicable to the industries. The standards were based on threshold energy limits for released gases, or for the coal mining industry. The aim was to prevent fire and explosion. Hence the MIE was equated to the minimum observed energy value resulting from a series of ignition tests carried out in a test apparatus.

Most researchers only report a single value of MIE for a particular gas. However, due to local fluctuations within a nominally uniform mixture, ignition of a gas mixture with a given energy level is not guaranteed even if the energy exceeds the MIE. Therefore, there is an associated probability of ignition for a given energy level and given gas concentration. In Table 2.15, Moorhouse, 1974 provides details of the associated probability (80%). Lewis and von Elbe reported MIE values when one ignition occurs in a hundred tests for the specified energy level. (i.e. MIE with an ignition probability of 1%). Komai et. al., (1994) has used a criteria of 50 % ignition probability for the experiments investigating the ignition of a methane - air mixture by frictional sparks. Kono et al. (1977) has determined MIE for three lean concentrations of propane in air. The minimum ignition energies were based on a 50 percent ignition frequency criterion. Parker (1985), using pulsed sparks, determined MIE, for 2.7 percent propane in air with an ignition probability of 10 percent. So the MIE values are reported at different values of ignition probability. To have uniformity in the energy values, it is necessary to associate ignition probability values to the ignition energy values.

Although data is available in the literature for the MIE of methane and for hydrogen, only a limited range of concentrations are normally examined and reported. There is strong variation in ignition energy as the lean limit is approached (Shepherd, J.E., et. al 2000). Moreover the ignition energies of methane-hydrogen-air gas mixtures are not available. Hence there is a need to determine these ignition energies and associated ignition probabilities experimentally.

Accordingly, an experimental apparatus was designed, primarily based on the capacitive spark discharge. The apparatus is similar to the Lewis and von Elbe apparatus with certain modifications to overcome the limitations outlined in section 2.5.4. The electric circuit was based on the spark ignition of dust clouds as proposed by Randeberg 2006. The entire experimental setup, its design and the operating procedure is presented in the next chapter.

Design and Operation of the Experimental facility

3.1 Introduction

The experiments reported in this thesis to study the ignition of the methane-hydrogen-air system were carried out in a facility, designed and built at the Department of Chemical Engineering, Loughborough University. Prior to actual experimental runs, the equipment was thoroughly checked. All the gas pipelines, gas regulators, manifold assembly and cylindrical vessel were pressure tested with soap solution at each joint, to eliminate leaks. A pressure holding test (with dry air) at 200 kPa pressure was carried out. The entire experimental setup was also vacuum tested at the lowest achievable vacuum (using a vacuum pump) to demonstrate that there was no ingress of any outside gases during experimental runs.

3.2 Conceptual design

The experimental facility comprised of: a cylindrical combustion chamber; a gas delivery system; an electrical circuit for generating a spark to ignite the gas-air mixture; and a data acquisition system. The cylindrical combustion chamber was equipped with instruments to measure the pressure and temperature within the chamber and also the potential difference across the electrodes at the time the spark was generated (i.e. the electrode voltage). A schematic of the experimental facility is presented in Figure 3.1.

The key components of the experimental setup are

- Gas supply, delivery and evacuation system
- Cylindrical combustion chamber
- Electrical circuit for ignition of the gas –air mixture
- Instrumentation
- Data acquisition system
- Safety systems

The experimental programme involved numerous explosions of flammable gas – air mixtures formed inside the combustion chamber. The flammable gas consisted of methane, hydrogen and mixture of methane and hydrogen. Fire and explosion hazards were present as a consequence of the highly flammable nature of the gases. In addition, the generation of a very high voltage (15 kV) across the spark electrodes made the experimental setup vulnerable to electrical hazards. Hence, safety interlocks were provided to mitigate the risks and these are discussed separately in Section 3.6 (Safety systems).

3.3 Gas supply, delivery and evacuation system

The gas supply and delivery system for the experimental facility comprised of three gas cylinders (methane, hydrogen and dry air) each at an initial gauge pressure of 20 MPa (200 bar – when the cylinders were full). These gas cylinders were secured in position with belts to prevent them being disturbed. Speciality gases two-stage regulator valves were fitted to each gas cylinder to deliver the gases to the cylindrical combustion chamber. Gases from BOC (Brin's Oxygen Company, Ltd.) were utilized in this work. The Cylindrical combustion chamber was evacuated with an Edwards vacuum pump.

The gas cylinder regulator valve was opened and the delivery pressures for the flammable gases were set at a gauge pressure of approximately 50 kPa (~ 7.5 psi). Limiting the maximum pressure upstream of the combustion chamber gave better control when introducing the flammable gases, since the gas introduction process was manually controlled through the operation of manifold valves (V1 to V3 - Figure 3.1). The delivery pressure for the dry air cylinder was set at a gauge pressure just above 100 kPa (~ 15 psi), since air was the last gas to be introduced into the combustion chamber and the final pressure within the combustion chamber prior to ignition was required to be approximately 1 atm.

Firstly, the vessel was evacuated using a vacuum pump. Once the gas cylinder delivery pressure was set to the desired values the gases were admitted one at a time and the desired mixture obtained through the method of partial pressures. The flammable gas (methane and/or hydrogen) was fed to the vessel by opening the appropriate manifold valve. Each manifold valve was completely closed once the

desired amount of methane (or hydrogen) was added to the vessel. The first gas fed to the chamber occupied the entire volume of the vessel and the actual amount of the gas was computed by the data acquisition system. For experiments with a mixture of flammable gases (methane and hydrogen), the desired quantity of the second flammable gas (either methane or hydrogen) was injected by opening the second manifold valve. The amount of the second flammable gas added to the vessel was also computed by the data acquisition system. The air inlet valve on the manifold was slowly opened, so as to increase the pressure inside the vessel to atmospheric. The pressure transducer reading P1 attached to the manifold was recorded to determine the vacuum achieved and after the addition of each of the component gases (including dry air), until near atmospheric pressure conditions were achieved. At the end of the gas filling process, the gas filler pipe was removed from the vessel by disconnecting the coupling (CV4) so that the explosion vessel was isolated from the feed gas system. This action also isolated the pressure transducer (P1) from the overpressure wave generated after gas ignition, thus preventing it from being damaged.

After the introduction of each gas and air (the last component), a settling period of 1 to 2 minutes was always provided before the data acquisition system was initiated to record the values of the parameters. The turbulence (eddies) generated due to gases introduction dissipates within this time, as observed through the stabilised pressure transducer readings (P1 and P2) and a quiescent well mixed gas-air mixture was achieved. Thus the gases conditions inside the experimental vessel were essentially quiescent and the experimental results reported relate to the ignition of a quiescent fuel gas-air mixture formed within the cylindrical combustion chamber.

Figure 3.1 : Schematics of the experimental setup

3.4 Cylindrical combustion chamber

The cylindrical combustion chamber was fabricated from a 152 mm (six inch) diameter, seamless, (schedule 40) stainless steel pipe of length 232 mm. (Figure 3.2). Flanges were welded on either side of the pipe and polycarbonate plates were used as windows to observe the onset of ignition and the subsequent burning process. Stainless steel flanges covering either side of the pipe were specially fabricated (see Appendix C, Figure C.1) to hold the polycarbonate plate in place with minimum compressive stress. The windows retaining flange was secured to the vessel flange using bolts. The calculations on the required thickness of the vessel shell and windows (incorporating a safety factor) for the expected pressure rise inside the vessel is presented in Appendix C.

Figure 3.2 : Schematic representation of the cylindrical combustion chamber

The cylindrical vessel had a total of six tappings, one each for gas feed and exhaust, two spark electrode insertion tappings diametrically opposite, one tapping for a thermocouple insertion and one tapping for a pressure transducer. As described in Section 3.2, the gases were introduced to the vessel through a manifold device after evacuation, using the vacuum pump to the lowest steady state pressure value achievable. The pressure transducer attached on the gas manifold (P1 - Figure 3.1) displayed the dynamic/transient pressure of the introduced gas. Steady state

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pressure values were recorded at vacuum condition and after the introduction of each gas component to the vessel.

A second pressure transducer (P2 - Figure 3.1) mounted on the vessel was used for recording the pressure rise inside the vessel after the ignition. The transducer had a pressure range from near complete vacuum conditions to 930 kPa (~ 135 psi). The sole purpose of having this pressure transducer in place was to check and ascertain the ignition of gases inside the vessel. A fast response time of 2 ms for this transducer made sure that the smallest overpressure wave generated through the burning process was captured, even though at times it was not possible to observe a flame through the viewing window.

A thermocouple was used to measure and record the gas temperature. The gas mixture inside the cylindrical vessel was prepared at ambient temperature. The temperature recording was initiated only after ignition of the vessel contents (after the onset of the spark). The dynamic temperature rise of the thermocouple sensor was recorded. The only purpose for temperature measurement/recording was to determine the occurrence of ignition inside the vessel. This was an additional instrument to the pressure transducer (P2 - Figure 3.1) mentioned above to ascertain ignition of the vessel contents.

The spark electrodes were 2 mm diameter copper rods. The copper rods were tightly housed in a solid teflon rod 10 mm outside diameter to provide the necessary support to the copper electrodes and also to act as electrical insulators from the stainless steel vessel. Details of the electrode assembly inside the vessel is shown in Figure 3.3. Approximately 10 mm of bare copper rod protruded from the teflon housing near the centre of the vessel. The tips of the copper rods were produced, initially with a 60° taper angle, but in order to reduce the generation of a corona, the electrode end tips were slightly rounded off. The electrode gap was set at 2 mm during the ignition energy measurement experiments performed inside the vessel. After every 4 to 5 successful ignition attempts of the vessel contents, the electrode tips (copper tips) were cleaned. This action prevented the deposition of combustion/oxidation products on the copper electrodes. The condition of the electrode tips was also observed through the polycarbonate windows and accordingly the decision to clean the electrodes was initiated.

Figure 3.3 : The electrode assembly

3.5 The Capacitive spark discharge circuit

The electrical circuit for the capacitive spark generation and energy measurement is shown in Figure 3.4. The spark was generated by the release of electrical charge stored in a capacitor when the voltage across the electrode tips overcame the dielectric strength of the gaseous atmosphere. The stored energy for a capacitor with capacitance CP, charged to voltage V is given by

$$E = 0.5 * CP * V^2 \quad \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots(3.1)$$

Where CP Capacitance (F)

   V Charged voltage of capacitor (V)

   E Stored energy in the capacitor (J)

Since the energy required for gas ignition is very small, the value of E is reported as mJ rather than J. In the present study the stored energy in the capacitor was used to create an electrical discharge which defines the ignition energy. The ignition energy tests follows the practice started by Lewis and von Elbe (1987) of reporting the stored energy rather than measuring the energy discharged into the spark. The circuit for measuring ignition energy for the gases, is based on standards BS EN 13821 : 2002 (Potentially explosive atmospheres – Explosion prevention and protection – Determination of minimum ignition energy of dust/air mixtures) and ASTM E582 1988 (Standard Test Method for Minimum Ignition Energy and Quenching Distance in Gaseous Mixtures)

Figure 3.4 : Simplified electrical circuit for Capacitive spark discharge

The ignition energy for gases (methane or hydrogen) at stoichiometric conditions is very low, hence an electrical circuit capable of producing sparks of very low energies was utilised in the present study. A similar circuit was utilised by Randeberg et. at (2006), to study the minimum ignition energy of (low energy) dust clouds, in the below 1 mJ region. Accordingly, the present spark generator was designed based on the feed of 230 V AC mains. This spark generator circuit is similar to the ordinary car induction coil, where instead of the 12 V DC (battery) supply, the circuit was modified to have a rectified 230 V AC mains signal (see Appendix F, Figure F.1 for the rectifier circuit). Safety interlocks were added to this circuit as a consequence of the flammable nature (explosion potential) of the gases and the handling high voltage, and to protect the low pressure transducer.

The circuit operates in two steps

1. Generation of Extra High Tension (EHT) / high voltage pulse through the EHT pulse generator.

2. Capacitor discharge to the spark in the gas air mixture, charged by this EHT pulse.

A trigger pulse, as shown in Figure 3.4 was fed to a thyristor. The triggering of the thyristor discharges the primary capacitor of 1 µF (initially charged to the rectified AC

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mains voltage) through the primary coil of the HV transformer. This induces a high voltage pulse with amplitude of around 15 kV on the secondary coil side. The high voltage/EHT pulse was fed through a charging resistor, causing a voltage build-up on the discharge capacitor, placed downstream of the charging resistor. Discharge of the discharge capacitor occurred when the voltage across the electrode gap (2 mm) reached the breakdown voltage (6 kV for a 2 mm electrode gap in air). Specific combinations of the resistor and discharge capacitor, resulted in different energies being produced as presented in Table 3.1. Hence, different spark energies could be studied starting from the lowest energy (smallest capacitor) to the maximum energy achievable (largest capacitor) from the electrical circuit.

Table 3.1 : Energy delivered by Capacitor

Discharge Capacitor (pFarads)	Discharge Resistance (k Ohms)	Charging time (µs)	Nominal Breakdown Voltage(kVolts)	Energy (milli Joules)
10	1000	10	6.0	0.18
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

The choice of charging resistor depends on the size of the discharge capacitor, the aim being to avoid recharging of the discharge capacitor during the duration of the discharge. This was achieved by choosing a charging resistor that gave a theoretical time. constant R^*CP (where R is the charging resistance in ohms and CP the discharge capacitance in farads) of at least 1 µs, which ensured insignificant recharging of the discharge capacitor during the spark discharge (normally nano seconds). During experiments, the time constant R^*CP was always kept above the theoretical value of 1 µs. However, if the charging resistor was too large (i.e. large time constant and long charging time), the spark gap voltage would not reach breakdown during the lifetime of the feed from the EHT pulse generator.

A system for the measurement of the spark voltage through a high voltage probe (HV probe) was included in the discharge circuit, enabling measurement of the spark energy according to Equation 3.1. The high voltage probe had a capacitance of 3.0 pF. This 3.0 pF capacitance adds to the discharge capacitance in parallel, and was taken into account when estimating the total capacitance involved in the discharge and also during the estimation of the spark energy. The voltage signal was captured, and the maximum voltage observed was recorded as the peak breakdown voltage. The signal was captured on the oscilloscope for a total duration of 400 ns by adjusting the scale of the oscilloscope.

The fact that the current flow through the charging resistor was negligible during the duration of the discharge is very important for evaluating the spark energy. If the charging resistance (R) is too small, the energy input to the spark process continues to increase until there is appreciable voltage on the secondary side of the HV transformer. Hence, during the experiments it was always assured that the charging time of the discharge capacitor ($R*CP$) was at least 5 µs (> 1 µs.). This assured that the assumption of a "pure" capacitive discharge from the electrical circuit was reasonably correct.

The capacitive spark discharge circuit is very compact, and without switches which can introduce stray capacitance and additional energy. Moreover, the time of spark discharge was much more precisely determined because of the initiation of a trigger pulse. Lewis and von Elbe (1987) used a static high-voltage source to slowly raise the capacitor voltage until breakdown and this required the use of very large resistors during the charging process. In addition, the gas mixture was exposed to high voltage for a much longer time until the actual spark occurred. This provided ample time for corona discharge through the gas medium.

3.6 Instrumentation

3.6.1 Pressure Transducers

Two pressure transducers (PX 219 - 30V15G5V and PX 219 - 30V135G5V, Omega Solid state pressure transducers) were used to measure the gas pressure at two different locations in the present study. The first pressure transducer P1 (PX 219 -

30V15G5V) generates an output from 0 to 5 V DC for a total gauge pressure range of 2.048 MPa (-14.7 psig to 15 psig). The second pressure transducer P2 (PX 219 - 30V135G5V) also generates a similar output from 0 to 5 V DC for a total gauge pressure range 10.321 MPa (-14.7 psig to 135 psig). The calibration data from the National Institute of Standards Technology, United States for these transducer was utilised to convert the output voltage (0 - 5 V DC) to the pressure (kPa) value. The calibration graphs for both these transducers are presented in Appendix G, Figure G.1 and G.2.

The first transducer (P1 -, Figure 3.1) was fixed to the manifold to measure the pressure from vacuum to near atmospheric (101 kPa) pressure. The pressure recordings from this transducer (P1) was utilised to estimate the gas composition (concentration of flammable gas in the gas-air mixture). The second pressure transducer P2 (PX 219 - 30V135G5V) was used to check/ascertain the ignition of the gas–air mixture.

3.6.2 Temperature thermocouple

A Chromel/Alumel (Ni Cr +ve and Ni Al –ve), K – type, RS Thermocouple (maximum continuous temperature range of 1100^0K) was used to record the gas temperature inside the cylindrical combustion chamber after ignition or non-ignition. The temperature recording was initiated only after the onset of the spark. The only purpose for the temperature measurement/recording was to check and ensure the occurrence of ignition inside the vessel. This was an additional instrument to the pressure transducer (P2) mentioned above. This double check on the burning process was useful during experiments with lean concentrations of hydrogen, since the flame was invisible.

3.6.3 Oscilloscope

A Tektronix Oscilloscope model no. TDS 3034 B, was used for data acquisition. The oscilloscope has a bandwidth of 300 MHz, a maximum sampling rate of 2.5 giga samples per second (GS/s) and four channels. Channel 1 was used to record the breakdown voltage by means of high voltage Tektronix probe (P 6015 A). Channel 2 was used to record the current during the spark process across the current sense

resistor by means of probes available with the oscilloscope. (Figure 3.4). The parameters were recorded using the single shot acquisition. (single sequence button). The scope was set to capture the spark voltage for a total duration of 400 ns. The spark discharge process was complete during this time as observed from the voltage trace signal shown in Figure 3.5.

Figure 3.5 : Voltage trace for the spark discharge captured on the scope

3.7 Data acquisition (LabVIEW interface)

National Instruments LabVIEW version 8.0 was used as the programming software tool for measurement (partial pressure of gases), automation of the spark generation, and simultaneous recording of transient temperature and pressure as shown in Figure 3.6. The experimental procedure was semi-automatic where the ignition process and subsequent data recordings were automatic and the preparation of the gas-air mixture (valve operation) was done manually. Human intervention was required during, vacuum creation, introduction of the gases (valves operation) and combustion products expulsion. The LabVIEW program was used for the automation process and data recording. Programming was done to record the dynamic pressure and temperature values at a click of the control button. Data recording could be either continuous, from the time of the click of the button, or just recording a single value parameter on the click of the button. Accordingly the control buttons were given specific modes as required for the recording of each particular parameter.

The pressure value from transducer P1 was utilised to record the initial pressure (vacuum reading) and also after addition of each of the component gases (methane and/or hydrogen and dry air). The signal from pressure transducer P1 was generated at an interval of 0.001 s. At this rate, there was constant fluctuation in the pressure value, and hence an arithmetic mean of 1000 readings was recorded as the pressure value. The control button (AM to File) was selected such that only one Arithmetic Mean pressure value was recorded at the click of the control button "AM to File". All the pressure values - transducer P1 readings (vacuum reading and after addition of each component gases including air) were recorded to the LabVIEW file PP.LMV. The readings from file PP.LMV were used to calculate the flammable gas concentration during each of the experimental runs.

Another control button "Fire & Record" was used to perform three tasks simultaneously. The electrical circuit required a small trigger pulse to activate spark generation (generating high voltage across the electrodes inside the gas chamber). It was observed that the electrical trigger was required for a stipulated duration of 1.5 s., to activate spark generation. The use of a timing function (in LabVIEW programming) coupled with the control button "Fire & Record" helped to deliver the trigger pulse for 1.5 s as required for the electrical circuit. The duration of this electrical trigger is shown as "Preset for firing" in the Figure 3.6.

Simultaneously, on activation of the trigger pulse, the temperature and pressure recordings were started. The temperature (thermocouple T1) and pressure (transducer P2) readings were recorded in separate files (out_temp.LMV and out_press.LMV respectively) for a specified duration of 5 s. through the use of another timing function coupled with the same control button "Fire & Record". The duration of 5 s. was chosen since the thermocouple (T1) and the pressure transducer (P2) were able to record the peak values during this stipulated time. The signals from the thermocouple T1 were at an interval of 0.0833 s. Hence temperature values were recorded in "out_temp.LMV" file at this time interval, starting from the click of "Fire & Record" button for a total time duration of 5 s.

The rate of data acquisition for pressure transducer P2 was at an interval of 0.001 s. and was recorded in file "out_press.LMV". The total duration for pressure recording was also equal to 5 s (same as the temperature recording) but the number of data

points obtained for the pressure value was far greater than for temperature. It was found that the pressure readings defined the success of ignition more clearly than the temperature readings. Appropriate digital values from LabVIEW files generated during each experimental run were recorded in a separate MS excel sheet and then all LabVIEW files were deleted, so that new files generated during the execution of the LabVIEW program carried the same name.

All the LabVIEW files and the control buttons are shown in Figure 3.6. The block diagrams for the LabVIEW program are illustrated in, Appendix F, Figure F.2.

Figure 3.6 : LabVIEW (front panel) with data file and readings

3.8 Safety Systems

The experiments involved handling of flammable gases (methane, hydrogen and mixtures of methane and hydrogen) and carrying out confined explosions inside the vessel. A Risk Assessment (RA) of the entire experiment was carried out. As a result of the RA study, various safety systems were included during the design of the rig. These systems are further discussed below.

Accidental release of flammable gases from the rig cannot be ruled out, hence Xgard (type 5) gas detectors (manufactured by Crowcon) for each of the flammable gases (methane and hydrogen) was installed just above the gas cylinders. An alarm sounded if a gas concentration of 20% of the lower flammability limit (LFL) was detected. If the flammable gas concentration exceeded 40% LFL then air ventilation for the laboratory area was activated.

The filling process to the vessel was manual, through a manifold system. The gas filler pipe feeding gas to the vessel acted as a link between the ignition vessel and the gas cylinders and was attached by means of a coupling (CV4 -Figure 3.1). This coupling CV4 was always detached and locked in a stored position before attempting any ignition. An interlock prevented use of the spark generation circuit if the coupling was not locked in the stored position. This action also isolated the pressure transducer (P1) thereby protecting it from the overpressure wave generated after ignition.

Prior to experimental runs commencing, all the gas pipelines, and connecting equipments were pressure tested so that leaks were eliminated. A pressure holding test at 200 kPa pressure was also carried out to identify any minor leaks. The entire experimental setup was also vacuum tested to detect ingress of gases.

The electrical circuit was provided with a safety interlock 'key switch'. The electrical circuit could only produce a high voltage (spark) after inserting the key and turning it ON. An interlock was also provided on the Electronic Component Unit (ECU) box, housing the capacitor and resistors. The electrical circuit was designed such that

opening of the ECU box discharged any voltage on the secondary side of the HV transformer. A spark could not be achieved, if this box was not closed and latched. A "Monitor Point" and an earth probe was provided to discharge any residual charge manually before replacing/changing electrical components.

Prior to actual experimental runs commencing, a written Risk Assessment Record, Electrical Assessment Record, and COSHH forms were submitted to the competent authorities. All these risk assessment forms are presented in Appendix D of this thesis. A written detailed operation and shutdown procedure was approved and followed during the experiments. Experiments were performed strictly following the laid down procedure.

3.9 Experimental procedure

Prior to conducting a series of experiments the distance between electrode tips was adjusted to 2 mm, so that the spark was able to jump across the electrode gap and the generator could produce sufficient voltage to break the dielectric strength (3 kV/mm) of the air. A confirmatory test was performed to obtain consistent sparks for the combination of largest capacitors (200 pF and 300 pF) with a charging resistance of 100 kΩ in the circuit. This was necessary, since larger capacitors have higher time constant 'R^*CP' (Table 3.1) and hence requires more time to reach the breakdown voltage. The generator provides the high voltage pulse only for a specified duration. The experimental procedure is based on the flowchart presented in Figure 3.7.

```
                        ┌─────────────┐
                        │    Start    │
                        └──────┬──────┘
                               ▼
        ┌──────────────────────────────────────────────┐
        │  Ensure following systems are ON             │
        │  1.  Gas supply to manifold                  │
        │  2.  Spark generation (EHT) system           │
        └──────────────────────────────────────────────┘

        ┌───────────────────────────────────────────────────┐
        │  Evacuate vessel completely using vacuum pump     │
        └───────────────────────────────────────────────────┘
                               ▼
            ┌─────────────────────────────────────┐
            │  Record absolute rig pressure       │
            └─────────────────────────────────────┘
                               ▼
        ┌───────────────────────────────────────────────────┐
        │  Introduce desired quantity of flammable gas      │
        │  and record steady state pressure                 │
        └───────────────────────────────────────────────────┘
                               ▼
        ┌───────────────────────────────────────────────────┐
        │  Introduce dry air and record steady state pressure│
        │  (around atmospheric). Estimate gas concentration  │
        └───────────────────────────────────────────────────┘
                               ▼
    ┌───────────────────────────────────────────────────────────┐
    │  Detach the gas filler pipe from the vessel, and lock it   │
    │  in position                                               │
    └───────────────────────────────────────────────────────────┘
                               ▼
    ┌───────────────────────────────────────────────────────────┐
    │  Initiate spark (& record energy) by activating EHT       │
    │  generator                                                 │
    └───────────────────────────────────────────────────────────┘
                               ▼
                       ◇ Gas Ignited ◇ ──Y──►
                               │ N
                               ▼
    ┌───────────────────────────────────────────────────────────┐
    │  Increase spark energy using larger capacitor             │
    └───────────────────────────────────────────────────────────┘
                               ▼
           N ◄──── ◇ Ignition attempts < 4 ◇ ────► Y
               │
               ▼
    ┌───────────────────────────────────────────┐
    │  Stop  (End series of experiments )        │
    └───────────────────────────────────────────┘
```

Figure 3.7 : Flowchart for the experiment

3.9.1 Gas mixture preparation

During the initial start-up for an experimental run, the combustion chamber was purged with dry air for at least 120 s. to drive out water vapours/flammable gases accumulated from the last experiment. For each of the subsequent experiments in the run, air purging was done with atmospheric ambient air, after evacuation of the vessel, so that combustion products (CO_2, CO, unburned flammables etc.) did not accumulate within the vessel chamber.

The regulators (two stage regulators - valves) on the gas cylinders required for the experiments were opened sufficiently to deliver gas to the cylindrical combustion chamber. The flammable gases were set at a delivery pressure of around 50 kPa (maximum 10 psig) and the dry air was set at a delivery pressure of around 100 kPa (maximum 15 psig). This action gave good control for the introduction of flammable gases into the cylindrical combustion chamber. This was necessary because the gas introduction process was manually controlled through the operation of the manifold valves.

Data acquisition (pressure and temperature recordings) and triggering of the spark process (EHT pulse generation) was carried out using the LabVIEW program specially prepared for the operation of the rig. The major steps for the operation of the vessel are outlined with an explanation for the necessary relevant actions.

The cylindrical combustion chamber was evacuated completely using the Edwards vacuum pump (HP no. G1099 – 80024) to achieve a condition of near complete vacuum (absolute pressure below 2 kPa). The vacuum pump was required to be run for 3 to 5 minutes. A steady state pressure reading as displayed by the pressure transducer (P1) was recorded and stored as a digital value (file : PP.LMV) on actuation of the LabVIEW front panel display button "AM to File" through a click of the mouse. This recording was done after switching off the vacuum pump and isolating the vessel by closing all inlet and outlet valves.

The flammable gas (methane and/or hydrogen) was fed to the cylindrical combustion chamber through the method of partial pressures by opening the appropriate manifold valve. The first gas fed to the chamber occupied the entire volume of the

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vessel and the amount of the gas was computed from the difference between the displayed pressure reading and the vacuum pressure reading as monitored by the pressure transducer and recorded by the LabVIEW program. The manifold valve was completely closed once the desired amount of methane (or hydrogen) was added to the vessel. A steady state pressure value was then recorded with a click of "AM to File". If the experiments were carried out for a mixture of flammable gases (methane and hydrogen), then the desired quantity of the second flammable gas (either methane or hydrogen) was injected by opening the appropriate manifold valve. A steady state pressure value, after closing the manifold valve, was then recorded with a click of "AM to File".

The air inlet valve on the manifold was opened slowly so as to increase the pressure inside the rig to around 101 kPa (pressure transducer P1 reading). The introduction of air in a jet creates turbulence, mixes the gases and results in a homogeneous mixture inside the vessel. The air inlet valve was completely closed after addition of dry air and the steady state pressure transducer reading recorded, (in the same file PP.LMV) with a click of "AM to File".

The gas filler pipe was then removed from the vessel (by disconnecting the coupling CV4) and locked in the stored position. This action isolated the flammable gas cylinders from the combustion vessel. The transducer P1 with a maximum pressure range of 2.048 MPa was also protected from exposure to the overpressure wave.

3.9.2 Gas Ignition

An appropriate value of discharge capacitor and charging resistor, as presented in Table 3.1, were connected into the electrical circuit (starting with the combination of R and CP delivering lowest ignition energy required for the experiment to be conducted).

A trigger pulse to the electrical circuit was actuated with a click of the mouse on the LabVIEW interface button "Fire & Record". This trigger induced a high voltage across the electrode tips inside the gas chamber and initiated a spark. The oscilloscope screen displayed the voltage as a function of time (ns) over the entire duration of the

spark process. The breakdown voltage i.e. maximum voltage achieved during the time duration was manually recorded (in a MS Excel spreadsheet) as shown in Figure 3.6 for the specified flammable gas composition prepared. The energy delivered by the spark was then calculated based on the discharge capacitor in the circuit and the breakdown voltage recorded by the oscilloscope. The energy was recorded irrespective of the success, or the failure of the ignition process. If the first ignition attempt was unsuccessful, then the spark was initiated with a different combination of resistance and capacitance values giving a higher value of energy for the same gas composition. A total of four such attempts were made to ignite the gas, increasing the spark energy incrementally, utilising higher capacitors during each attempt. In the case of ignition failure, a very small amount of mixture burnt by the spark near the electrode tips, can contaminate the un-burnt gases and influence the ignition energy of the mixture. The spark process also generates free radicals, which can modify the ignition characteristics. Hence the maximum number of successive unsuccessful ignition attempts was restricted to four with the same gas-air mixture. If all the four attempts were unsuccessful then the vessel was again evacuated and filled with fresh gases to give the same overall composition.

If the spark, ignited the flammable mixture inside the cylindrical combustion chamber, as observed through the visual inspection, sound, recorded pressure or temperature rise, then the spark energy was recorded as a successful ignition. The cylindrical combustion chamber was evacuated to remove the combustion products, purged with ambient atmospheric air and then again evacuated to start up the process of filling with fresh gases to give the same composition. Attempts were made to prepare exactly the same flammable gas concentration during each planned set of tests and the actual composition achieved was reported along with the spark energies for successful and un-successful ignitions in the MS Excel spread sheet.

The ignition of methane can be visually observed since the vessel had transparent polycarbonate plate flanges. The flame initiation, flame growth and the flame quenching on the walls of the vessels, can be distinctly observed because of the nature of the methane - air combustion process. Hydrogen combustion was often invisible (especially near the lean limit) to naked eyes, and hence the criteria for ascertaining the success of ignition was based on values of pressure and/or temperature recorded after each attempt of spark initiation. The gases within the

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vessel were considered to have been ignited, if the pressure increase within the chamber was more than 7% of the initial pressure value. Cashdollar, et. al. (2000) initially suggested this 7% pressure rise, and this was confirmed through the observations during this work. The temperature rise for lean hydrogen burning, as recorded during these experiments, was negligible, possibly because of the high thermal capacitance of the vessel.

A written operating procedure for the experiments leading to an estimation of the ignition energy and the ignition probability for a quiescent flammable gas concentration was formulated. This is given in Appendix E. The human actions performed during the operation of various valves are based on the valves nomenclature described earlier and shown in Figure 3.1.

3.10 Experimental programme

An experimental programme was formulated to study the entire range of flammable gases concentrations for methane-air, hydrogen-air and methane–hydrogen-air mixtures. The main purpose of this work was to study methane–hydrogen–air mixtures, since this had not been done before and was required for the Naturalhy project. Measurements of MIE for methane–air and hydrogen–air had been made by previous workers and these served as a good check on the experimental method used for the work described in this thesis. As the experiments involved two flammable gases, methane and hydrogen, experiments were initially proposed in which the concentration of each of these gases was studied separately over each of their flammable ranges. Then three different flammable gas mixtures of methane and hydrogen were studied which were 25%, 50% and 75 % hydrogen in methane. Thus a total of five different gases; pure methane, 25% hydrogen in methane, 50% hydrogen in methane, 75 % hydrogen in methane and pure hydrogen were mixed with air and the ignition of these mixtures studied over their flammable ranges. The composition of the fuel-air mixture used during each experiment was identified using the Equivalence Ratio (ER). The Equivalence Ratio is defined in Section 3.10.1 below and an estimation of the range of flammable concentrations studied during the experiments is given in Section 3.10.2.

3.10.1 Equivalence Ratio

The Equivalence Ratio (ER) is often used to define a fuel-air mixture undergoing a combustion process and is defined as the ratio of fuel to air compared to the stiochometric fuel to air ratio. A stoichiometric fuel-air mixture is one that leads to complete combustion since the air provides just enough oxygen to achieve the final combustion products of carbon dioxide and water vapour. For example, a methane-air stoichiometric mixture contains 9.5% by volume methane in air. Similarly for hydrogen, a stoichiometric mixture contains 29.5% by volume hydrogen in air. Experimentally, it is very easy to ignite a mixture which is close to the stoichiometric ratio and often the ignition energies for stoichiometric flammable gases are reported in the literature. The deviation of an actual gas composition from the stoichiometric composition is often reported using the equivalence ratio. Mathematically Equivalence Ratio (ER) is defined as

$$ER = \frac{Ratio\ of\ number\ of\ moles\ of\ flamm.\ gas\ to\ air\ in\ actual\ mixture\ composition}{Ratio\ of\ number\ of\ moles\ of\ flamm.\ gas\ to\ air\ in\ stoichiometric\ composition}$$

A 'lean' mixture will have ER < 1, due to the excessive dilution by unconsumed oxygen and its associated nitrogen. ER > 1 defines the condition of a 'rich' fuel-air mixture. Equivalence Ratios for methane, hydrogen and methane-hydrogen mixtures can be worked out from the Stoichiometric combustion reaction as follows

Combustion reaction for methane	Combustion reaction for hydrogen
$CH_4 + 2 O_2 \longrightarrow CO_2 + 2 H_2O$	$H_2 + 0.5 O_2 \longrightarrow H_2O$
Moles of CH_4 = 1	Nos. of moles of H_2 = 1
Moles of O_2 = 2	Nos. of moles of O_2 = 0.5
Moles of Air = 2 * (100 / 21)	Nos. of moles of Air = 0.5 * (100 / 21)
Stoi. molar ratio CH_4 : Air = 1 : (200 / 21)	Stoi. molar ratio H_2 : Air = 1 : (50 / 21)
Stoi. molar ratio CH_4 : Air = (21 / 200)	Stoi. molar ratio H_2 : Air = (21 / 50)

$$ER_{CH_4} = \frac{(molar\ ratio\ CH_4\ :\ Air)_{actual\ composition}}{(molar\ ratio\ CH_4 : Air)_{stoichiometric}}$$

$$ER_{H_2} = \frac{(molar\ ratio\ H_2\ :\ Air)_{actual\ composition}}{(molar\ ratio\ H_2 : Air)_{stoichiometric}}$$

$$ER_{CH_4} = \frac{(molar\ ratio\ CH_4\ :\ Air)_{actual\ composition}}{(21/200)}$$

$$ER_{H_2} = \frac{(molar\ ratio\ H_2\ :\ Air)_{actual\ composition}}{(21/50)}$$

$$.ER_{CH_4} = \frac{200}{21} * \left(\frac{moles\ of\ CH_4}{moles\ of\ Air}\right)_{actual\ composition}$$

$$ER_{H_2} = \frac{50}{21} * \left(\frac{moles\ of\ H_2}{moles\ of\ Air}\right)_{actual\ composition}$$

For a two component (methane and hydrogen) mixture the equivalence ratio can be worked out as follows

$$aCH_4 + bH_2 + (2a+0.5b)O_2 \quad \text{--------->} \quad aCO_2 + (2a+b)H_2O$$

where a is the moles of CH_4 in the mixture

b is the moles of H_2 in the mixture

Ratio of fuel ($CH_4 + H_2$ mixture) to air at stoichiometric concentration will be

$$\left[\frac{Fuel}{Air}\right]_{stoichiometric} = \frac{(a+b)}{(2a+0.5b)*\left(\frac{100}{21}\right)}$$

Equivalence ratio for the mixture will be

$$ER_{mixture} = \frac{\left[\dfrac{Fuel}{Air}\right]_{actual\,composition}}{\left[\dfrac{Fuel}{Air}\right]_{stoichiometric}}$$

$$ER_{mixture} = \frac{\left[\dfrac{(a+b)}{moles\ of\ Air}\right]_{actual\,composition}}{\left[\dfrac{(a+b)}{2a+0.5b*\left(\dfrac{100}{21}\right)}\right]}$$

$$ER_{mixture} = \left[\frac{(2a+0.5b)*\left(\dfrac{100}{21}\right)}{moles\ of\ Air}\right]_{actual\,composition}$$

$$ER_{mixture} = \frac{200}{21}*\left(\frac{a}{moles\ of\ Air}\right)_{actual\,composition} + \frac{50}{21}*\left(\frac{b}{moles\ of\ Air}\right)_{actual\,composition}$$

Hence for the two component (methane and hydrogen) mixture, the equivalence ratio can be worked out as follows

$$ER_{mixture} = ER_{CH_4} + ER_{H_2}$$

Therefore the equation used for estimating ER of two component mixture is

$$ER_{mixture} = \frac{(200/21)*(moles\ of\ CH_4)+(50/21)*(moles\ of\ H_2)}{(moles\ of\ Air)}$$

Experimentally the gases mixture were prepared on a volume percentage basis. Since for gases, the volumetric percentage is equivalent to the molar percentage, accordingly the equation for the equivalence ratio is modified as :

$$ER_{mixture} = \frac{(200/21) * (volumetric\ percentage\ of\ CH_4) + (50/21) * (volumetric\ percentage\ of\ H_2)}{(volumetric\ percentage\ of\ Air)}$$

3.10.2 Flammability limits

The flammability limits LFL and UFL for Methane were taken as 5% and 15% respectively. For hydrogen the LFL and the UFL values were taken as 4% and 75% respectively (Lewis and von Elbe, 1987). For the two component methane hydrogen mixtures (25%, 50% and 75% hydrogen in methane) the flammability limits was estimated from the Le Chatelier's equation (Lees, F.P. 1980) as follows:

$$LFL_{mix} = \frac{1}{\dfrac{mole\ fraction\ CH_4}{LFL_{CH_4}} + \dfrac{mole\ fraction\ H_2}{LFL_{H_2}}}$$

$$UFL_{mix} = \frac{1}{\dfrac{mole\ fraction\ CH_4}{UFL_{CH_4}} + \dfrac{mole\ fraction\ H_2}{UFL_{H_2}}}$$

This Le Chatelier's equation is empirically derived equation and is not universally applicable. In the present experiments, the equation was utilised to get some idea about the limiting ranges for the mixtures, so that gas concentrations could be determined. The LFL and UFL values thus computed from the Le Chatelier's equation for the gas mixtures are presented in Table 3.2.

Table 3.2 : Flammability limits of gas mixture

Gas mixture	LFL	UFL
25 % Hydrogen in Methane	4.7	21.1
50 % Hydrogen in Methane	4.4	27.7
75 % Hydrogen in Methane	4.2	40.5

3.10.3 Targeted gas concentrations

Methane has flammability limits of 5% to 15% (vol. percentage) at ambient atmospheric conditions in air. Even though the flammable limits for methane are well known it is difficult to ignite mixtures close to the limiting conditions. Hence the extreme end values of the flammability range were discarded from the proposed experimental concentrations. The ignitions of the lean mixtures is more important from a safety perspective than rich mixtures. Any accidental flammable gas release within a confined space, will result in a concentration build-up first crossing the lean limit.

For the first two gases (methane and 25% hydrogen in methane) four different concentrations groups were studied. The next two gases (50% hydrogen in methane and 75% hydrogen in methane) five different concentrations were targeted. Hydrogen has the widest flammability limits (4% to 75%) and hence was subdivided into six concentrations so that the entire flammable range could be covered during the experimental programme. The target concentrations for the 5 different flammable gases/gas mixtures are shown in Table 3.3.

Table 3.3 : Target gas concentrations for the experimental programme

Gas	LFL-UFL (vol.%)	Target conc. of total flammable gas (vol.%)					
CH_4	5 – 15	6	8	10	14		
75% CH_4 + 25% H_2	4.7 - 21.1	8	12	16	20		
50% CH_4 + 50% H_2	4.4 - 27.7	6	10	16	20	26	
25% CH_4 + 75% H_2	4.2 - 40.5	8	14	21	28	38	
H_2	4 – 75	6	10	20	30	45	60

The targeted concentrations of flammable gases were based on the theoretical calculations of the flammability ranges. These targeted concentrations were initially prepared inside the vessel, considering the practicality aspect of achieving a consistent repeatable concentration during each run. The flammable mixtures were prepared from pure gases methane and hydrogen directly inside the vessel through the method of partial pressures.

Results, Data analysis and Discussions

A total of approximately 2000 ignitions were attempted on the flammable gas–air mixtures. These covered the flammable gases methane, hydrogen, and three different mixtures of methane and hydrogen. Different targeted concentrations for each of these gases were given in Table 3.3. In each case a range of ignition energies was used by varying the capacitor-resistor combination. Both ignition and non-ignition of the flammable gas–air mixtures was recorded. The basic results giving percentage flammable gas concentration, (methane and/or hydrogen mixture), total flammable gas concentration, air concentration, spark energy and success of ignition are presented in Appendix H, from Tables H.1 to H.5. In this section the experimental data is analysed to arrive at the results from the experimental observations. Firstly the data analysis of the measured parameters is considered in Section 4.1. In Section 4.2 the errors in the measurements and derived values are assessed. The results obtained for methane, hydrogen and methane–hydrogen mixtures respectively are presented in Section 4.3 to 4.5. The overall results are discussed in Section 4.6.

4.1 Data analysis

During the gas mixture preparation, spark initiation and subsequent ignition or non-ignition of the flammable gas–air mixtures, various parameters were measured and recorded. The measured parameters were analysed to estimate the gas concentration, spark energy and the ignition probability from the series of experiments. The actual calculations performed during the estimation of the parameters are discussed in more detail in subsequent sections.

4.1.1 Concentration estimation

Experimental runs were targeted for various gas concentrations across the flammable range. The gas concentration (C_{gas}) was estimated from the values of recorded pressure (kPa) sensed by the pressure transducer (P1 - Figure 3.1).

Pressure recordings were made at each stage of the gas filling process (Figure 4.1) and used to calculate gas concentration as follows.

$$C_{gas} = \frac{P_{gas} - P_{vac}}{P_{gas-air}} \times 100 \qquad\qquad ...(4.1)$$

Where

P_{vac} pressure when vessel is under vacuum (kPa)

P_{gas} pressure when flammable gas is introduced into the vessel (kPa)

$P_{gas-air}$ total pressure of flammable gas and air inside the vessel (kPa)

Figure 4.1 : Pressure recording during gases introduction

For two a component system comprising of a methane-hydrogen mixture, when methane was the first flammable gas injected during the gas filling process (see Figure 4.2) the concentration was estimated as follows

$$C_{CH_4} = \frac{P_{CH_4} - P_{vac}}{P_{total\ gas-air}} \times 100$$

$$C_{H_2}^{\cdot} = \frac{P_{CH_4 + H_2} - P_{CH_4}}{P_{total\ gas-air}} \times 100$$

$$C_{Total\ flammable\ gases} = C_{CH_4} + C_{H_2}$$

$P_{total\ gas-air}$

P_{H2+CH4}

P_{CH4}

P_{vac}

Figure 4.2 : Pressure recording during methane and hydrogen introduction

The Equivalence Ratio (ER) for the flammable gas-air mixture (for two components) was also calculated as described in Section 3.8.1.

$$ER_{mixture} = \frac{(200\,/\,21)*(volumetric\ percentage\ of\ CH_4) + (50\,/\,21)*(volumetric\ percentage\ of\ H_2)}{(volumetric\ percentage\ of\ Air)}$$

When only one flammable gas (either methane or hydrogen) was mixed with air, then ER, was calculated as

For Methane

$$ER_{CH_4} = \frac{200}{21}*\left(\frac{volumetric\ percentage\ of\ CH_4}{volumetric\ percentage\ of\ Air}\right)$$

For Hydrogen

$$ER_{H_2} = \frac{50}{21}*\left(\frac{volumetric\ percentage\ of\ H_2}{volumetric\ percentage\ of\ Air}\right)$$

4.1.2 Energy estimation

Ignition assurance

In a closed vessel the success of ignition for invisible flames was difficult to establish. In order to determine the ignition energy, a criteria for successful ignition is required. Most tests resulted in visually observed ignitions but for the borderline cases, the pressure transducer was relied on to define ignition. The overpressure wave as

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observed for methane (6.7% and 8%) and hydrogen (30%) ignition are presented in Figure 4.3.

Figure 4.3 : Overpressures during methane and hydrogen ignitions

When the pressure rise was very small, at lean ignitions of hydrogen (Figure 4.4), assurance was required that the pressure rise was due to gas combustion. It was initially confirmed that there was no pressure rise, associated with the spark by recording the pressure for test runs with air. To distinguish ignitions from non-ignitions for lean concentrations of hydrogen, the combustion reaction should result in a quantifiable pressure rise within the vessel. The smallest pressure rise observed during the combustion process was just over 7%.

Figure 4.4 : Overpressure for lean hydrogen ignitions

Energy determination

Electrical sparks have been used over a number of years to measure the Minimum Ignition Energy (MIE) of gases. Electric sparks are a very hot and fast acting mode of ignition. The discharge time of an electric spark is very short (of the order of 10^{-8} to 10^{-7} s), the energy that is imparted to the gas at the end of the discharge period is highly concentrated, so that a very steep temperature profile with a very high temperature at the centre is established. (Lewis and von Elbe 1987). In the initial stage of the flame development, the combustion process is insufficient to maintain such a steep temperature profile, so the profile broadens and the temperature at the centre decreases. Depending on the physical and chemical properties of the gas and the discharge energy (if sufficient to establish sustained combustion) the flame continues to propagate as a steady state wave. The temperature at the centre settles down to the value of the flame temperature.

Spark discharges are typically two electrode discharges. They occur between conductors (electrodes) with different electric potentials. A spark is a discrete discharge that bridges the gap between the two conductors in the form of an ionisation path, in which the stored energy is quickly transferred. The spark is

triggered when the breakthrough field strength is reached at a certain point in the gap between the electrodes, normally at the surface of the electrodes. The field strength between the electrode space must be sufficiently high, that the discharge can travel through that space. For a homogeneous electric field between the electrodes, spark discharge will occur when the ratio of potential difference and the gap between the electrodes reaches about 3 kV/mm (dielectric strength of air). For an electrode spacing of 2 mm a potential of 6 kV would be required for a spark to occur. Although the dielectric strength of air is 3 kV/mm, the dielectric strength of hydrogen is only 1.75 kV/mm. (Astbury and Hawksworth, 2007). The variation of dielectric strength with concentration of hydrogen in air mixtures is unknown.

Studies of nanosecond duration spark discharges suggest that the process proceeds in several stages: (i) deposition of electrical energy to create an ionized channel of hot gas between the electrodes; (ii) expansion of the hot gases and creation of a shock wave; (iii) recirculation of fresh gases into the channel; (iv) turbulent mixing of hot and cold gases (Kono et. al 1989). As a result of these processes, the kernel of hot gas created in the discharge region initiates a reaction in the surrounding cooler gas, buoyancy may also come into play. If the ignition energy is sufficiently large, then a propagating flame emerges from the vicinity of the kernel of hot gas. Several ideas (Lewis and von Elbe 1987, Kondo et. al 2003) have been proposed to correlate ignition energy with other combustion parameters. Because the process is very complex, there was limited success in the theoretical prediction of ignition energy.

The standard way to characterize capacitive discharge sparks is in terms of the quantity of stored electrical energy, measured as millijoule (mJ). Precise measurement or estimation of stored electrical energies is essential when determining the ignition energy. The actual amount of energy that is deposited in the gas by the discharge is lower than the actual spark energy, particular for short duration sparks. A distinction can be made between measurement of gross capacitor energy prior to breakdown and integration of spark power versus time. In the former case, the energy is simply assumed equal to the difference between stored capacitor energy before and after the discharge:

$$E = 0.5CP\left(V_{before}^2 - V_{after}^2\right) \qquad \qquad ...(4.2)$$

where E   is the calculated energy (J)

                since E, is very small it is reported as mJ in the entire thesis

       CP   is the capacitance (F)

    V_{before} is the capacitor voltage before discharge (V)

    V_{after} is the voltage after discharge (V)

Usually, $V_{before} \gg V_{after}$, and thus the energy can be approximated by

$$E \cong 0.5CP * V_{before}^2 \qquad \qquad ...(4.3)$$

Whether the spark energy can be accurately estimated by this simple expression depends on the characteristics of the discharge circuit. The discharge circuit inevitably contains some resistive elements in series with the spark, where some of the energy is lost, and not delivered to the spark. The energy lost in the electric circuit, to the electrodes, through a corona and radiation is disregarded when stating the spark energy. When estimating the spark energy purely based on stored capacitor energy, information about the spark duration, voltage and current waveforms are not obtained.

Randeberg E. (2006) measured the circuit variables (voltage and current trace waveforms) and calculated energy from the integral of the power, i.e. the product of voltage 'V' and current 'I' of the spark, over the duration of the discharge.

$$E = \int V I \, dt \qquad \qquad ...(4.4)$$

Where V is the voltage on capacitor at a point in time (during the duration of spark)

Thus, only the energy delivered by the spark was found, and circuit capacitance and losses are implicit. A major challenge to this approach is the fact that the different spark phases (e.g. creation of ionized channel of hot gas, breakdown, and spark etc.) have durations differing by orders of magnitude, and the currents and voltages are varying by orders of magnitude in the different phases. (Maly R. and Vogel M. 1979).

The actual energy delivered to the spark can be estimated by subtracting the resistive losses from the spark energy over the duration of the discharge using :

$$E = \int V\,I\,dt - \int R\,I^2\,dt \qquad \qquad ...(4.5)$$

where R - is the circuit resistance in series with the spark gap (Ω)

      I - is the current flowing through the spark (A)

      V - is the voltage measured as a function of time during the spark (V)

However the resistive losses are small, for the test rig used for this work, so the second term in the Equation 4.5 can be neglected. Moreover, to measure the spark energy as per Equation 4.5, it was important to measure parameters 'V' and 'I' as functions of time. (nanosecond discharge over the spark duration). The current 'I' was found to oscillate over the spark duration. The metallic stainless steel vessel (a very good electrical conductor) might have impacted upon the frequency and amplitude of this current trace. These difficulties with energy measurement prevented the use of Equation 4.5 to calculate the energy. Moreover most of the earlier work (including those of Lewis and von Elbe) utilised Equation 4.3, to calculate the ignition energy. Accordingly the stored capacitor energy and the maximum value of the voltage trace waveform (captured by the oscilloscope) were used to derive the ignition energy throughout this work.

$$E = 0.5CP * V^2 \qquad \qquad ...(4.6)$$

4.1.3 Ignition probability computation

The result presented in Appendix H, Tables H.1 to H.5, show that for higher spark energy levels, more successful ignitions were achieved, indicating an increase in the probability of ignition with increasing energy. The ignition energy results for each gas concentration for each flammable gas were analysed to derive the associated ignition probabilities as follows :

The entire energy range (zero to maximum value) was sub divided into bands of 2 mJ (except for test results of hydrogen). In each band there would be number of ignition and non-ignition results. However, if a mixture had been successfully ignited by an energy within a low energy band, then clearly this mixture would also have

been ignited had the energy 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. That is :

Assume the energy range used for a series of experiments at a particular concentration is divided into 'M' bands and that for a given band, m. (1 $\leq$ m $\leq$ M) there were IG_m successful ignitions and NI_m non – successful ignitions.

Then the probability of ignition (IP_m) for the energy level associated with band 'm' is given by

$$IP_m = \frac{\sum\limits_{j=1}^{m} IG_j}{\sum\limits_{j=1}^{m} IG_j + NI_m} \qquad \qquad ...(4.7)$$

The ignition probability for each of these individual bands was determined accordingly and then assigned to the average energy value within this band.

The first band was lowest value of non-ignition energy to 2 mJ. Below the lowest successful ignition energy value within this first band, the ignition probability has to be considerably less than the ignition probability for the band. Accordingly, a value of zero ignition probability was assumed for the average energy value, calculated from lowest non-ignition energy value and lowest ignition energy value, within this first energy band.

The ignition probability (IP) was observed to be an exponential function of the ignition energy (E). Hence an equation of the following form was assumed to relate the ignition probability to the energy E.

$$IP = 1 - \exp\left[-b(E - E_{min})\right] \qquad \qquad ...(4.8)$$

Where 'E_{min}' is the energy when IP equals zero.

The above equation tends to 'unity' for higher values of 'E' and is 'zero' at 'E_{min}', and well describes the observed variation in the ignition probability with energy.

Since standard Microsoft excel worksheet does not correlate this type of curve fitting, Equation 4.8 was rearranged as follows :

$$Ln(1 - IP) = bE_{min} - bE$$ ...(4.9)

A best fit plot of '$Ln(1 - IP)$' against energy 'E' provides a line with :

slope $= -b$

Intercept $= bE_{min}$

Accordingly the parameters 'b'and 'E_{min}' was derived for each of the target concentrations for each of the flammable gas. The energy for 1% ignition probability (often reported by many authors) was then calculated from Equation 4.8 as:

$$E_{IP=1\%} = E_{min} - \frac{1}{b} * Ln(1 - 0.01)$$ ...(4.10)

Where $E_{IP=1\%}$ is the energy for 1% ignition probability

E_{min} is the energy when ignition probability is zero

4.2 Error Analysis

The quantifiable parameters of pressure, temperature, capacitance and voltage were measured by various instruments during the experiments. Measurement of these parameters can not be 100% perfectly accurate or exact. The gas concentration in air of the mixture produced is determined from measurements of pressure during the filling process. The accuracy of the concentrations determined is considered in Section 4.2.1. The energy delivered by the spark is determined from the voltage and capacitance as given in Equation 4.6. The accuracy of the spark energy is assessed in Section 4.2.2.

4.2.1 Error analysis for concentration

The absolute error in the measured parameter and accordingly the maximum and the minimum measured values are presented in Table 4.1.

Table 4.1 : Error in pressure measurement

Measured parameter	Measurement error	Maximum value	Minimum value
Pressure	$\pm \Delta P$	$P_{max} = P + \Delta P$	$P_{min} = P - \Delta P$
Concentration	$\pm \Delta C$.	$\left(C_{gas}\right)_{max} = \left(C_{gas}\right) + \Delta C$	$\left(C_{gas}\right)_{min} = \left(C_{gas}\right) - \Delta C$

Referring to specification of the pressure transducer (PX 219-30V15G5V) the error ($\pm \Delta P$) in pressure measurement is ± 0.25%.

i.e. $\pm \Delta P = \pm 0.0025 * P$ ...(4.11)

The error in concentration of the gas (C_{gas}) was determined from the pressure transducer error ($\pm \Delta P$) used during gas mixture preparation stage. The concentration was measured as per Equation 4.1 discussed earlier.

$$C_{gas} = \frac{P_{gas} - P_{vac}}{P_{gas-air}} \times 100$$

The gas concentration (C_{gas}) is the volume percentage concentration of the flammable gas in the gas-air mixture.

The basic equation for error in the concentration measurement of the gas is

$$2 * \Delta C = \left(C_{gas}\right)_{max} - \left(C_{gas}\right)_{min} \quad ...(4.12)$$

The maximum value of concentration will be

$$\frac{\left(C_{gas}\right)_{max}}{100} = \frac{\left(P_{gas} + \Delta P_{gas}\right) - \left(P_{vac} - \Delta P_{vac}\right)}{\left(P_{gas-air} - \Delta P_{gas-air}\right)}$$

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$$\frac{\left(C_{gas}\right)_{max}}{100} = \frac{\left(P_{gas} + 0.0025 P_{gas}\right) - \left(P_{vac} - 0.0025 P_{vac}\right)}{\left(P_{gas-air} - 0.0025 P_{gas-air}\right)}$$

$$\frac{\left(C_{gas}\right)_{max}}{100} = \frac{\left(P_{gas} - P_{vac}\right) + 0.0025\left(P_{gas} + P_{vac}\right)}{P_{gas-air}\left(1 - 0.0025\right)}$$

Similarly the minimum value of concentration will be

$$\frac{\left(C_{gas}\right)_{min}}{100} = \frac{\left(P_{gas} - P_{vac}\right) - 0.0025\left(P_{gas} + P_{vac}\right)}{P_{gas-air}\left(1 + 0.0025\right)}$$

$$\frac{2 * \Delta C}{100} = \frac{\left(P_{gas} - P_{vac}\right) + 0.0025\left(P_{gas} + P_{vac}\right)}{P_{gas-air}\left(1 - 0.0025\right)} - \frac{\left(P_{gas} - P_{vac}\right) - 0.0025\left(P_{gas} + P_{vac}\right)}{P_{gas-air}\left(1 + 0.0025\right)}$$

$$= \frac{\left(1 + 0.0025\right)\left[\left(P_{gas} - P_{vac}\right) + 0.0025\left(P_{gas} + P_{vac}\right)\right] - \left(1 - 0.0025\right)\left[\left(P_{gas} - P_{vac}\right) - 0.0025\left(P_{gas} + P_{vac}\right)\right]}{P_{gas-air}\left(1 - 0.0025\right)\left(1 + 0.0025\right)}$$

$$= \frac{\begin{array}{c} P_{gas} - P_{vac} + 0.0025\left(P_{gas} + P_{vac}\right) + 0.0025\left(P_{gas} - P_{vac}\right) + 0.0025^2\left(P_{gas} + P_{vac}\right) \\ -\left(P_{gas} - P_{vac}\right) + 0.0025\left(P_{gas} + P_{vac}\right) + 0.0025\left(P_{gas} - P_{vac}\right) - 0.0025^2\left(P_{gas} + P_{vac}\right) \end{array}}{P_{gas-air}\left(1 - 0.0025^2\right)}$$

$$\frac{2 * \Delta C}{100} = \frac{2 * 0.0025\left(P_{gas} + P_{vac} + P_{gas} - P_{vac}\right)}{P_{gas-air}\left(1 - 0.0025^2\right)}$$

$$\frac{\Delta C}{100} = \frac{0.0025\left(2 P_{gas}\right)}{P_{gas-air}\left(1 - 0.0025^2\right)} \qquad \qquad ...(4.13)$$

Considering $(1 - 0.0025^2) = 1$ and that P_{vac} is small so $P_{gas} = P_{gas} - P_{vac}$

$$\frac{\Delta C}{100} \cong 0.0025 * 2\left(\frac{P_{gas} - P_{vac}}{P_{gas-air}}\right) \qquad \qquad ...(4.14)$$

$$\frac{\Delta C}{100} \cong 0.0025 * 2C_{gas}$$

$$\frac{\Delta C}{C_{gas}} \cong 0.5 \qquad \text{or} \qquad \frac{\Delta C}{C_{gas}} \cong (2\Delta P) \qquad ...(4.15)$$

The percentage error in concentration is 0.5% of the measured gas concentration (C_{gas}) or the error in concentration is twice the error in pressure measurement $(2\Delta P)$. Even if Equation 4.13 is utilised to calculate the percentage error in concentration, the maximum error was 0.65% for the range of pressure values used during the experiments. Accordingly Equation 4.13 and Equation 4.15 give a similar error.

The error estimated for the gas concentration is very small ($\Delta C = 0.5$%) as compared to the range of the gas concentrations studied (6.5% to 10.5%). The error for the methane gas concentration as shown by the horizontal spread of the concentration value is presented in Figure 4.5. Two data sets of 6.5% and 8% methane concentrations only are plotted to show the horizontal spread clearly. Since the error is very small the concentration parameters are displayed as a single value for all results presented hereafter.

Error estimation for the methane-hydrogen mixture was estimated from the individual errors of methane and hydrogen concentrations. The error in ratio of methane to hydrogen was estimated as follows

$$R = \frac{C_{CH4}}{C_{H2}}$$

$$R(\max) = \frac{C_{CH4}(\max)}{C_{H2}(\min)} \qquad \text{and} \qquad R(\min) = \frac{C_{CH4}(\min)}{C_{H2}(\max)}$$

$$2\Delta R = R(\max) - R(\min)$$

$$2\Delta R = \frac{C_{CH4}(\max)}{C_{H2}(\min)} - \frac{C_{CH4}(\min)}{C_{H2}(\max)}$$

$$2\Delta R = \frac{C_{CH4} + \Delta C_{CH4}}{C_{H2} - \Delta C_{H2}} - \frac{C_{CH4} - \Delta C_{CH4}}{C_{H2} + \Delta C_{H2}}$$

$$2\Delta R = \frac{C_{CH4}\left[1 + \dfrac{\Delta C_{CH4}}{C_{CH4}}\right]}{C_{H2}\left[1 - \dfrac{\Delta C_{H2}}{C_{H2}}\right]} - \frac{C_{CH4}\left[1 - \dfrac{\Delta C_{CH4}}{C_{CH4}}\right]}{C_{H2}\left[1 + \dfrac{\Delta C_{H2}}{C_{H2}}\right]}$$

Since $R = \dfrac{C_{CH4}}{C_{H2}}$ and $\dfrac{\Delta C_{CH4}}{C_{CH4}} = \dfrac{\Delta C_{H2}}{C_{H2}} = 0.005$

$$2\Delta R = \frac{R(1 + 0.005)}{(1 - 0.005)} - \frac{R(1 - 0.005)}{(1 + 0.005)}$$

$$2\frac{\Delta R}{R} = \frac{(1 + 2*0.005 + 0.005^2) - (1 - 2*0.005 + 0.005^2)}{1 - 0.005^2}$$

$$2\frac{\Delta R}{R} = \frac{4*0.005}{1 - 0.005^2}$$

Considering $(1 - 0.005^2) = 1$

$$\frac{\Delta R}{R} \cong 2*0.005$$

$$\frac{\Delta R}{R} \cong 0.01 \qquad \text{or} \qquad \frac{\Delta R}{R} \cong (2\Delta C) \qquad \qquad ...(4.16)$$

Accordingly the error in the ratio of the flammable gases is 1% (twice the gas concentration error). The error is still small compared to the gas concentration ratios considered in the experiments for the methane-hydrogen mixtures.

4.2.2 Error analysis for energy

The specifications of capacitors suggest that the capacitance values for each capacitor have around 10-15 % error. The actual capacitance of each capacitor was measured using RLC (Resistance Inductance and Capacitance) meter to arrive at the correct capacitance value. Table 4.2 presents the nominal and actual measured values of capacitors used in experiments.

Table 4.2 : Nominal and actual capacitance of the capacitors

Capacitor (nominal capacitance) (pF)	Actual capacitance measured by RLC meter (pF)
10	10.8
47	55.8
100	111.3
150	160
200	206
300	336

A Tektronix Oscilloscope model no. TDS 3034 B was used for measuring the voltage. The specifications of the voltage measuring instrument (oscilloscope associated with the high voltage probe) suggest that the error ($\pm \Delta V$) is 0.018% of the measured voltage.

Actual error in voltage measurement = $\pm \Delta V$

$$= \pm 0.00018 * V \qquad \text{volt}$$

A similar exercise was carried out for the errors in voltage (Table 4.3) and energy measurement, as was carried out during error analysis of gas concentration measurement. The results are shown in Table 4.3.

Table 4.3 : Error in voltage measurment

Measured parameter	Measurement error	Maximum / Upper value	Minimum / Lower value
Voltage	$\pm \Delta V$	$V_{max} = V + \Delta V$	$V_{min} = V - \Delta V$
Energy	$\pm \Delta E$	$E_{upper} = E + \Delta E$	$E_{lower} = E - \Delta E$

Energy $E = 0.5\,CP*V^2$

Error in the energy can be evaluated from upper and lower values :

$$2*\Delta E = E_{upper} - E_{lower}$$

Accordingly the upper and lower values of energy are

$$E_{upper} = 0.5CP*V^2_{max}$$

$$E_{lower} = 0.5CP*V^2_{min}$$

$$2*\Delta E = 0.5CP*V^2_{max} - 0.5CP*V^2_{min}$$

$$2*\Delta E = 0.5CP*\left(V^2_{max} - V^2_{min}\right)$$

$$2*\Delta E = 0.5CP*\left(V_{max} - V_{min}\right)*\left(V_{max} + V_{min}\right)$$

$$2*\Delta E = 0.5CP*\left(2\Delta V\right)*\left(2V\right)$$

$$2*\Delta E = 0.5CP*V^2\left(2*0.00018*V\right)*\left(2V\right)$$

$$2*\Delta E = 0.5CP*V^2\left(4*0.00018\right)$$

$$\Delta E = 0.5CP*V^2*\left(2*0.00018\right)$$

$$\frac{\Delta E}{E} = 0.00036 \qquad\qquad \text{or} \qquad\qquad \frac{\Delta E}{E} = (2\Delta V) \qquad\qquad ...(4.17)$$

Hence, the error for the energy measurement was calculated as twice the voltage error. i.e. 0.036% of the measured energy value. The error estimated for the ignition energy is very small as compared to the range of the energy values observed during experiments. The typical measurement errors for the methane test runs are shown in the Figure 4.5, by the vertical spread of the energy values plotted against the gas concentration. Indeed, the error was too small to be clearly seen on the plots, hence only a single value for the ignition energy is used for all results presented hereafter.

Figure 4.5 : Errors analysis for experiments with 6.5% & 8% methane concentrations

4.3 Results for methane

Experimental test runs with methane gas at the nominally lean (5%) and rich (15%) flammability limits never resulted in any successful ignitions. The concentration range of methane was then narrowed and ignition attempted with the highest spark energy. No ignitions were observed even after a hundred attempts outside the range 6% to 12% methane concentration. Experiments were then conducted with the target concentrations shown in Table 4.4. The corresponding equivalence ratio is also given in the same table.

Table 4.4 : Experimental conditions for methane tests

Parameter	Concentration of methane (% by volume)					
Observed range outside of which there were no ignitions	6	<------------------------------>				12
Target concentrations for methane experiments	--	7	8	9	11	--
Equivalence Ratio (ER)	0.61	0.72	0.83	0.94	1.06	1.30

Twenty different gas mixtures for each target concentration were prepared and ignition attempted up to 4 times (maximum) for each. The actual gas concentrations were calculated from the pressure transducer readings. A plot of the actual gas concentration prepared in each run and the observed ignitions (Yes) and non–ignitions (No) for a range of energies is presented in Figure 4.6.

Figure 4.6 : Results of ignition experiments for methane

The data is summarised in Table 4.5. An arithmetic average of the different nominal concentrations for the specified set is also presented. If the first ignition attempt was unsuccessful, the energy was progressively increased (by introducing a larger capacitor into the circuit), until the vessel contents were ignited or a maximum of four attempts had been made. The range of ignition energies; lowest to the largest, for the specified nominal concentration set, as observed during test runs is also presented. Nominal concentrations at the near stoichiometric (8% and 9%) compositions were almost all ignited with only a very small increase in energy value. Hence energies were not increased further for the 8% and 9% composition mixtures. Concentrations of lean (7%) and rich (11%) mixtures for the twenty samples (each) could not be ignited for the largest energy that could be generated inside the rig, even though a few ignitions were observed at lower energies. The range of ignition energies (lowest to largest) as observed for methane is presented in Table 4.5. Ignition probabilities calculate from this range of observed energies is discussed in Section 4.3.2.

There is a large variability in the ignition energy for 7% and 11% methane concentrations. The reasons for non-ignitions can be due to many reasons, including: the condition of electrode tips (covered with combustion products) may prevent the combustion, even though the sparks are visible across the electrodes. Similarly minor changes in temperature and humidity can also play an important role for deciding the success or failure of ignition. Moorhouse J. et. al. (1974) , Esseghir, M. and Polymeropoulos, C.E. (1988), Shepherd, J.E. et. al. (2000) and Randeberg E. (2006) observed similar variability in the ignition energy when working with their experimental rigs. All these parameters suggest a probabilistic element to the ignition energy as discussed in section 4.3.2.

Table 4.5 : Observed Ignition energies for various methane concentrations

Target concentration (%)	7	8	9	11
Actual Concentration (%) (Mean ± std. dev)	6.69 ±0.15	7.96 ±0.17	8.95 ±0.20	10.63 ±0.10
Actual Equivalence ratio (ER) (Mean ± std. dev)	0.68 ±0.015	0.82 ±0.018	0.94 ±0.022	1.13 ±0.011
Lowest observed Ignition energy for gases (mJ)	1.82	0.24	0.22	0.83
Range of observed Ignition energies for gases (mJ)	1.82 to 13.13	0.24 to 3.75	0.22 to 2.27	0.83 to 10.85

4.3.1 Comparison with previous data

Previous work on the ignition of quiescent methane-air mixtures at atmospheric pressures was examined. The ignition energy results of the present work are compared with Lewis & von Elbe, 1987, Bjerketvedt et. al. (1997) and calculated MIE data of Kim et. al. (2004) for methane in Figure 4.7 which shows the lowest observed ignition energy plotted against mean methane concentration. Kim et. al., 2004 have solved one dimensional unsteady state conservation equation numerically to arrive at the MIE values.

Figure 4.7 : Comparision of methane ignition energy results with those from the literature

The lowest observed ignition energies agree reasonably well with the reported data, except at the leanest mean concentration. The experimental method used by earlier researchers allows high voltage to be present on the electrodes for a considerable duration. Long duration of high voltage permits the condition of a corona discharge. High voltage before the spark can ionise the gas, reducing the dielectric strength of gas-air mixture. This in turn reduces the actual voltage at which spark occurs directly affecting the calculation of energy ($E = 0.5 \; CP^*V^2$). Thus the energy required for sparking process is reduced resulting in reporting of lower MIE value at the lean concentration. Lewis and von Elbe (1987) further introduced radium capsules of various strengths inside the bomb (experimental vessel) to reduce the time lag between the charging process (voltage elevation) and the onset of spark. The introduction of radium capsules may have lowered the MIE values. Ignitions at near stoichiometric are always guaranteed and hence these effects may not have impacted in lowering of MIE, which is in good agreement with this work.

4.3.2 Ignition probability

The success of ignitions quantified through the experiments was reported as ignition probability at the specified concentration of methane in air. Ignition probability in each of the 2 mJ energy bands was determined as described in Section 4.1.3 and the

computed values are presented in Table 4.6. The maximum ignition probability value was restricted to 99% to get finite value for the term '$Ln(1-IP)$'. There was an observed increase in the ignition probability from lower to higher energies, with higher energies resulting in more successful ignitions.

Table 4.6 : Ignition probability for various methane concentrations

Mean methane concentration	Percentage ignition probability for energy bands					
	0 - 2 mJ	2 - 4 mJ	4 - 6 mJ	6 - 8 mJ	8 - 10 mJ	10 - 12 mJ
CH_4 = 6.7%	5	20	59	69	89	89
CH_4 = 8%	29	82	99	99	99	99
CH_4 = 9%	49	99	99	99	99	99
CH_4 = 10.6%	15	47	75	68	88	94

The coefficients within the exponential function, Equation 4.8 (values 'b'and 'E_{min}') and the calculated ignition energy at 1% ignition probability using the Equation 4.10 is presented in Table 4.7. The lowest experimentally observed ignition energy is also shown for comparison with the $E_{IP=1\%}$ value.

Table 4.7 : Ignition energy at 1% ignition probability for methane

Mean methane concentration	Lowest observed ignition energy (mJ)	Value 'b'	Value E_{min} (mJ)	Energy $E_{IP=1\%}$ (mJ)
CH_4 = 6.7%	1.8163	0.2285	1.1803	1.2243
CH_4 = 8%	0.2402	0.6245	0.3339	0.3499
CH_4 = 9%	0.2242	1.7017	0.3874	0.3933
CH_4 = 10.6%	0.8349	0.2535	0.5538	0.5935

The value of E_{min} defines the energy level at a theoretical ignition probability of 'zero'. Whereas the more significant lowest value (often reported by various researchers) is at an ignition probability of 1% i.e. $E_{IP=1\%}$. The lowest observed ignition energy is based on the limited number of experiments carried out, whereas $E_{IP=1\%}$ (theoretically calculated), is based on one in a hundred chance of ignition.

A plot of ignition probability against energy as observed for methane–air is presented in Figure 4.8. Also shown in Figure 4.8 is the line representing the Equation 4.8 fitted to each data set.

Figure 4.8 : Ignition probability at various energies along with exponential trend lines for methane

4.4 Results for hydrogen

Similar to methane–air experiments, test runs with hydrogen–air at the lean (4%) limit never resulted in any successful ignitions. The concentration of hydrogen was increased and ignition attempted with the highest spark energy. The highest lean concentration below which no ignitions were observed even after a hundred attempts is presented in Table 4.8. The target concentration prepared for the experimental runs are also depicted in the Table 4.8. Since the concentration range examined for hydrogen was very wide seven different concentrations were targeted. The corresponding equivalence ratios are also given.

Table 4.8 : Experimental conditions for hydrogen

Parameter	Concentration of hydrogen (% by volume)							
Observed lean limit below which there were no ignition	5	<------------------------------------>						
Target concentrations for hydrogen experiments	--	6	10	20	25	30	45	60
Equivalence ratio (ER)	0.13	0.15	0.26	0.60	0.79	1.02	1.95	3.57

Experimental runs for different target concentrations for hydrogen were conducted similar to that for methane. A plot of the gas concentration prepared in each run and the observed ignitions (Yes) and non–ignitions (No) for a range of energies is presented in Figure 4.9.

Figure 4.9 : Ignition energies for a range of hydrogen concentrations

An arithmetic average of the different nominal concentrations was calculated and presented along with a mean and a standard deviation value in Table 4.9. The range of ignition energies as observed, is also presented in the same table. The range of ignition energies studied are narrower at the 20%, 25% and 30% concentrations as these mixtures are easily ignited at low energy levels (near stoichiometric concentration of hydrogen-air mixtures).

Table 4.9 : Observed Ignition energies for various hydrogen concentration

Nominal concentration (%)	6	10	20	25	30	45	60
Actual concentration (%) (Mean ± std. dev)	6.32 ±0.44	10.65 ±0.89	20.96 ±0.74	25.47 ±0.64	30.38 ±0.94	45.68 ±1.19	62.11 ±1.83
Equivalence ratio (ER) (Mean ± std. dev)	0.16 ±0.01	0.28 ±0.03	0.63 ±0.03	0.81 ±0.03	1.04 ±0.05	2.00 ±0.10	3.90 ±0.30
Lowest observed Ignition energy for gases (mJ)	0.76	0.10	0.04	0.03	0.02	0.06	0.22
Range of observed Ignition energies for gases (mJ)	0.76 to 11.96	0.10 to 4.63	0.04 to 0.87	0.03 to 0.36	0.02 to 0.24	0.06 to 0.56	0.22 to 4.96

4.4.1 Comparison with Previous data

Previous work on the ignition of quiescent hydrogen-air mixtures at atmospheric pressures was examined. The ignition energy results of the present work are compared with Lewis & von Elbe, 1987 and D.W.V. (German Hydrogen Association, 2002) for hydrogen. Further the MIE values were also compared with experimental work of Ono et. al. (2007) and Kim et. al. (2004)

The experimental results of Ono et. al were aimed at understanding the influence of spark gap length on the MIE values. The lowest values of MIE were obtained with spark gap of 0.5 mm. MIE increases with increasing distance between spark gap. The minimum spark gap delivers the lowest MIE which are compared with the present work.

Kim et. al., 2004 have solved one dimensional unsteady state equation numerically for hydrogen-air mixture to arrive at the MIE values. Detail chemical kinetic mechanisms are used to predict the MIE for hydrogen-air mixtures. The results showed that MIE is a sensitive function of the radius of ignition energy source considered as half of quenching distance and the duration of supplied energy as 0.52 μs. The MIE values at ambient pressure as reported by Kim et. al are compared with the present work.

Figure 4.10 : Comparision of hydrogen igntion energy results with those from the literature

The lowest ignition energies for hydrogen have a 'U' shape dependence on concentration (similar to that of methane). The energy increases asymptotically at the lean and rich limits. The lowest observed values of the ignition energy agree reasonably well with the reported data.

4.4.2 Ignition probability

The success of ignitions quantified through the experiments was used to derive ignition probabilities for each concentration of hydrogen gas. Different energy bandwidths were selected for the different concentrations. At the leanest concentration (6%), there were not many ignitions in each bandwidth, therefore a 4 mJ bandwidth was selected and the results are presented in the Table 4.10. At near stoichiometric (20 – 30%) concentration and also at 45%, ignition was achieved easily at low energy levels, hence a 0.5 mJ bandwidth was selected as presented in Table 4.11. For other concentrations 10% and 60% an energy bandwidth of 1 mJ was considered appropriate as shown in Table 4.12. Ignition probability in each of these individual bands was determined as described in Section 4.1.3. The results of the 20%, 25% and 30% were grouped together when drawing the exponential curve fit line.

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Table 4.10 : Ignition probability for very lean (6%) hydrogen concentration

Mean Hydrogen concentration	Percentage ignition probability for energy bands		
	0 – 4 mJ	4 – 8 mJ	8 – 12 mJ
H_2 = 6.32%	3	10	15

Table 4.11 : Ignition probability for 20% to 45% hydrogen concentration

Mean Hydrogen concentration	Percentage ignition probability for energy bands	
	0 – 0.5 mJ	0.5 – 1 mJ
H_2 = 20%, 25% and 30 %	75	99
H_2 = 45.68%	61	99

Table 4.12 : Ignition probability for lean (10%) and rich (60%) H_2 concentration

Mean Hydrogen concentration	Percentage ignition probability for energy bands				
	0 – 1 mJ	1 – 2 mJ	2 – 3 mJ	3 – 4 mJ	4 - 5 mJ
H_2 = 10.65%	22	62	79	89	95
H_2 = 62.11%	17	42	77	84	91

The coefficients of the exponential function as described in equation 4.8 (values 'b' and 'E_{min}') and the calculated ignition energy at 1% ignition probability using the Equation 4.10 is presented in Table 4.13. The lowest experimentally observed ignition energy is also shown for comparison with the $E_{IP=1\%}$ value.

Table 4.13 : Ignition energy at 1% ignition probability for hydrogen

Mean Concentration	Lowest observed ignition energy (mJ)	Value 'b'	Value E_{min} (mJ)	Energy $E_{IP=1\%}$ (mJ)
H_2 = 6.32%	0.7607	0.0173	0.3815	0.9624
H_2 = 10.65%	0.1049	0.6744	0.1481	0.1630
H_2 = 20% – 30%	0.0244	6.3469	0.0287	0.0302
H_2 = 45.68%	0.0576	6.7521	0.0841	0.0856
H_2 = 62.11%	0.2242	0.5632	0.2317	0.2450

The lowest observed ignition energy is based on the limited number of experiments carried out, whereas $E_{IP=1\%}$, is the one in hundred chance of ignition. A constant ignition energy value ($E_{IP=1\%}$) for 20%, 25% and 30% is in agreement of D.W.V data presented in Figure 4.10.

A plot of ignition probability against energy as observed for hydrogen gas is presented in the Figure 4.11. Also shown in Figure 4.11 is the line representing Equation 4.8 fitted to each data set.

Figure 4.11 : Ignition probability at various energies along with exponential trend lines for hydrogen

4.5 Results for methane hydrogen mixtures

4.5.1 Gas with 75 % methane and 25 % hydrogen

The flammable range for the gas mixture (methane with 25% hydrogen) was initially determined using the Le Chatelier's equation (see Table 3.2). The lowest lean concentration practically achievable was 4% flammables in air (3% methane and 1% hydrogen). It was quite evident that, since this concentration was below the LFL given in Table 3.2 (4.7%), ignition was not possible. Experiments performed with the next practically achievable concentration, 8% flammables in air (6% methane and 2% hydrogen) resulted in ignitions, hence this was considered as the lowest practical lean concentration. The rich limit where no ignitions were observed even after a hundred attempts and the target concentration are presented in the Table 4.14.

Table 4.14 : Experimental conditions for 75% CH_4 + 25% H_2 mixture

Parameter	Concentration of flammable gas mixture (% vol.)											
Gases	CH_4	H_2	CH_4	H_2	CH_4	H_2	CH_4	H_2	CH_4	H_2	CH_4	H_2
End limits (no ignition)	3	1	<--->								15	5
Total (CH_4 + H_2)	4						.				20	
Target concentration	--	--	6	2	8	3	10	3	12	4	--	--
Total (CH_4 + H_2)	---		8		11		13		16		---	
Equivalence ratio (ER)	0.44		0.67		0.94		1.18		1.5		1.6	

Gas mixtures at the target concentrations were prepared and the actual gas concentrations determined from the partial pressures of methane and hydrogen as described in Section 4.1.1. A plot of the observed energies, ignitions (Yes) and non-ignitions (No) against the concentration are presented in Figure 4.12.

Figure 4.12 : Ignition energies for 75% CH_4 + 25% H_2 mixture

An arithmetic average of the total flammable concentration (75 % CH4 + 25 % H2 mixture) was calculated for each nominal specified data set, and presented in Table 4.15. The lowest ignition energy and the range of ignition energies observed for each concentration are also presented in the same table.

Table 4.15 : Observed Ignition energies for 75% CH_4 + 25% H_2 mixture

Gases	CH_4	H_2	CH_4	H_2	CH_4	H_2	CH_4	H_2
Targeted concentration (%)	6	2	8	3	10	3	12	4
Actual concentration of flammable gas mixture (%)	6.01	2.01	8.03	2.64	10.1	3.39	12.42	4.28
Ratio (CH_4 : H_2) Mean ± std. dev.		3.02 ±0.31		3.07 ±0.34		2.99 ±0.25		2.91 ±0.19
Total conc. (CH_4 + H_2) Mean ± std. dev.		8.02 ±0.32		10.67 ±0.39		13.49 ±0.42		16.70 ±0.39
Equivalence ratio (ER) Mean ± std. dev.		0.67 ±0.31		0.93 ±0.34		1.20 ±0.25		1.54 ±0.19
Lowest Ignition energy for gas mixture (mJ)		0.58		0.10		0.52		5.19
Range of Ignition energies for gas mixture (mJ)		0.58 to 5.05		0.10 to 2.32		0.52 to 9.63		5.19 to 11.74

4.5.2 Ignition probability of 75% CH₄ + 25% H₂ mixture

Ignition probabilities for the flammable gas mixtures were calculated in a similar way to that used for the methane tests. The entire energy range (Minimum to Maximum) was divided into bands of 2 mJ each. The ignition probability in each of these individual bands was determined as described in Section 4.1.3 and the computed values are presented in Table 4.16.

Table 4.16 : Ignition probability for 75% CH₄ + 25% H₂ mixture

Total flammables and $CH_4 : H_2$	Percentage ignition probability for energy bands					
	0 - 2 mJ	2 - 4 mJ	4 - 6 mJ	6 - 8 mJ	8 - 10 mJ	10 - 12 mJ
8.01 (6.01:2.00)	5	94	99	99	99	99
10.67 (8.03: 2.64)	73	99	99	99	99	99
13.49 (10.10:3.39)	10	56	79	94	99	99
16.70 (12.42:4.28)	0	0	7	15	40	55

The coefficients of the exponential function (values 'b' and 'E_{min}') as described in Equation 4.8 and the calculated ignition energy at 1% ignition probability using the Equation 4.10 is presented in Table 4.17. The lowest experimentally observed ignition energy is also shown for comparison with $E_{IP=1\%}$ value.

Table 4.17 : Energy at 1% ignition probability for 75% CH₄ + 25% H₂ mixture

Concentration Total $(CH_4 : H_2)$	Lowest observed ignition energy (mJ)	Value 'b'	Value E_{min} (mJ)	Energy $E_{IP=1\%}$ (mJ)
8.02 (6.01: 2.01)	0.5840	1.0115	0.3499	0.2368
10.67 (8.03 : 2.64)	0.1049	1.603	0.1591	0.1591
13.49 (10.10 : 3.39)	0.5160	0.5135	1.0365	1.1608
16.70 (12.42 : 4.28)	5.1900	0.1260	4.973	5.05278

The lowest experimentally observed ignition energies (column 2) are comparable with the $E_{IP=1\%}$ values, which are based on one in hundred chance of ignition.

Figure 4.13 : Ignition probability at various energies along with exponential trend lines for 75% CH₄ + 25% H₂ mixture

A plot of ignition probability against ignition energy as observed for the gas mixture is presented in the Figure 4.13 together with the exponential curves fitted to the data. Similar to the methane tests there was an observed increase in the ignition probability from lower to higher energies, with higher energies resulting in more successful ignitions.

4.5.3 Gas with 50% methane and 50% hydrogen

The flammable range for the gas mixture (methane with 50% hydrogen) was initially determined using the Le Chatelier's equation (see Table 3.2). The lowest lean flammability limit practically achievable was 4% flammables in air (2% methane and 2% hydrogen). It was quite evident that, since this concentration was below the LFL given in Table 3.2 (4.4 %), ignition was not possible. Experiments performed with the next practically achievable concentration, 6% flammables in air (3% methane and 3% hydrogen) resulted in ignitions, hence this was considered as the lowest practical lean concentration. The rich limit where no ignition were observed even after a hundred attempts and the target concentration are presented in Table 4.18. Since the flammability range was wider than for 75% CH₄ + 25% H₂ gas mixture, five different gas concentrations were targeted within this range.

Table 4.18 : Experimental conditions for 50% CH$_4$ + 50% H$_2$ mixture

	Concentration of flammable gas mixture													
Gases	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$
End limits (no ignition)	2	2	<--->										12	12
Total (CH$_4$ + H$_2$)	4												24	
Targeted conc.	--	--	3	3	5	5	7	7	8	8	10	10	--	--
Total (CH$_4$ + H$_2$)	---		6		10		14		16		20		---	
ER	0.25		0.38		0.66		0.97		1.13		1.49		1.88	

Gas mixtures at the target concentrations were prepared and the actual gas concentrations determined from the partial pressures of methane and hydrogen as described in Section 4.1.1. A plot of observed energies, ignitions (Yes) and non-ignitions (No) against concentration are presented in Figure 4.14.

Figure 4.14 : Ignition energies for 50% CH$_4$ + 50% H$_2$ mixture

An arithmetic average of the total flammable concentration (50% CH$_4$ + 50% H$_2$ mixture) was calculated for each nominal specified data set, and presented in Table 4.19. The lowest ignition energy and the range of ignition energies observed for each concentration are also presented in Table 4.19.

Table 4.19 : Observed Ignition energies for 50% CH$_4$ + 50% H$_2$ mixture

Gases	Concentration of flammable gas mixture									
	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$
Targeted conc.	3	3	5	5	7	7	8	8	10	10
Actual conc. of flammable gases	3.01	3.13	4.99	5.04	6.80	6.68	8.09	8.07	10.10	10.00
Ratio (CH$_4$: H$_2$) Mean ± Std. dev	0.97 ±0.10		0.99 ±0.09		1.02 ±0.03		1.00 ±0.04		1.01 ±0.05	
Total conc. Mean ± Std. dev	6.14 ±0.10		10.03 ±0.09		13.48 ±0.03		16.16 ±0.04		20.10 ±0.05	
ER Mean ± Std. dev	0.39 ±0.03		0.69 +0.03		0.93 +0.03		1.15 +0.03		1.50 +0.06	
Lowest Ignition energy for gas mixture (mJ)	2.72		0.16		0.09		0.10		4.11	
Range of Ignition energies for gas mixture (mJ)	2.72 to 13.34		0.16 to 3.16		0.09 to 2.31		0.10 to 2.98		4.11 to 11.40	

4.5.4 Ignition probability of 50% CH$_4$ + 50% H$_2$ mixture

Ignition probabilities for the flammable gas mixtures were calculated in a similar way to that used for the methane tests. The entire energy range (Minimum to Maximum) was divided into bands of 2 mJ each. The ignition probability in each of these individual bands was determined as described in Section 4.1.3 and the computed values are presented in Table 4.20.

Table 4.20 : Ignition probability for 50% CH$_4$ + 50% H$_2$ mixture

Total flammables and CH$_4$: H$_2$	Percentage ignition probability for energy bands					
	0 - 2 mJ	2 - 4 mJ	4 - 6 mJ	6 - 8 mJ	8 - 10 mJ	10 - 12 mJ
6.14 (3.01: 3.13)	0	11	29	43	54	71
10.03 (4.99 : 5.04)	67	99	99	99	99	99
13.48 (6.80 : 6.68)	72	99	99	99	99	99
16.16 (8.09 : 8.07)	65	99	99	99	99	99
20.10 (10.1 : 10.0)	0	0	11	18	38	50

The coefficients of the exponential function as described in Equation 4.8 (values 'b' and 'E_{min}') and the calculated ignition energy at 1% ignition probability using the equation 4.10 is presented in Table 4.21. The lowest experimentally observed ignition energy is also shown for comparison with $E_{IP=1\%}$ value. As can be seen they are almost of the same magnitude.

Table 4.21 : Energy at 1% ignition probability for 50% CH$_4$ + 50% H$_2$ mixture

Concentration Total (CH$_4$: H$_2$)	Lowest observed ignition energy (mJ)	Value 'b'	Value E_{min} (mJ)	Energy $E_{IP=1\%}$ (mJ)
6.14 (3.01: 3.13)	2.7209	0.1456	2.6353	2.7043
10.03 (4.99 : 5.04)	0.1590	1.6527	0.2630	0.2690
13.48 (6.80 : 6.68)	0.0894	1.606	0.1633	0.1695
16.16 (8.09 : 8.07)	0.1044	1.6221	0.2190	0.2252
20.10 (10.1 : 10.0)	4.1084	0.0975	4.1733	4.2764

A plot of ignition probability again energies as observed for the gas mixture is presented in the Figure 4.15 together with the exponential curves fitted to the data.

Figure 4.15 : Ignition probability at various energies along with exponential trend lines for 50% CH_4 + 50% H_2 mixture

4.5.5 Gas with 25% methane and 75% hydrogen

The flammable range for the gas mixture (methane with 75% hydrogen) was initially determined using the Le Chatelier's equation (see Table 3.2). The lowest lean flammability concentration practically achievable was 4% flammables in air (1% methane and 3% hydrogen) as was the case with 75% CH_4 + 25% H_2 mixture described in Section 4.5.1. It was quite evident that, since this concentration was below the LFL given in Table 3.2 (4.2%), ignition was not possible. Experiments performed with the next practically achievable concentration, 8% flammables in air (2% methane and 6% hydrogen) resulted in ignitions, hence this was considered as the lowest practical lean concentration. The rich limit where no ignitions were observed even after a hundred attempts and the target concentration as prepared for the experimental runs is presented in Table 4.22. Five different gas concentrations were targeted within this range, similar to the 50% CH_4 + 50% H_2 mixture.

Table 4.22 : Experimental conditions for 25% CH$_4$ + 75% H$_2$ mixture

Parameter	Concentration of flammable gas mixture													
	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$
End limits (no ignition)	1	3				<--->							9	27
Total (CH$_4$ + H$_2$)	4												38	
Targeted conc.	--	--	2	6	3	10	4	13	5	16	7	20	--	--
Total (CH$_4$ + H$_2$)	---		8		13		17		21		28		---	
ER	0.17		0.36		0.60		0.83		1.08		1.57		2.34	

Gas mixtures at the target concentrations were prepared and the actual gas concentrations determined from the partial pressures of methane and hydrogen as described in Section 4.1.1. A plot of observed energies, ignitions (Yes) and non-ignitions (No) against concentration are presented in Figure 4.16.

Figure 4.16 : Ignition energies for 25% CH$_4$ + 75% H$_2$ mixture

An arithmetic average of the total flammable concentration (25% CH$_4$ + 75% H$_2$ mixture) was calculated from each nominally specified dataset and presented in Table 4.23. The lowest ignition energy and the range of ignition energies observed for each concentration are also presented in Table 4.23.

Table 4.23 : Observed Ignition energies for 25% CH$_4$ + 75% H$_2$ mixture

Parameter	Concentration of flammable gas mixture									
	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$	CH$_4$	H$_2$
Targeted conc.	2	6	3	10	4	13	5	16	7	20
Actual conc. of flammable gases	2.27	6.16	3.5	10.14	4.56	13.34	5.59	15.84	6.38	20.39
Ratio (CH$_4$: H$_2$) Mean ± Std. dev.	8.43 ±0.04		13.64 ±0.03		17.90 ±0.02		21.43 ±0.02		27.22 ±0.01	
Total conc. Mean ± Std. dev.	8.43 ±0.32		13.64 ±0.41		17.90 ±0.47		21.43 ±0.49		27.22 ±0.53	
ER Mean ± Std. dev.	0.40 ±0.03		0.67 ±0.03		0.92 ±0.04		1.16 ±0.05		1.56 ±0.05	
Lowest ignition energy for gas mixture (mJ)	2.06		0.10		0.07		0.11		1.24	
Range of Ignition energies for gas mixture (mJ)	2.06 to 12.42		0.10 to 2.59		0.07 to 1.73		0.11 to 2.64		1.24 to 11.74	

4.5.6 Ignition probability of 25% CH$_4$ + 75% H$_2$ mixture

Ignition probabilities for the flammable gas mixtures were calculated in a similar way to that used for the methane tests. The entire energy range (Minimum to Maximum) was divided into bands of 2 mJ each. The ignition probability in each of these individual bands was determined as described in Section 4.1.3 and the computed values are presented in Table 4.24.

Table 4.24 : Ignition probability for 25% CH_4 + 75% H_2 mixture

Total flammables and CH_4 : H_2	Percentage ignition probability for energy bands					
	0 - 2 mJ	2 - 4 mJ	4 – 6 mJ	6 - 8 mJ	8 - 10 mJ	10 - 12 mJ
8.43 (2.27 : 6.16)	0	8	38	63	69	75
13.63 (3.50 : 10.14)	69	99	99	99	99	99
17.90 (4.56 : 13.34)	74	99	99	99	99	99
21.43 (5.59 : 15.84)	65	99	99	99	99	99
27.22 (6.84 : 20.39)	5	29	55	75	85	88

The coefficients of exponential function (values 'b'and 'E_{min}') as described in Equation 4.8 and the calculated ignition energy at 1% ignition probability using Equation 4.10 is presented in Table 4.25. The lowest experimentally observed ignition energy is also shown for comparison with $E_{IP=1\%}$. As can be seen they are almost of the same magnitude.

Table 4.25 : Energy at 1% Ignition probability for 25% CH_4 + 75% H_2 mixture

Concentration Total (CH_4 : H_2)	Lowest observed ignition energy (mJ)	Value 'b'	Value E_{min} (mJ)	Energy $E_{IP=1\%}$ (mJ)
8.43 (2.27 : 6.16)	2.0639	0.1531	1.5336	1.5993
13.63 (3.50 : 10.14)	0.1049	1.6131	0.1882	0.1944
17.90 (4.56 : 13.34)	0.0663	1.5844	0.1161	0.1224
21.43 (5.59 : 15.84)	0.1115	1.6221	0.2190	0.2252
27.22 (6.84 : 20.39)	1.2358	0.2240	1.2063	1.2511

Similar to the methane tests there was an observed increase in the ignition probability from lower to higher energies, as presented in the Figure 4.17. Higher magnitude of energies resulting in more successful ignitions. The exponential curves fitted to the data are also shown in Figure 4.17.

Figure 4.17 : Ignition probability at various energies along with exponential trend lines for 25% CH_4 + 75% H_2 mixture

4.6 Overall results

Sections 4.3 to 4.5 shows that an exponential function, Equation 4.8, can be used to represent the variation in ignition probability with energy for all the gas-air mixtures studied. Values of b and E_{min} were identified for the gas-air mixture and for each set of flammable gas concentration. The value of E_{min} represents the hypothetical energy for which there is no possibility of ignition (Ignition probability has theoretically zero value). Any small increase in the ignition energy above E_{min} gives a small probability of ignition. Therefore, the values of E_{min} represent the hypothetical minimum ignition energies for the fuel-air mixtures studied. The values of b, in combination with the corresponding values of E_{min}, define the shapes of the ignition probability-energy curves.

The consistency of the experimentally obtained ignition probability was compared with the calculated value of ignition probability using Equation 4.8. The values of

b and E_{min} utilised in Equation 4.8 are derived for each fuel-air mixture. This comparison is shown in Figure 4.18.

Figure 4.18 : Observed Ignition probability plotted against the calculated value

Figure 4.18 shows that for the range of gas-air mixtures studied, the ignition probability is well represented by Equation 4.8 and that this equation could be used to determine the ignition probability at a particular ignition energy if the relevant values of b and E_{min} are known. This is further discussed in the next section.

4.6.1 Ignition energy

The overall experimental results for the variation of lowest ignition energy with equivalence ratio for the range of flammable gases studied (100% CH_4, 75% CH_4 + 25% H_2, 50% CH_4 + 50% H_2, 25% CH_4 + 75% H_2, and 100% H_2) are presented in Figure 4.19.

Figure 4.19 : Lowest ignition energy at various ER of flammable gases

The nature of the curve for all the gases indicates that, it may be possible to collapse the data into a single curve. Accordingly the Equivalence Ratio (ER) and the ignition energy for lean and rich flammable gas–air mixtures were normalised to form dimensionless parameters as follows:

Lean mixture

$$Normalised\ ER\ (NER) = \frac{ER - ER_{MIE}}{ER_{MIE} - ER_{LFL}} \qquad (when\ ER\ <\ ER_{MIE}) \qquad ...(4.18)$$

Rich mixture

$$Normalised\ ER\ (NER) = \frac{ER - ER_{MIE}}{ER_{UFL} - ER_{MIE}} \qquad (when\ ER\ >\ ER_{MIE}) \qquad ...(4.19)$$

Where

ER_{MIE} Equivalence Ratio of the gas at the MIE value

ER_{LFL} Equivalence Ratio at the Lower Flammability Limit

ER_{UFL} Equivalence Ratio at the Upper Flammability Limit

The Ignition Energy (IE) for each gas was normalised by using the MIE value as follows:

$$Normalised\ IE = \frac{IE}{MIE} \qquad \qquad ...(4.20)$$

The lean and the rich Equivalence Ratios with their corresponding ignition energy values are considered separately. Logarithmic values of normalised IE are plotted against the normalised ER (for lean values) for all the gases under consideration and is presented in the Figure 4.20. All the test results of methane, hydrogen and the methane-hydrogen mixtures (25%, 50% and 75% hydrogen in methane) were collated and the data points were plotted from the most lean mixture (ER < ER_{MIE}) to ER equals ER_{MIE}, i.e. NER between the range -1 to 0. Mirror images of the lean normalised ER values (i.e. for NER between the range 0 to 1) were then plotted along with the normalised ignition energy. Curve fitting was done for the entire set of data points (including the mirror image points) with an intercept at the origin. This type of curve fitting achieves a zero slope at the origin. The lowest point on the curve is described by 'NER = 0' and the corresponding ordinate of the lowest point i.e. normalised ignition energy = 1 describes the MIE for the gas under consideration.

Ignition energy data at various ER values for Ethane and Propane gas (Lewis and von Elbe, 1987) were also analysed in similar way. The normalised IE for Ethane and Propane are also plotted and presented in the same Figure 4.20. The data analysis for all these gases (CH_4, H_2, CH_4 + H_2 mixtures, C_2H_6 and C_3H_8) suggest that the ignition energy for flammable gases can be collapsed through such dimensionless parameters and the data correlates well for the lean compositions.

Figure 4.20 : Normalised ignition energy vs normalised ER for lean gas composition

A second degree polynomial equation fitted for the lean values gives.

$Ln(Normalised\ IE) = 3.2342 * NER * NER + 2\ x\ 10^{-17} * NER$

$Normalised\ IE = \exp\left(3.2342 * NER * NER + 2\ x\ 10^{-17} * NER\right)$

$IE = MIE * \exp\left(3.2342 * NER * NER\right)$ ...(4.21)

A similar exercise was carried out for the rich mixtures and the variation of normalised Ignition Energy against normalised ER is presented in the Figure 4.21. The rich mixtures were plotted initially and then mirror images for the normalised ER values were plotted. The best fit second degree polynomial equation for the rich gas mixture is

$Ln(Normalised\ IE) = 10.119 * NER * NER + 5\ x\ 10^{-17} * NER$

$Normalised\ IE = \exp\left(10.119 * NER * NER + 5\ x\ 10^{-17} * NER\right)$

$IE = MIE * \exp\left(10.119 * NER * NER\right)$ ...(4.22)

The curve fitting for the rich values also gives a zero slope at the origin, similar to the lean values. The ordinate of the lowest point (normalised ignition energy = 1) describes the MIE for the gas under consideration.

Figure 4.21 : Normalised Ignition energy vs normalised ER for rich gas composition

The trend line for the entire range (lean to rich mixture composition) is presented in the Figure 4.22. Data points for Ethane and Propane gas are also plotted on the same graph, suggesting good agreement with the correlated equation of the trend line.

Figure 4.22 : Normalised Ignition energy vs normalised ER for all gases

A correlation to determine the minimum ignition energy in Equations 4.21 and 4.22 can also be obtained in terms of the concentration of hydrogen in the methane–hydrogen mixture as presented in Figure 4.23.

Figure 4.23 : MIE variation for various concentration of hydrogen in mixture

The reported values of MIE for methane are from Lewis & Von Elbe, 1987 and Bjerketvedt et. al., 1997. An average value of the literature data is presented as single value in the in Figure 4.23. Similarly for hydrogen the average value of MIE from Lewis and D.W.V (German hydrogen association) data is presented. The exponential curve fit line suggests an equation for MIE as follows:

$$MIE = 0.2433 * \exp\left(-0.221 * C_{H2}\right) \qquad \ldots(4.23)$$

Where C_{H2} is the percentage of hydrogen in the methane-hydrogen mixture.

A generalised correlation to estimate the ignition energy for a methane-hydrogen-air mixture was derived from the test results and the range of observed data. The equation for calculating ignition energy for a methane-hydrogen-air mixture (including those of pure methane (when C_{H2} = 0) and pure hydrogen (when C_{H2} = 100) obtained through the correlation is:

For lean gas mixtures

$$IE = MIE * \exp(3.2342 * NER * NER) \qquad (when \ ER \ < \ ER_{MIE})$$

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$$IE = 0.2433 * [\exp(-0.221 * C_{H2})] * \exp(3.2342 * NER * NER) \qquad \ldots(4.24)$$

For rich gas mixtures

$$IE = MIE * \exp(10.119 * NER * NER) \qquad (when \ ER > ER_{MIE})$$

$$IE = 0.2433 * [\exp(-0.221 * C_{H2})] * \exp(10.119 * NER * NER) \qquad \ldots(4.25)$$

Equations 4.24 and 4.25, above presents the generalised correlation based on the experimental observations for the methane-hydrogen gas mixture. The equations can also be used to calculate the ignition energy for pure gases methane and hydrogen at various concentrations. However the equations are restricted to methane, hydrogen and mixtures of methane and hydrogen.

The equivalence ratio at MIE for the flammable gases (methane, hydrogen, and methane–hydrogen mixtures) was then correlated to the molecular weight of the flammable gas. A plot of the Equivalence Ratio at MIE (ER_{MIE}) against Molecular Weight (MW) is presented in Figure 4.24 for the methane-hydrogen system studied.

Figure 4.24 : Equivalence Ratio variation with molecular weight of flammable gas

The value of ER at the MIE for the gases (methane, hydrogen and methane-hydrogen mixtures) can be calculated from the Molecular Weight (MW) of the flammable gas using the following equation.

$$ER_{MIE} = 0.0014 * MW * MW - 0.0307 * MW + 1.081 \qquad \ldots(4.26)$$

Equations 4.24, 4.25 and 4.26 are restricted to the methane hydrogen system. Figure 4.25 presents analysis of the data considering ethane and propane along with methane-hydrogen system.

Figure 4.25 : Equivalence Ratio variation with molecular weight of flammable gas including ethane and propane gas

Based on Figure 4.25 the coefficients of Equation 4.26 modify to the following :

$$ER_{MIE} = 0.0004 * MW * MW - 0.012 * MW + 1.0338 \qquad \ldots(4.27)$$

The analysis was extended further, to establish relationship between the minimum ignition energy and the laminar burning velocity. The variation in MIE for gases were plotted against the reported values of laminar burning velocity for hydrogen, methane (Lewis and von Elbe, 1987) and predicted values (based on a correlation proposed by Huanga, et. al. 2006) for methane-hydrogen mixtures. The reported values of laminar burning velocity (Su) of fuel-air mixtures near the stoichiometric composition

were utilised. The best fit curve through the data points is presented in Figure 4.26. Accordingly the equation to calculate MIE for these gases is

$$MIE = 0.0693 * Su^{-1.3028}$$
...(4.28)

Figure 4.26 : MIE variation with Laminar burning velocity

Other hydrocarbons ethane, propane, n-butane, pentane, hexane acetylene, ethylene and propylene also satisfy the trend line equation as shown in the Figure 4.26.

A generalised equation to calculate ignition energy for gas mixtures can be obtained from Equation 4.21 and Equation 4.22 as follows:

$$IE = 0.0693 * Su^{-1.3028} * \exp(3.2342 * NER * NER) \qquad (when \ ER < ER_{MIE}) \qquad ...(4.29)$$

$$IE = 0.0693 * Su^{-1.3028} * \exp(10.119 * NER * NER) \qquad (when \ ER > ER_{MIE}) \qquad ...(4.30)$$

A plot of ignition energy as calculated through the equations above and the actual observations is presented in Figure 4.27. The actual observation agrees well with the correlated equations for all the gases methane, hydrogen and methane - hydrogen mixtures. Good agreement is also obtained for ethane and propane. The lines are plotted as per the equations for ignition energy (Equation 4.29 and Equation 4.30) and the points are the actual experimental observations.

Figure 4.27 : Ignition energy for gases at various Equivalence Ratios

The consistency of the approach taken to determine the minimum ignition energy and the variation in lowest ignition energy with equivalence ratio is illustrated by plotting the observed and predicted ignition energy. This comparison is shown in Figure 4.28.

Figure 4.28 : Observed and predicted values of Ignition energy

4.6.2 Prediction of Ignition probability

After the success of the use of the correlations obtained from the experimental results to predict the ignition energy illustrated above, attention was given to the ignition probability. The relationship between b, E_{min} (energy at zero ignition probability) and the lowest ignition energy (as referred in Equation 4.8) was examined.

Figure 4.29 : Variation of parameter 'b' with Lowest Ignition energy

As shown in Figure 4.29, the relationship between the value of 'b' and the lowest ignition energy is given by:

$$b = 0.3209 * IE^{(-0.7235)} \qquad\qquad ...(4.31)$$

Figure 4.30 : Variation of parameter 'E_{min}' against Lowest Ignition energy

The relationship between E_{min} and the lowest ignition energy (as shown in Figure 4.30) is:

$$E_{min} = 0.95 * IE \qquad\qquad ...(4.32)$$

The Ignition probability (IP) equation after substitution of parameters 'b' and 'E_{min}' is

$$IP = 1 - \exp\left[-0.3209 * IE^{-0.7235} * (E - 0.95 * IE)\right]$$

Referring to Equation 4.21 and 4.22 ignition probability can be calculated as

<u>For lean mixtures</u> $(ER < ER_{MIE})$

$$IE = MIE * \exp(3.2342 * NER * NER)$$

Hence

$$IP = 1 - \exp\left[\begin{array}{l} -0.3209 * \{MIE * \exp(3.2342 * NER * NER)\}^{-0.7235} \\ * (E - 0.95 * \{MIE * \exp(3.2342 * NER * NER)\}) \end{array}\right] \qquad ...(4.33)$$

Where $NER = \dfrac{ER - ER_{MIE}}{ER_{MIE} - ER_{LFL}}$

ER_{LFL} Equivalence Ratio at the Lower Flammability Limit

$$ER_{MIE} = 0.0014 * MW * MW - 0.0307 * MW + 1.081$$

$$MIE = 0.0693 * Su^{-1.3028}$$

<u>For Rich mixtures</u> $(ER > ER_{MIE})$

$$IE = MIE * \exp(10.119 * NER * NER)$$

Hence

$$IP = 1 - \exp\left[\begin{array}{l} -0.3209 * \{MIE * \exp(10.119 * NER * NER)\}^{-0.7235} \\ * (E - 0.95 * \{MIE * \exp(10.119 * NER * NER)\}) \end{array}\right] \qquad ...(4.34)$$

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Where $NER = \dfrac{ER - ER_{MIE}}{ER_{UFL} - ER_{MIE}}$

ER_{UFL} Equivalence Ratio at the Upper Flammability Limit

MIE and ER_{MIE} has same meaning as referred for lean mixtures.

As per Equation 4.33 and 4.34 above, IP is a function of Energy (E in mJ) and Equivalence Ratio (ER) at lean and rich flammable gas concentrations.

Accordingly in this section a generalised correlation for the ignition energy (Equation 4.21 and Equation 4.22) and Ignition probability (referred above) was developed from the test results of the flammable gases. The generalised ignition energy equation was also found useful for other flammable gases like ethane and propane as presented in the Figure 4.22. Ignition probability can not be compared for other gases, since data for other gases are not available. The overall result of the ignition energy tests are further discussed based on ignition probability at various energies and the impact of hydrogen on MIE of gases.

4.6.3 Ignition probability computation

The Ignition probability calculated from the Equation 4.8 for fixed energy levels (2 mJ, 5 mJ and 10 mJ) and at a three different equivalence ratios (lean, near stoichiometric and rich) for the flammable gases are presented in Table 4.26.

Table 4.26 : Ignition probability of flammable gases at various energies

Gases	Equivalence ratio	Ignition probability at energies of		
		2 mJ	5 mJ	10 mJ
Methane		0.17	0.58	0.87
75% CH$_4$ + 25% H$_2$		0.81	0.99	1.00
50% CH$_4$ + 50% H$_2$	ER = 0.67	0.94	1.00	1.00
25% CH$_4$ + 75% H$_2$		0.95	1.00	1.00
Hydrogen		1.00	1.00	1.00
Methane		0.94	1.00	1.00
75% CH$_4$ + 25% H$_2$		0.95	1.00	1.00
50% CH$_4$ + 50% H$_2$	ER = 0.93	0.95	1.00	1.00
25% CH$_4$ + 75% H$_2$		0.95	1.00	1.00
Hydrogen		1.00	1.00	1.00
Methane		0.31	0.68	0.91
75% CH$_4$ + 25% H$_2$		0.40	0.87	0.99
50% CH$_4$ + 50% H$_2$	ER = 1.2	0.95	1.00	1.00
25% CH$_4$ + 75% H$_2$		0.95	1.00	1.00
Hydrogen		1.00	1.00	1.00

The ignition probability for all gases increases with energy across all the lean (ER = 0.67) to rich (ER = 1.2) limits. Methane ignition probability varies a large amount from 17 % to 87 % (70 % variation) for energies from 2 mJ to 10 mJ respectively. This observation justifies that sources with energies around 2 mJ can result in only few ignitions of the released methane gas, but with higher energy sources around 10 mJ, ignition is virtually guaranteed as shown in Figure 4.31.

Figure 4.31 : Ignition probability variation for flammable gases at ER = 0.67

Hydrogen is excluded from Figure 4.31, Figure 4.32 and Figure 4.33 since its ignition probability (at energy levels 2, 5 & 10 mJ) is unity. (see Table 4.26.)

The wide range in ignition probability is narrowed down to a very small percentage change of 5 to 6% (94% to 100 % for methane and 95% to 100% for methane - hydrogen mixtures) when the gases approach the stoichiometric concentration. Ignition probability at close to a stoichiometric mixture (ER = 0.93) increases rapidly for all gases under consideration as compared to lean mixtures. (Figure 4.32)

Figure 4.32 : Ignition probability variation for flammable gases at ER = 0.93

Ignition probability for rich mixtures (ER = 1.2) again expands to a range of about 60% wide variation (31% to 91% for methane, 40% to 99% for 75% CH_4 + 25% H_2 mixture) for energies of 2 mJ to 10 mJ. The observations are presented in Figure 4.33. The variation in ignition probability is only 5% (not significant) for the other two gases (50% CH_4 + 50% H_2 mixture and 25% CH_4 + 75% H_2 mixture).

Figure 4.33 : Ignition probability variation for flammable gases at ER = 1.2

Addition of hydrogen increases the ignition probability values for all gases under consideration. The next section exclusively discusses the impact of increasing hydrogen concentration on various gas characteristics related to ignition energy and ignition probability.

4.6.4 Impact of hydrogen

Hydrogen minimum ignition energy is about 1/15th that of methane, and the lowering of the ignition energy for methane–hydrogen mixtures is quite evident if an increasing concentration of hydrogen is injected into methane. The experiments have enabled these variations in the ignition energy to be quantified. The first observation was the lowering of the lowest ignition energy as the hydrogen content was increased. A plot of lowest ignition energy against total flammables concentration is presented in Figure 4.34. The shift of the U shape curve towards the right for each of the gas mixtures under consideration, shows that the most easily ignitable mixtures have increasing concentrations of hydrogen (total flammables gas). The bottom of the U curve is brought down with increasing addition of hydrogen, suggesting the lowering of the MIE values for the flammable gases under consideration. The width of the U curve, i.e. the rage of concentrations over which ignitions were achieved, increases with increasing concentration of hydrogen.

Figure 4.34 : Lowest ignition energy at various flammable concentrations

The variation in ignition energy at 1%, 50% and 100% ignition probability was studied for increasing addition of hydrogen. The ignition energies for different methane – hydrogen mixtures were compared at ER of 0.67, 0.93 and 1.2. The result of the comparison is presented along with experimental observations of ignition energy in Table 4.27. Ignition probability increases for increasing ignition energy for all the gases under consideration. Increasing the concentration of hydrogen reduces the ignition energy, for all probability values under consideration.

Table 4.27 : Ignition energy at various ignition probabilities

Gases	Equivalence ratio	Ignition energy at various probabilities (mJ)			Observed Lowest ignition energy (mJ)
		IP = 1%	IP = 50%	IP = 100%*	
Methane	ER = 0.67	1.2243	4.2138	41.4881	1.8163
75% CH_4 + 25% H_2		0.3499	1.0253	9.4456	0.5840
50% CH_4 + 50% H_2		0.2691	0.6824	5.8359	0.1590
25% CH_4 + 75% H_2		0.1944	0.6179	5.8979	0.1049
Hydrogen		0.0303	0.1379	1.4799	0.0365
Methane	ER = 0.93	0.3933	0.7947	5.7998	0.2242
75% CH_4 + 25% H_2		0.1591	0.5852	5.8985	0.1049
50% CH_4 + 50% H_2		0.1696	0.5949	5.8983	0.0894
25% CH_4 + 75% H_2		0.1224	0.5536	5.9292	0.0663
Hydrogen		0.0303	0.1379	1.4799	0.0244
Methane	ER = 1.2	0.5934	3.2881	36.8865	0.8349
75% CH_4 + 25% H_2		1.0365	2.3668	18.9533	0.5160
50% CH_4 + 50% H_2		0.1877	0.6117	5.8979	0.1044
25% CH_4 + 75% H_2		0.2252	0.6463	5.8970	0.1115
Hydrogen		0.0303	0.1379	1.4799	0.0244

(* Ignition energy values are calculated for probability of 99.99%)

Ignition probability was quantified for the increased addition of hydrogen. The variation in the experimentally observed lowest ignition energy against hydrogen concentration (for different ER) is presented in Table 4.27 and Figure 4.35. The lowest ignition energy for the lean concentration (ER = 0.67) is reduced by 1.24 mJ (from 1.82 mJ to 0.58 mJ) when 25% of the methane is replaced by hydrogen. Further increases in the proportion of hydrogen does not bring further appreciable lowering of the ignition energy. At a rich concentration (ER = 1.2) ignition energy changes are quantitatively smaller in magnitude as compared to a lean concentration (ER = 0.67). The ignition energy change is smallest for ER=0.93 (near stoichiometric mixture) across the entire range of increasing concentration of hydrogen (ref. Table 4.27 and Figure 4.35).

Figure 4.35 : Lowest ignition energy varation with increasing hydrogen percentage

The variation in the ignition energy and the associated ignition probability for the flammable gases with addition of increasing concentrations of hydrogen, is likely to change the probability of fire and explosion accidents arising as a result of gas releases. Overviews of these changes along with the main findings of the study are discussed in the next chapter.

Conclusions of the Experimental study

In this thesis minimum ignition energy (MIE), lowest ignition energy at various concentrations and ignition probability are presented and discussed for methane-air, hydrogen-air and methane-hydrogen-air mixtures. The addition of hydrogen to the gas characteristics (ignition energy and ignition probability) is also discussed with an observed and predicted ignition probability for each methane–hydrogen mixture. Hydrogen, if injected into the natural gas infrastructure (natural gas considered as methane only) is likely to result in many changes to the gas industry and also to the end user. As a part of the safety work package of the Naturalhy project, the objective was to determine experimentally the ignition energy for methane–hydrogen–air mixtures, along with ignition energies for methane–air and hydrogen–air mixtures at ambient temperature and pressure. Ignition probabilities associated with the ignition energy values were also determined from the observed results.

MIE's for most of the flammable hydrocarbon gases are very small (0.3 mJ and less). Measurement of the MIE is done through various techniques; Capacitive spark discharge; Laser; and energy delivered by passing electric current through a wire placed in a flammable gas–air mixture (Martin Hattwing, 2004 and Lewis 1987). Komai et. al., 1994 has carried out experiments of igniting flammable gases (methane, hydrogen etc.) in air using frictional sparks from light alloys. The aim of the experiment was to examine the incendivity of the sparks and to determine the safety associated with handling flammable and explosive gases.

Capacitive spark discharge utilised during this work has been the preferred method for determining MIE for gases. The stored capacitor energy prior to the electrical discharge (spark) is considered as the energy delivered to the gas-air mixture. MIE is evaluated from the quantified spark discharge energy and success of ignition. The method is also standardised by British, European (BS EN 13821 - 2002) and American standards (ASTM E 582 -1988).

5.1 Main findings of the study

5.1.1 Minimum Ignition Energy

The experimental test runs has provided information on the MIE of the flammable gases under consideration. The observed MIE for methane (0.22 mJ) is marginally less than the historical data (0.29 mJ) reported by Lewis and von Elbe, 1987 and Bjerketvedt et. al., 1997 (Gas explosion handbook). Concentration of methane corresponding to the MIE is reported as 8.5% by Lewis and von Elbe, 1987 and 9% by Bjerketvedt et. al., 1997. This work suggest MIE (0.22 mJ) at a concentration of 9% methane in air. A methane concentration of 8% resulted in a lowest ignition energy of 0.24 mJ, which is less than the reported MIE values. This lowest ignition energy value at 8% concentration cannot be described as the MIE because of the definition of MIE. The value of MIE obtained for hydrogen of 0.024 mJ is in agreement with the German Hydrogen Association (D.W.V) value of 0.029 mJ, and is slightly greater than the Lewis and von Elbe value of 0.019 mJ.

MIE values for other gases (various methane-hydrogen mixtures) were also determined and found to be between those of methane and hydrogen MIE results. The most easily ignitable mixture (the concentration at which the MIE is determined) has an increasing concentration of total flammables, with the addition of hydrogen. Increasing concentration of hydrogen has the effect of lowering the MIE values of the gases. The correlation obtained from the test results as presented in Figure 4.23 is :

$$MIE = 0.2433 * \exp\left(-0.221 * C_{H2}\right) \qquad \qquad ...(5.1)$$

Where C_{H2} is the percentage of hydrogen in the methane-hydrogen mixture.

The variation in MIE for gases plotted against the reported values of laminar burning velocity for hydrogen, methane (Lewis and von Elbe, 1987) and predicted values (based on correlation proposed by Huanga, et. al. 2006) for methane-hydrogen mixtures are presented in Figure 4.26. The relationship obtained between the MIE and the laminar burning velocity (Su) is :

$$MIE = 0.0693 * Su^{-1.3028} \qquad \qquad ...(5.2)$$

Where Su   Laminar burning velocity (m/s)

Equations 5.1 and 5.2 provide means of predicting the MIE for either methane, hydrogen, or methane–hydrogen mixtures.

5.1.2 Ignition Energy

The lowest observed ignition energy for methane and hydrogen within the flammable range agree reasonably well with the reported data, except at the leanest mean concentration of methane. The lowest ignition energy for methane–hydrogen–air mixtures decreases with increasing concentration of hydrogen in the mixture. These values cannot be compared to the literature, because of the absence of any previous work on methane–hydrogen mixtures. The lowest ignition energies have a 'U' shape dependence on concentration. The characteristic U shape curve was observed for all the gases under consideration (see Figure 4.27). Ignition energy increases asymptotically at the lean and rich limits of flammability. The nature of the curve for all the gases indicated that, it may be possible to collapse the data into a single curve. Accordingly the Equivalence Ratio (ER) and the Ignition Energy (IE) was normalised as presented in Equations 4.18 to 4.20. The Normalised Equivalence Ratio (NER) and Normalised Ignition Energy were correlated as presented in Equations 5.3 and 5.4.

For lean gas mixtures $(ER < ER_{MIE})$

$$IE = MIE * \exp(3.2342 * NER * NER) \quad and \quad NER = \frac{ER - ER_{MIE}}{ER_{MIE} - ER_{LFL}} \qquad ...(5.3)$$

For rich gas mixtures $(ER > ER_{MIE})$

$$IE = MIE * \exp(10.119 * NER * NER) \quad and \quad NER = \frac{ER - ER_{MIE}}{ER_{UFL} - ER_{MIE}} \qquad ...(5.4)$$

Where MIE Minimum Ignition Energy for flammable gas (mJ)

        ER_{LFL} Equivalence Ratio at the Lower Flammability Limit

        ER_{UFL} Equivalence Ratio at the Upper Flammability Limit

        ER_{MIE} Equivalence Ratio of the gas at the MIE value

ER_{MIE} was correlated to the molecular weight (MW) of the gas as presented in Equation 5.5 (for methane, hydrogen and their mixtures) and Equation 5.6 (for methane, hydrogen, methane-hydrogen mixture, ethane and propane).

$$ER_{MIE} = 0.0014 * MW * MW - 0.0307 * MW + 1.081 \qquad ...(5.5)$$

$$ER_{MIE} = 0.0004 * MW * MW - 0.012 * MW + 1.0338 \qquad ...(5.6)$$

The Ignition energy predicted through Equations 5.3 and 5.4 was compared to the experimentally observed lowest ignition energy. The correlated and the observed values agree reasonably well with the reported data (for all the gases) as presented in Figure 4.28. Thus, the experimental data collated in this thesis, has enabled relationships to be developed that allows the lowest ignition energy of methane-air, hydrogen-air, methane–hydrogen–air mixture, ethane-air and propane-air mixtures to be calculated over their flammable ranges.

5.1.3 Ignition probability

The ignition probability for methane increases with increasing (spark) energy. The gradient of ignition probability against energy is greatest when the fuel-air ratio of the mixture is close to stoiciometric. The gradient is least when the mixture is lean or rich.

Ignition probability for hydrogen also increases with increasing energy. Concentrations of 20% to 30% (hydrogen percentage in air) have a very rapid increase in ignition probability with energy. The rate of increase (in ignition probability) with energy for 20%, 25% and 30% can not be differentiated from each other. Lean and rich mixtures have slower rates of increase in ignition probability as compared to a near stoichiometric mixture. Hydrogen ignition probability was observed to be always higher than methane, when compared at similar energy levels.

Ignition probability for all the gases increases exponentially with energy ultimately reaching a value of unity described by Equation 4.8.

$$IP = 1 - \exp\left[-b\left(E - E_{\min}\right)\right] \qquad \ldots (5.7)$$

The coefficients of the ignition probability equation 'b' and '$E_{\min}$' (Energy at zero ignition probability) determines the nature of the ignition probability curve. Coefficients b and $E_{\min}$ each were correlated to the lowest ignition energy as follows :

$$b = 0.3209 * IE^{(-0.7235)} \qquad \ldots (5.8)$$

$$E_{\min} = 0.95 * IE \qquad \ldots (5.9)$$

The Ignition probability (IP) equation after substitution of parameters 'b' and '$E_{\min}$' is

$$IP = 1 - \exp\left[-0.3209 * IE^{-0.7235} * \left(E - 0.95 * IE\right)\right] \qquad \ldots (5.10)$$

IP as observed for methane-air, hydrogen-air and methane-hydrogen-air mixtures was ultimately correlated to equivalence ratio (ER) and source energy (E). A generalised correlation to calculate 'IP' from the experimental result is :

<u>For lean mixtures</u> $\left(ER < ER_{MIE}\right)$

$$IE = MIE * \exp\left(3.2342 * NER * NER\right)$$

$$IP = 1 - \exp\left[\begin{array}{l} -0.3209 * \left\{MIE * \exp\left(3.2342 * NER * NER\right)\right\}^{-0.7235} \\ * \left(E - 0.95 * \left\{MIE * \exp\left(3.2342 * NER * NER\right)\right\}\right) \end{array}\right] \qquad \ldots (5.11)$$

Where $NER = \dfrac{ER - ER_{MIE}}{ER_{MIE} - ER_{LFL}}$

ER_{LFL} Equivalence Ratio at the Lower Flammability Limit

$$ER_{MIE} = 0.0014 * MW * MW - 0.0307 * MW + 1.081$$

$$MIE = 0.0693 * Su^{-1.3028}$$

For Rich mixtures $(ER > ER_{MIE})$

$$IE = MIE * \exp\left(10.119 * NER * NER\right)$$

$$IP = 1 - \exp\left[\begin{array}{l} -0.3209 * \left\{MIE * \exp\left(10.119 * NER * NER\right)\right\}^{-0.7235} \\ *\left(E - 0.95 * \left\{MIE * \exp\left(10.119 * NER * NER\right)\right\}\right) \end{array}\right] \qquad ...(5.12)$$

Where $NER = \dfrac{ER - ER_{MIE}}{ER_{UFL} - ER_{MIE}}$

ER_{UFL} Equivalence Ratio at the Upper Flammability Limit

MIE and ER_{MIE} has the same meaning as referred for lean mixtures

The IP relationships help in predicting the ignition probability for methane-air, hydrogen-air and methane-hydrogen-air mixture from a knowledge of equivalence ratio (ER) and source energy (E). It may be possible to extend the relationships to include other flammable gas such as ethane-air and propane-air mixtures by substituting ER_{MIE} value (Equation 5.6) in the Equations 5.11 and 5.12. However, experimental data on the ignition probability of these gases is not available to confirm whether or not this approach is reasonable.

Increasing concentration of hydrogen in methane-hydrogen mixtures decreases MIE and Ignition energy for the gas mixture. An increase in ignition probability is observed due to increasing the hydrogen content of the gas mixture. It can be expected that such changes will result in an increase in incidents of fire and/or explosion. Chapter 6, discusses this issue, using deterministic values of leakages (for methane and hydrogen) and ignition source energies. A quantified change in the gas incident probability from existing knowledge of gas release frequency is presented in Chapter 6.

Changes in gas incident probability

Introduction of hydrogen into the natural gas infrastructure will change the gas properties, which were discussed in earlier chapters. The experimental results indicate that there is greater risk of ignition of hydrogen-air mixtures, compared to methane–air mixtures. The ignition risks are compared for methane and hydrogen based on the experimental observations of minimum ignition energy (MIE), lowest ignition energy at various concentrations and ignition probability.

6.1 Comparing methane and hydrogen releases

Comparison of the probability of ignition of gas releases was done using methane and hydrogen. Properties of other gases (various mixtures of hydrogen and methane) were considered to be somewhere between these two extremes. Comparison of the results for hydrogen and methane show that, hydrogen has an increased flammability range, a lower ignition energy and a higher ignition probability. Further comparison based on the energy density shows that hydrogen energy (120 MJ/kg) is more than twice that of methane (50 MJ/kg). Thus, hydrogen has a somewhat higher flammability hazard and likely to deliver more destructive energy for equal quantum (1 kg) of flammable gas oxidised/consumed. Swain et. al., 2005 expressed the view that hydrogen disperses so quickly that it is unlikely to create a flammable mixture of any large volume. Swain's experiments were carried out in open space. In confined spaces, such as homes, garages, etc., hydrogen will not disperse so readily and the buoyancy of hydrogen will have less effect, if it releases inside domestic properties.

This is the major concern of this work and the effect of adding hydrogen to natural gas on the frequency of fires and explosions. Hence, this analysis is based on confined spaces of domestic gas releases and its fire and explosion potential.

Gas ingress within the domestic property is considered as the starting point of this analysis. The gas release can have an origin within or outside the house. The analysis is based on experimental studies carried out for natural gas releases

(HSE, 1998). Hydrogen gas releases are considered to occur under similar conditions, but with a density correction factor to calculate the hydrogen flow rate from the corresponding methane release rate.

Three methane gas releases; minor, significant and major 0.36 m^3/hr, 1.44 m^3/hr and 7 m^3/hr, are considered in the analysis, in line with the HSE CRR report 168, 1998. The minor release corresponds to the gas flow rate from a single gas burner on a domestic gas cooker. The significant release condition is, when all four burners on a cooker are ON. The major release condition is corresponding to the maximum likely leak from a single open end carcass/internal pipe, within the house downstream of the meter. Typical values of the background ventilation rate of 0.6 air changes per hour and a room volume of 25 m^3 is considered as per the HSE report. Conditions of perfect mixing are assumed within the room to calculate the steady state gas concentration. The rate of build up of the gas concentration to the steady state level as a function of time is presented in Figure 6.1. The actual values of the maximum steady state concentration calculated under these typical conditions of room volume and ventilation rates are presented in Table 6.1. The ignition probability for the energy level of 2 and 5 mJ (energy delivered by various ignition sources present within the house) are also presented in Table 6.1. Ignition sources have energies far greater than 2 – 5 mJ, but it's the energy delivered to the gas that is important for successful ignition, hence the lower values considered in the analysis. Experiments in which gas concentrations build up within a particular room, also lead to gas-air mixture accumulation in an adjacent room, even if an interconnecting doorway is closed (one of the major findings of the HSE, 1998 report). This practical observation restricts us from reducing the ignition probability value, when gas concentration surpasses the stoichiometric concentration. Accordingly under practical situations the ignition probability can not be considered as negligible (zero) at gas concentration exceeding UFL values. A conservative approach of risk assessment is adopted here. If the room concentration exceeds the stoichiometric concentration, ignition probability is considered as the maximum value observed at the stoichiometric concentration, for the gases under consideration (either methane or hydrogen in this present case).

Table 6.1 : Ignition probability for methane releases within confined areas

| Methane release | m³/hr | Conc.(%) | Percentage ignition probability at energy | |
			2 mJ	5 mJ
Minor	0.36	2.34	0	0
Significant	1.44	8.8	49	100
Major	7.0	31.82	49	100

Hydrogen is 1/8th times less dense than methane, accordingly the volume leakage rate for hydrogen is expected to be 2.8 times that of methane. If, when hydrogen is used to replace some of the natural gas, the pressure is increased in order to supply an equivalent amount of energy, then this factor would be even greater. This, coupled with lowering the ignition energy for hydrogen results in increased ignition probability for all categories of releases; minor, significant and major cases under consideration. The minor release of hydrogen is likely to form a flammable cloud (with concentration of 6.4% hydrogen in air, inside the room), which was not the case with a minor release of methane (concentration of 2.3% methane in air was achieved under identical conditions, inside the room). The results of the deterministic analysis are presented in Table 6.2.

Figure 6.1 : Gas concentration built up for methane, hydrogen release in room

Table 6.2 : Ignition probability for hydrogen releases within confined areas

| Hydrogen release | m³/hr | Conc.(%) | Percentage ignition probability at energy | |
			2 mJ	5 mJ
Minor	1.02	6.4	3	8
Significant	4.07	21.4	100	100
Major	19.80	56.9	100	100

Comparison of Table 6.1 and Table 6.2 for Methane and Hydrogen, (using the deterministic releases of flammable gases suggest some understanding of the increase in the ignition probability. For minor releases, the rise in ignition probability was estimated to be 3%, for an ignition source delivering 2 mJ energy to flammable gas–air mixture. If the source energy is increased to 5 mJ, then the probability increases to 8%. Significant and major releases result in an increase in ignition probability from around 50 % to 100 %, (i.e. almost twice the number of cases with hydrogen will result in fire and/or explosion as compared to the base case) for a 2 mJ energy. With higher energies (> 2 mJ) the ignition is always certain (ignition probability is unity) for significant and major releases, hence further increase in ignition probability because of fuel change is ruled out. This analysis predicts quantitative increase in the ignition probability when domestic fuel (flammable gas) is changed from methane to hydrogen.

In real world situations successful ignition of released gas is dependent on many factors. Thus ignition probability is dependent on numerous parameters. In the present analysis, ignition probability is considered dependent on probability of flammable vapour cloud formation and then ignition of this vapour cloud using surrounding ignition sources. The probability of formation of a flammable cloud is calculated from the leakage and ventilation rates, which were deterministic in the above analysis. Probabilistic consideration of leakage and ventilation rates within the residential area will help to arrive at the probability of flammable cloud formation. Experiments with common ignition sources within a flammable gas air mixture, will determine the ratio of source energy and the delivered energy. These studies will reduce the uncertainties associated with the determination of ignition probability. The same methodology can then be applied to estimate the quantitative increase in the ignition probability with the change of domestic fuel from methane to hydrogen.

Issues for future work

The experimental facility used to carry out the ignition energy measurements for methane–air, hydrogen–air and methane–hydrogen–air gas mixtures was designed and built at the Department of Chemical Engineering, Loughborough University. The rig is equipped to handle other flammable gases e.g. propane, butane simply by replacing the feed gas. Accordingly, ignition energy, ignition probability and minimum ignition energy for other flammable gases and mixture of gases can be determined using the existing experimental facility. A few of the limitations of the existing experimental arrangement are discussed in details in Section 7.1. Issues for future work are discussed in Section 7.2.

7.1 Limitations of the experimental setup

The operating procedure for the experiments was semi-automatic where the preparation of the gas mixture (valve operation) was done manually and the ignition process, with subsequent data recording, completed automatically.

The mixtures for the exeriments involving mixtures of methane and hydrogen were prepared inside the vessel by the method of partial pressures, by first injecting one of the gases (either methane or hydrogen) and then adding the second gas as required for the targeted concentration. The recorded pressure of the first and second gases, with a consistent pressure difference (between the two gases), gives the flammable gas mixture concentration to be prepared. The lowest ignitable (practically achievable), gas mixture of 25 % hydrogen in methane and 75 % hydrogen in methane were prepared with the lean limit of 8% flammables in air. The lean limit for these gases can be far below this 8% concentration. The existing method of preparing the gas mixture from individual gases, prevented, experiments to be performed below 8% concentration (for $75\%CH_4 + 25\%H_2$ and $25\%CH_4 + 75\%H_2$). This technique also meant that the ratio of methane to hydrogen was likely to be slightly different for each test. Therefore, it is suggested that, rather than mixing the flammable gases directly in the combustion chamber, premixed gases should be used.

The experimental setup was designed and fabricated in order to produce sparks of very low energies to investigate the ignition energy and the ignition probability for flammable gas air mixture at atmospheric pressure. The energy delivered by the capacitive spark is considered as the ignition energy during the test runs. The High Voltage (HV) transformer inside the Extra High Tension (EHT) pulse generator determines the maximum voltage obtained during the spark process. The electrical circuit for the spark process operates by triggering a thyristor. A high-voltage pulse with amplitude of maximum 15 kV (in the present case) is generated by discharging a primary capacitor of 1 µF, initially charged at about 300 V, through a HV transformer. The energy stored on the primary capacitor, which indicates a theoretical upper energy limit of the spark energy. Losses in the HV transformer and the charging resistor cause the actual upper energy limit to be significantly lower. The maximum consistent delivered spark energy achieved during the ignition process was 10 mJ only. It was observed that the voltage delivered by the HV transformer was insufficient to charge capacitors with higher capacitance (390 pF). This larger value capacitor was unable to deliver a spark after triggering the thyristor. The charging time for the large capacitor was greater compared to the other capacitors. This was also one of the reasons that no spark was observed inside the vessel when working with the larger capacitors.

The experimental flammability limits for the gases are heavily dependent on the ignition energy of the sparking process. Ignition energies have a characteristic 'U' shape dependence on concentration and this was observed for all the gases under consideration. Ignition energy increases away from the stoichiometric mixture at an ever increasing rate and increases, asymptotically, at the limiting values of flammability. The experimentally observed flammability limits were narrower than the reported data. Increasing the spark energy (by due modification of the electrical circuit) will help broaden the range of experimental gas compositions that can be studied as the lean and rich limits are approached. Extending the range of experiments, particular in the vicinity of the lean flammability limit, is important since little information is available near to the flammability limits.

7.1.1 Modifications proposed to the experimental setup

Human intervention was required during, vacuum creation, gases introduction (valves operation) and combustion products expulsion. The manual operation of in-situ

preparation of methane-hydrogen mixtures, severely limits the lowest (lean) limit of the gas to be prepared and experimentally tested inside the vessel. Procurement of premixed gas cylinders with 25% H_2 in CH_4, 50% H_2 in CH_4, and 75% H_2 in CH_4, (similar compositions to the one studied in this work) for further experimental runs, will help reduce the errors in manual preparation of these gases. Moreover the ratio of the methane and hydrogen gases will be consistent over the entire flammability range, when gases are drawn from same premixed cylinder.

The pure gases (methane and hydrogen) or the premixed gases would still require manual operation of feed valves, since various flammable gases will be introduced to the evacuated vessel, followed by air injection to achieve various flammable concentrations. Manual preparation often required expulsion of gases, when more than the desired quantity of flammable gas was introduced to the combustion chamber. This resulted in loss of flammable gas. Hence high precision metering pumps can be installed on each of the feed gas lines (including air), to get the exact predetermined composition of the flammable concentration inside the combustion chamber.

The stored energy in the primary capacitor was the theoretical upper limit of the spark energy delivered by the rig. If the primary capacitor is not large enough, the discharge capacitor never reaches the breakdown voltage during the high-voltage pulse charge duration. The maximum achievable upper energy limit from the circuit was only 10 mJ. A higher value primary capacitor is proposed to obtain higher energy from the circuit. The primary capacitor cannot be increased disproportionately. The circuit can accommodate a certain increase in capacitance, which needs to be determined only after installation of the increased value capacitance. To increase the spark energy further, a higher turns ratio HV transformer is required to achieve higher voltage on the secondary side of the transformer. The higher voltage will be fed through a charging resistor, causing a voltage build-up on the discharge capacitor placed downstream of the charging resistor. A spark occurs when the discharge capacitor finally gets charged to the breakdown voltage of the electrode gap. This helps in generating a spark with higher value capacitors, delivering higher energy. Accordingly modifications in the EHT pulse generator unit is proposed to achieve higher spark energy.

The ignition process is visually observed and the transient effects of the ignition process confirmed through the overpressure wave recordings as generated during the process. The observation windows provided on the experimental setup can be utilised to measure various other parameters, if a high speed camera (camera exceeding the flame speed) is installed outside the windows. The camera can be interlocked with the spark triggering process to capture photographs of the burning process. Images of the development of the flame kernel, and the subsequent flame, including video records can be obtained. Various parameters, like flame speed, burning velocity, overpressure wave, temperature gradients etc. can be correlated with each other for the flammable gases under consideration (Ilbas et. al. 2006 and Fairweather et. al. 2009). Initial disturbances created by the electrical discharge, flame initiation, flame size, flame development, flame growth can also be measured. The birth of the ignition process can be studied through the flame photographs. More information of the ignition process can be obtained if a high speed camera is synchronised with the experimental setup. Accordingly a detailed insight into the burning process and the ignition process can be obtained.

7.2 Future work

Modifications suggested to the experimental rig, will allow additional parameters to be monitored (discussed in Section 7.1.1). This will enable a better understanding of the ignition process to be obtained. The combustion chamber and its associated ancillary units improve the certainty with which various gas compositions can be prepared and increase the accuracy of the measurement of ignition energy. The cylindrical combustion chamber is capable of withstanding the overpressure waves generated during the combustion process for all flammable gas-air mixtures prepared at ambient conditions. Such a vessel can be used to study other combustion process parameters. The parameters which can be measured are numerous, on the subject matter of ignition, flame development, flame propagation, burning process and combustion.

Ignition probability for gas release cases are often considered from historical data or from expert judgement to arrive at an understanding of the risk of fire and/or explosion. Results from the present experiments can be considered a beginning phase to determining the proportion of gas release incidents that can proceed to fire

and/or explosion. In order to further analyse real world situations to arrive at ignition probability for gas releases, all ignition sources need to be categorised with their ignition sources energies. As already discussed, the energy delivered during the sparking process and the energy actually received by the gas is different. Moreover the enclosure of the ignition sources prevents the flammable gas-air mixture actually penetrating to the spark location. The degree of confinement or openness of the sources can be studied for common ignition sources observed in our daily life. Ignition sources can be placed inside the cylindrical combustion chamber and then tested with various flammable gas-air mixture compositions for their ignition potential. The certainty with which these ignition sources can result in the ignition of the flammable gas can be measured experimentally. Accordingly, various common sources can be tested and assigned ignition potential. Experiments with different gas composition for methane and also with hydrogen will help determine the impact of each gas and its nature on the ignition properties.

Distribution of various ignition sources within the house/confined area and the ignition potential will help predict the ignition probability of the released gas. The uncertainty in the ignition probability can thus be reduced (if not eliminated) to arrive at the actual value of the ignition probability under realistic gas release scenarios in future.

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Appendix A

Research work presentation and meetings attended

S.No.	Name of event	Venue	Date
I	**Conferences / Technical meetings**		
1	NATURALHY Project - Safety Work Package meeting	Loughborough University, UK	26 March 2008
2	NATURALHY Project meeting – Technical discussions on ignition energy	Shell Global Solutions, Cheshire, UK	16 October, 2007
3	The 2nd International Conference on Hydrogen Safety (HYSAFE)	Miramar Palace, San Sebastian, SPAIN	11- 13 September, 2007
4	NATURALHY project - fourth meeting of the full consortium of the project	Holiday Inn, Stratford on Avon, UK	28 –29 June, 2007
5	NATURALHY Project - Safety Work Package meeting	National Grid Office, Warwick, UK	27 June, 2007
6	Defence-in-depth: Prevention of Escalation of Fires and Explosions, FABIG technical meeting.	Institution of Structural Engineers, London	28 February, 2007
7	Indian Chemical Engineering Congress Conference (CHEMCON) 2006	GNFC Ltd., Gujarat, INDIA	27 – 30 December, 2006
8	Safety Implications of a Hydrogen Economy: Risk Assessment and Design against Fires and Explosions, FABIG technical meetings	Institution of Structural Engineers, London	15 February, 2006
II	**Large scale experiments**		
9	Large scale experiments for build up of gas in confined /vented enclosures and gas explosions at test site	Spadeadam, Cumbria, UK	05 November 2005
III	**Training**		
10	Introduction to PIPESAFE and ORDER - risk assessment packages	Advantica, Loughborough	10 - 11 November 2005
11	Hazard Identification, Consequence analysis and Risk assessment (Module - CGP 073)	Loughborough University, UK	18 – 22, July 2005

Properties of Hydrogen and Methane gas

Gas property	Methane	Hydrogen
Gas Density at NTP (kg/m^3)	0.72 kg/m^3	0.09 kg/m^3
Energy density (Higher heating value, Lower heating value as MJ per kg)	50 MJ / kg (55.53, 50.02)	120 MJ / kg (141.86, 119.93)
Flammability limits (% vol)	5 % - 15 %	4 % - 75 %
Minimum Ignition Energy (mJ)	0.29	0.02 mJ
Diffusivity (cm^2/sec)	0.16	0.61
Autoignition temperature (K)	813	853
Water solubility in 100 mL	3.5 mg @ 17° C	0.16 mg @ 20° C
Melting point (K)	91	14
Boiling point (K)	109	20
Flash point (K)	85	20
Flame temperature (K)	2148	2318
Max. Laminar burning velocity in air (m/s)	0.43	3.46
Detonation limits (% Vol.)	6.3-13.5	11/18-59
Flame quenching distance (gap)	2.16 mm	0.6 mm
Flames visibility	visible	invisible
Molecular weight	16	2

The above data is gleaned from various sources (books, standards, research papers and internet search) and hence references are not provided.

Appendix C

Calculations for the cylindrical combustion chamber design

The cylindrical combustion chamber (Figure C.1) was fabricated from a 6 inch (schedule 40) stainless steel (SS) seamless pipeline section with welded flanges on either side of the pipe ends. The ends were closed using stainless steel flanges specially fabricated to house the polycarbonate plate and then bolted to the cylinder ends through covering flanges.

The actual cylindrical combustion chamber measurements are:

Internal diameter (ID) = 155 mm
Outer diameter (OD) = 169 mm
Wall thickness   = 7 mm
Chamber Length   = 232 mm
Flange (welded to pipe) outside diameter   = 279 mm
Flanges (welded to pipe) thickness   = 24 mm
Flanges (for closures) ID   = 155 mm
Flanges (for closures) OD   = 279 mm
Flanges (for closures) thickness   = 24 mm
Polycarbonate housing diameter   = 197 mm
O ring gasket nominal bore   = 175 mm
O ring gasket diameter   = 5 mm

Total thickness of polycarbonate plate (for window) = 25 mm
Polycarbonate plate inside the shell   = 10 mm
Polycarbonate plate inside flanges   = 15 mm
Bolt pitch circle diameter   = 242 mm
Diameter of hole for bolts   = 10 mm
Diameter of each bolts   = 9.5 mm
Nos. of bolts   = 16

(all dimensions are in mm)

Figure C.1 : Details of the windows and the flange assembly for the cylindrical combustion chamber

C.1   Sample calculation for Overpressure expected during the explosion

The maximum overpressure for confined gas explosion within experimental cylindrical combustion chamber was based on the assumption, that during the combustion of a stoichiometric mixture of fuel gas with air, the adiabatic flame temperature is reached for the burning gases. In practice the actual flame temperature achieved will be less than the adiabatic flame temperature, but the cylindrical combustion chamber was designed for the worst case scenario (EN 13673 : 2003). Accordingly two different cases were considered to determine in maximum adiabatic flame temperature during each process.

Case I : Stoichiometric mixtures of methane – air
Case II : Stoichiometric mixtures of hydrogen– air

Technical data used

Fuel	M. wt.	% fuel gas at stoic. ratio	Adiabatic flame temp T_f (K)	Ambient (initial) temp. T_i (K)	Ambient (initial) pressure $P_{ambient}$ (MPa)
Methane	16	9.5	2148	288	0.1
Hydrogen	2	29.5	2318	288	0.1

During the combustion process define

   N_u = number of moles or all reactants
   N_b = number of moles or all products formed

Case I : Combustion reaction for methane

$CH_4 + 2 O_2 + 7.52 N_2$ ----> $CO_2 + 2 H_2O + 7.52 N_2$

NR = 1+ 2 + 7.52 = 10.52   NP = 1+ 2 + 7.52 = 10.52

NP / NR = 10.52 / 10.52 = 1

Expansion factor 'EF' =   (Tf / Ti) * (NP/ NR)

                              =   (2148 / 288) * (1)

                              =   7.45

Maximum Pressure generated during explosion

Pmax = $P_{ambient}$ * EF

Pmax = 0.1 * 7.45 MPa.
Case II : Combustion reaction for hydrogen

$H_2 + 0.5\ O_2 + 1.88\ N_2 \quad ----> \quad H_2O + 1.88\ N_2$

NR = 1+0.5 + 1.88 = 3.38   NP = 1+1.88 = 2.88

NP / NR = 2.88 / 3.38 = 0.85

Expansion factor 'EF' =   (Tf / Ti) * (NP/ NR)

                      =   (2318 / 288) * (0.85)

                      =   6.84

Maximum Pressure generated during explosion

Pmax = $P_{ambient}$ * EF

Pmax =   0.1 * 6.84 MPa.

Flammable mixture	Max. Pressure (MPa)
Case I : Stoichiometric mixtures of methane – air	0.745
Case II : Stoichiometric mixtures of hydrogen– air	0.684

For cylindrical combustion chamber design purposes the maximum peak overpressure generated during the combustion process in Case I or Case II was assumed to be $p_{(combustion)}$ = **0.8 MPa.**

C.2   Sample calculation for the shell thickness

Cylindrical combustion chamber parameters

Shell thickness   (th_s) = 7   mm   (0.007 m)
Shell diameter   (d)   = 155 mm   (0.155 m)

Allowable working stress for SS, considered (f)   = 150 MPa (1500 kg/cm^2)
Overpressure expected $p_{(combustion)}$ =   0.8 MPa

Based on the hoop stress (p_h) equation, the cylindrical combustion chamber can withstand a maximum pressure of

        p_h =   2 * f * th_s / d

                  =   2 * 150 * 0.007 / 0.155

                  =   13.5 MPa

Safety factor for the vessel shell   = p_h / $p_{(combustion)}$

            =   17

The safety factor of 17 was based on the fact that the pressure inside the cylindrical combustion chamber was considered static, whereas the pressure exerted during the explosion and subsequent overpressure wave impact was dynamic. Even if the dynamic pressure (overpressure) was twice the static pressure, the cylindrical combustion chamber shell thickness would be strong enough to carry out the experiments (explosions inside the cylindrical combustion chamber)

C.3   Sample calculation for polycarbonate plate window thickness

Window parameters

Window diameter   (d_w)   = 155 mm (0.155 m)
Window thickness   (th_w)   = 15   mm (0.015 m)
Polycarbonate Tensile strength   (f)   = 50 MPa
Overpressure   (p)   = 0.8 MPa

$th_w = d_w * \sqrt{(3/16) * (p/f)}$ (ref. Brownell L.E. and Young E.H.
               Process equipment design, 1977)
Or

Limiting pressure ' p ' for a window thickness ' th_w ' will be

$$p = (16/3) * f * (th_w / d_w)^2$$

$$= (16/3) * 50 * (15/155)^2$$

$$= 2.50 \text{ MPa}$$

Safety factor for polycarbonate window   = p / p_h

               = 3.13

The safety factor of 3.13 for the windows was based on static pressure inside the cylindrical combustion chamber, similar to the analysis carried out for the shell thickness. The same logic of static and dynamic pressure was applied here, for the calculation of window thickness. It was inferred that in the case of dynamic pressure also, the windows would be capable of withstanding the overpressure wave generated through the explosion.

C.4 Sample calculation during vacuum formation

Cylindrical combustion chamber parameters
Vessel Length (l) = 232 mm
Outside diameter (d_o) = 169 mm .
Vessel wall thickness (th_s) = 7 mm

Vessel Length / Outside diameter (l / d_o)= 232 / 169 = 1.37

Outside diameter / thickness (d_o / th_s) = 169 / 7 = 24

Factor B = p (do/t) = 1400 (Ref. Brownell L.E. and Young E.H. Process equipment design, 1977, pp. 147,)

where

p (allowable pressure)= 1400 / (24)

= 58 psi

Since the atmosphere can exert a pressure of only 15 psi. (outside pressure = 14.7 psi) in case of a full vacuum (pressure inside vessel = zero psi), where as the cylindrical combustion chamber can withstand a negative pressure of 58 psi.

Comment : The cylindrical combustion chamber is able to withstand stresses generated due to explosive pressure and during the complete vacuum formation inside the cylindrical combustion chamber.

C.5   Flange specification

As per ASTM A 181 (BS 4504 : 1989) flange specifications for the 6 inch Stainless steel pipe are as follows:

Outside diameter   = 13.5 in.   (350   mm)
Min. thickness   = 1.125 in.   (28.6   mm)
Bolt pitch circle diameter = 11.75 in.   (298.45 mm)
Diameter of hole for bolts = 0.875 in.   (22.2   mm)
Diameter of bolts   = 0.75 in.   (19   mm)
Nos. of bolts   = 8

Actual flange specifications used

Outside diameter   = 279 mm
Flange thickness   = 24 mm
Bolt pitch circle diameter = 242 mm
Diameter of hole for bolts = 10 mm
Diameter of bolts   = 9.5 mm
Nos. of bolts   = 16

The standard (BS / ASTM) flange specifications are based on industrial use of the 6 inch pipe, utilised for very high pressures. The maximum pressure achieved in our experimental cylindrical combustion chamber is disproportionately less than the industrial pressures. Hence present flange specifications utilised in fabrication of the experimental cylindrical combustion chamber are adequate as confirmed through the calculations presented above.

Risk Assessment Record for the experimental rig

Assessment Number

Department Chemical Engineering	Location 2 nd. Pilot Laboratory Room No./Area : S- 038 Ignition energy measurement rig	Assessor Hemant Mathurkar	Date 08.05.2007

Risk Identified *(see Hazard prompt list over page)*	Persons at Risk *(Groups or nos.)*	Controls in Place	Assessors Risk Rating *(H.M.L. see over page)*	Assessors Actions to Further Reduce Risk
Methane : Methane is a colourless and odourless gas. It is flammable and may form flammable and explosive mixtures with air. Asphyxia may result if the oxygen concentration is reduced to below 18% by displacement.	All persons working in lab A Area (S - 038)	Fumes / flammable gases detection and extraction system	Medium	Evacuate area ((S - 038), use all appropriate protective equipments, Eliminate ignition sources. The rig is at ambient pressure and its volume is 4 litres, so only minor leaks from vessel are envisaged in case of valves malfunction
Hydrogen: Hydrogen gas is colourless, odourless and flammable. It forms flammable and explosive mixtures with air over a wide range of concentrations. Hydrogen burns with an almost invisible blue flame Gas is non-toxic, and the primary health hazard is asphyxiation by displacement of oxygen.	All persons working in lab Area (S - 038)	Fumes / flammable gases detection and extraction system.	Medium	Evacuate area ((S - 038), use all appropriate protective equipments, Eliminate ignition sources. The rig is at ambient pressure and its volume is 4 litres, so only minor leaks from vessel are envisaged in case of valves malfunction
Assessors Comment : Average risk rating is medium		Supervisors Comment		
Assessors Signature		Supervisors Signature		
Reassessment Date			Date: 10.05.2007	

Loughborough University

Risk Assessment Record

	Assessment Number

Department Chemical Engineering	Location 2 nd. Pilot Laboratory Room No./Area : S- 038 Ignition energy measurement rig	Assessor Hemant Mathurkar	Date 08.05.2007

Risk Identified *(see Hazard prompt list over page)*	Persons at Risk *(Groups or nos.)*	Controls in Place	Assessors Risk Rating *(H.M.L. see over page)*	Assessors Actions to Further Reduce Risk
Overpressure of experimental rig, before or following Ignition.	All persons working in lab Area (S - 038)	The line pressure and vessel pressure is continuously monitored and controlled. Excessive pressure can be vented through three way valve. Rig is over-designed with safety factor = 17. Windows designed with safety factor = 3.	Medium	Evacuate area, use all appropriate protective equipments. Adopting the safe procedure of disconnecting the hose from the ignition vessel and then venting the excess pressure to atmosphere.
Gas leak in line: Methane / Hydrogen : Gases are colourless, odourless and flammable. It forms flammable and explosive mixtures with air over a wide range of concentrations. Gases are non-toxic, and health hazard are asphyxiation by displacement of oxygen.	All persons working in lab area	Fumes / flammable gases detection and extraction system. Check for leaks using leak detection fluid	Medium	Close all cylinder valves and regulators. Minor leaks will be diluted to below the LFL limits. For major leaks Contact emergency telephone number (888).
Assessors Comment : Average risk rating is medium		Supervisors Comment		
Assessors Signature		Supervisors Signature		**Loughborough University**
Reassessment Date			Date: 10.05.2007	

Hazard Prompt Lists

Machinery & Work Equipment – Category 1
- Mechanical Hazards
- Electric Shock
- Vehicles/Transport
- Hand Tools

Hazards associated with Place of Work – Category 2
- Slips, trips and falls on a level
- Falls from a height
- Falling objects/materials
- Striking objects
- Contact with hot/cold materials/surfaces
- Storage and stacking
- Space and confined work area

Hazards associated with materials, substances & physical agents – Category 3
- Dust, fume and gases
- Biological hazards/infection
- Noise/Vibration
- Compressed gases

- Hazardous chemicals (CHIP P.10)
  - Explosive
  - Oxidising
  - Extremely flammable
  - High flammable
  - Flammable
  - Very toxic
  - Toxic
  - Harmful
  - Corrosive
  - Irritant
  - Sensitising
  - Carcinogenic
- Entry into confined space/lack of oxygen

Hazards associated with activity, methods of work – Category 4
- Manual handling
- Upper Limb Disorders/Repetitive Strain Injury
- Visual fatigue
- Posture

Hazards associated with Work Organisation (Contractors/Service) – Category 5

Hazards associated with Work Environment – Category 6
- Temperature
- Heating
- Ventilation
- Lighting

Other types of hazard – Category 7
- Violence
- Stress
- Drugs
- Substance abuse

Risk Ratings (High, Medium, Low)

CONSEQUENCE:

3   **Major** (e.g. death or major injury as per RIDDOR or irreversible health damage)

2   **Serious** (e.g. injuries causing absence of more
than three days or significant health effects – reversible)

1   **Slight** (other injuries requiring first aid and minor ill health effects)

LIKELIHOOD:

3   **High** (where certain or near certain harm will occur

2   **Medium** (where harm will frequently occur)

1   **Low** (where harm will seldom occur)

RISK RATING = CONSEQUENCE x LIKELIHOOD

High 6 – 9
Medium 2 – 4
Low 1

Department Chemical Engineering	Equipment Description:- Ignition energy measurement rig Area:- **Room No. S- 038 (2ⁿᵈ Pilot Laboratory)**		Assessor Hemant Mathurkar	Date 08.05.2007	
OPERATION/ ACTIVITY	**Risk Identified** (see Hazard prompt list over page)	**Persons at Risk** (Groups of numbers)	**Controls in Place**	**Risk Rating** (H, M, L see over page)	**Action-Control Required to further reduce risk**
Direct contact to any of the high voltage device	Electrical hazard (direct contact)	Single person	Ignition can only be carried out when key is inserted	MEDIUM	Operations strictly as per the operating procedure. Avoid wet conditions on the floor and near the devices.
Changing the capacitor / resistors in the ECU	Electrical hazard (direct contact)	Single person	Interlock control system	MEDIUM	Earth capacitors before changing / removing and resistor / capacitor
Mechanical impact by a conductor material / dropped object on ECU	Electrical hazard (only if it is simultaneous with the triggering pulse, OR manual triggering	Single person	ECU is closed	LOW	No shelf above the ECU. Nothing is kept on the ECU.
Assessors Comment: No further risks identified			Managers Comments/Approval:		
Reassessment Comments:		Date 09.05.2007			
Assessors Signature		Date 10.05.2007	Managers Signature		Date 10.05.2007

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Machinery Hazard Prompt List

MECHANICAL HAZARDS

Crushing
Shearing
Cuttings/Severing
Entanglement
Drawing-in/Trapping
Impact
Stabbing/Puncture
Friction/Abrasion
High Pressure Fluid Injection
Slips/Trips/Falls
Falling/Moving object
Other mechanical hazards

ELECTRICAL HAZARDS

Direct contact
Indirect contact
Electrostatic phenomena
Short circuit/Overload
Source of ignition
Other electrical hazards

RADIATION HAZARDS

Lasers
Electro-magnetic effects
Ionising/Non-ion. radiation

Other radiation hazards

HAZARDOUS SUBSTANCES

Toxic fluids
Toxic/gas/mist/fumes/dust
Flammable liquids
Flamm.gas/mist/fumes/dust
Explosive substances
Biological substances
Other hazardous substances

WORK ACTIVITY HAZARDS

Highly repetitive actions
Stressful posture
Lifting/Handling
Mental overload/Stress
Visual fatigue
Poor workplace design
Other workplace hazards

WORK ENVIRONMENT

Localised hot surfaces
Localised cold surfaces
Significant noise
Significant vibration
Poor lighting
Hot/cold ambient temperature
Other work environment

Hazards

Risk Assessment Method & Estimation Ratings

| RISK related to the considered hazard | is a function of | severity of the possible harm for the considered hazard | and | PROBABILITY OF OCCURRENCE of that harm |

PROBABILITY OF OCCURRENCE of that harm

frequency and duration of exposure

probability of occurrence of hazardous event

possibility to avoid or limit the harm

RISK RATINGS (HIGH, MEDIUM, LOW)

SEVERITY -
3 - **MAJOR (eg. death or major injury as per RIDDOR or irreversible health damage)**
2 - **SERIOUS (eg. injuries causing absence of more than three days or significant health effects - reversible).**
1 - **MINOR (eg. first aid treatments and other lost time injuries)**

PROBABILITY
3 - **HIGH (where certain or near curtain harm will occur).**
2 - **MEDIUM (where harm will frequently occur).**
1 - **LOW (where harm will seldom occur)**

RISK RATING = SEVERITY X PROBABILITY
HIGH 6-9 MEDIUM 2-4 LOW 1

CONTROL OF SUBSTANCES HAZARDOUS TO HEALTH

RISK ASSESSMENT FOR THE EXPERIMENTAL RIG

Before filling in this form, please read the *Notes*, as indicated.

This form **MUST BE COMPLETED** prior to the commencement of any work involving risks to health from a hazardous substance, so that a suitable and sufficient assessment of health risks is made.

Note 1 : Persons completing this form should make themselves aware of the Health and Safety Commission Approved Codes of Practice "Control of Substances Hazardous to Health" and "Control of Carcinogenic Substances", the HSE booklet "COSHH Assessments".

PART A   **RISK EVALUATION**

A1	Department	Chemical Engineering
A2	**Title of Work Activity**	Research activity (Ignition energy measurement for gas mixture)

Note:2 Choose a title or give a serial number so as to facilitate departmental filing and/or risk assessments.

A3   **Locations(s) of Work**

2 nd. Pilot laboratory (S – 038)

A4   **Hazardous Substance(s) Classification**
(*Note 3*) Tick 1 or more boxes

Very Toxic	
Toxic	
Harmful	Highly flammable
Corrosive	
Irritant	
Substance(s) with MEL or OES	
Dust	
Carcinogen(or suspect carcinogen)	
Micro-organism	

Specify particularly dangerous or hazardous substance(s) (Note5)

Specify particularly dangerous or hazardous substance(s) (Note5)

Methane (gas), Hydrogen (gas)

Total number of hazardous substances involved in the work activity

2

Note 3: The COSHH Regulations do **NOT** apply where either the Control of Asbestos at Work or theControl of Lead at Work Regulations apply or where the risk to health is solely from radiation noise, pressure, explosive or flammable properties, heat or cold, nor to medicines administered to patients.

A substance should be regarded as hazardous to health if it is hazardous in the form in which it occurs in the work activity, including by-products and waste residues.

(a) Any substance which is listed as "very toxic","toxic","corrosive","harmful", or"irritant" in Part1A of the Approved List for the Classification, Packaging and Labelling of Dangerous Substance Regulations, 1984, (2nd edition onwards) is a substance hazardous to health.

(b) Any substance which has a MEL(Maximum Exposure Limit) or OES(Occupational Exposure Standard) given in the HSE Guidance Note EH.40 (current year date) is a substance hazardous to health.

(c) Micro-organisms which create a hazard to the health of any person, where the hazard arises out of or in connection with a work activity. Hazard classification of pathogens is given in the booklet "Categorisation of Pathogens etc", Advisory Committee on Dangerous Pathogens. See also the relevant 'Advisory Committee on Genetic Manipulation/Health and Safety Executive Notes.

(d) A dust of any kind is a substance hazardous to health when present in a "substantial"concentration. See the Approved Code of Practice,para 2(f) and HSE Guidance Note EH.40.

(e) Any other substance is hazardous if it creates a risk to health comparable to any of the above

Note 4: Refer to the HSC Approved Code of Practice,"Control of Carcinogenic Substances".
Note 5: A Part B2 "Scheme of Work" must be completed for this type of work activity.

A5   Grounds for Concluding Exposure is not a Risk to Health

Quantities or rate of use of substance(s) are too small to constitute any risk to health under foreseeable circumstances or use, even if control measures broke down. (Yes / No) | N |

Note 6: If there are reasonable grounds for reaching the conclusion that risks are insignificant, finish this assessment now by signing page 6.

A6   Route by which the Substances are Hazardous to Health.
(Tick 1 or more boxes)

Inhalation	N/A
Ingestion	N/A
Skin Absorption	N/A
Direct Contact,Skin or eyes	N/A
Injection (via sharps)	N/A

A7   What could be the Effect of Exposure to the above Hazardous Substances?   (Tick boxes)

Single Acute Exposure :	Serious	√
	Not Serious	
	Not Known	
Repeated Low Exposures :	Serious	
	Not Serious	
	Not Known	√
Adverse Effect Could Be :	Long Term	
	Short Term	
	Not Known	√
Effects could be harmful to the Human Reproductive System:	Yes	
	No	√
	Not Known	
The Micro-organism could infect an individual	Yes	
	No	√
	Not Known	
An infected person could infect others	Yes	
	No	√
	Not Known	

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A8   **Engineering Control Measures:**
(Tick 1 or more boxes)

Work to be carried out on an open bench | √ |
Work to be carried out in fume cupboard(s) | |
Specify location(s) of fume cupboards below

Note 7: The fume cupboard must be used in accordance with the University's local guidance or code
of practice

The work will require some other local exhaust ventilation (Yes / No) | Yes |
If Yes please specify below :

EPS detection system to introduce to ------------ extract

The work will be carried out in a glove box (Yes / No) | No |
or other sealed system (Yes / No | Yes |
If Other please specify below :

Contaminant within --------------

The work will be carried out in a laboratory at the required biological containment | N/A |
level (Yes /No)
and in a biological safety cabinet, class(insert number where relevant) | N/A |
ACDP Category | N/A |
Genetic Manipulation Category | N/A |

A9   **Personal Protective Equipment Requirements**
The following personal protective equipment may be necessary for a part or all of the work.

Eye Protection (Yes / No) | Yes |
Face Protection (Yes / No) | No |
Hand Protection (Yes / No) | No |
Foot Protection (Yes / No) | No |
Respiratory Protection (Yes / No) | No |
Other (e.g. protective clothing) (Yes / No) | Yes |
If Other please specify below :

Labcoat

PART B   **DESCRIPTION OF THE WORKING PRACTICE**

Note 8: Part B2 of this form must always be completed for work by postgraduate research students
          and for 3rd and 4th year undergraduates carrying out similar research work.

B1   Instructions for the Work Activity
          (Tick 1 box only)

> The work activity consists of well documented routine procedures carried out
> frequently in a controlled environment and requiring only simple and easily
> understandable verbal instructions

Note 9: Where an assessment of risk is simple and obvious and where the work activity is straight-
          forward and clear verbal instructions can be given easily, a written scheme of work (Part B2)
          is unnecessary. Complete the other sections of Part B.

> The work activity consists of procedures requiring a specific scheme of work. √

Note 5: A Part B2 "Scheme of Work" must be completed for this type of work activity.

B2   Scheme of Work (Continue on a separate sheet, if necessary)

Note 10: The scheme of work is a statement of how the work activity is going to be carried out safely. It
          should specify the ways in which the hazardous substances are to be used or handled, and
          should give sufficient details to identify the precautions necessary to control the risks that
          arise from working with the hazardous substances.

Please refer to operating procedure

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B3   Training for the Work Activity

Specific training will be required :(Yes / No) | Yes |

Note 11: Any special training required to ensure that persons involved in the work activity can operate safely should be described here. This is particularly important so that persons can understand and comply effectively with the scheme of work (B2), where this had been formulated.

> Training in use and handling of pressurised cylinders.
> Training in handling high voltage equipments and interlock system

B4   Supervision

Note 12: The level of supervision must always be appropriate to the competence of the individuals involved in the work activity.

The supervisor will approve straightforward routine work in progress: (Yes / No) | Yes |
The supervisor will specifically approve the scheme of work, B2 (Yes / No) | Yes |
The supervisor will provide supervision personally to control the work (Yes / No) | No |

B5   Monitoring

Note 13: For the majority of work, atmospheric monitoring should not be necessary for protecting health, providing sufficient thought has gone into ensuring the adequacy of control measures in relation to risks, and the control measures are properly used and maintained. For further information on monitoring and health surveillance see the Approved code of Practice, paragraphs 66 to 92, inclusive.

Monitoring for airborne contaminant will be required: (Yes / No) | No |
Biological monitoring of workers will be required: (Yes / No) | No |

B6   Contingency Planning

Note 14: Contingency planning is required to limit the extent of the risk arising from an uncontrolled release of a hazardous substance and for regaining control as quickly as possible.

Written emergency instructions will be provided for workers and others who might be affected, on site: (Yes / No) | No |

Provision of the following may be required in an emergency:

Spill Neutralisation Chemicals: (Yes / No) | No |
If Yes please specify below :

Eye Irritation Point	N/A
Body Showe	N/A
Other First Aid Provisions	N/A
Breathing Apparatus (with trained operator)	N/A
External Emergency Services	N/A
Poison antidote	N/A
If Yes please specify below :	

B7   Disposal of Waste Residues   - (Not Applicable)

In-house to District Council Waste Collection, after rendering safe (Yes / No)
In-house to drain, after rendering safe (Yes / No)
In-house to incinerator, after rendering safe if appropriate (Yes / No)
To specialist licensed Waste Disposal contractor (Yes / No) (Please specify):
Other (e.g. inter-departmental (Yes / No)   (Please specify below):

B8   Implications for other Persons

Academic Staff (Yes / No)	Yes
Postgraduate Staff (Yes / No)	Yes
Postgraduate Students (Yes / No)	Yes
Undergraduate Students (Yes / No)	Yes
Technical Staff (Yes / No)	Yes
Cleaning Staff (Yes / No)	No
Contractor (Yes / No)	No
Visitors (Yes / No)	Yes
Others (Yes / No)	N/A

If Other please specify below :

B9   Accreditation

(i)   Signature of Assessor

                                                     Name   H. Mathurkar   Date   08.05.07

          Signature of Supervisor

                                                     Name   G. Hankinson   Date   08.05.07

(ii)   Signature of All Persons receiving a copy of this Risk Assessment.

Note 15: A copy of this assessment must be given to each postgraduate research student and/or to each
          4th year undergraduate doing like work, and he/she must sign a receipt, at B9(11)

          I/We have received a copy of Parts A and B of this Risk Assessment.

Signature	Name	Date
	Tony Eyre	08.05.2007

(Continue on a separate sheet, if necessary)

(iii)   Date of Next Assessment

1 _____ 2 _____ 3 _____

Note 16: This assessment should be reviewed immediately if there is any reason to suppose that the
          original assessment is no longer valid due to significant changes in the work activity, arising
          for example, from the introduction of new hazardous substances, new personnel, changes in
          procedures or reported ill-health. Otherwise, the assessment should be reviewed annually.

**A·COPY OF THIS ASSESSMENT MUST BE RETAINED BY THE HEAD OF
DEPARTMENT, OR HIS/HER REPRESENTATIVE, FOR AS LONG AS IT IS RELEVANT.**

Operating procedure

This section details the sequence of actions / activities to be performed (Operating procedure) during the experimental test runs. The abbreviations used and the valves referred during the description of the operating procedure are presented in the Figure 3-1.

Start power supply

Switch ON the following devices

- The computer (PC) and start the LabVIEW interface (hnmfinal.VI)
- The Extra High Tension (EHT) pulse generator unit - i.e. Spark generator
- The Oscilloscope TDS 3034 B
- The National Instruments housing box
- The Vacuum pump

Line pressurization
1. Close manifold valves (V1, V2 and V3) and vessel valves (V5 and V6)
2. Open the gas cylinder regulator valves and set flammable gases delivery pressure of around of around 50 kPa (max 10 psig) and the dry air delivery pressure of around 100 kPa (max 15 psig)
3. Ensure coupling CV4 is connected to the vessel

Vessel filling through the method of partial pressure
4. Start the vacuum pump, open valve V5 and turn valve V6 to vacuum pump
5. Evacuate the vessel completely till steady state pressure transducer (P1) reading is obtained
6. Close valves V5 & V6, stop the vacuum pump and record the pressure transducer reading P1, with a click on "AM to File"
7. Slowly open valve V3 to introduce CH_4,(and / or valve V2 for H_2) and record the steady state pressure transducer P1 reading again after closing V3, with a click on "AM to File"
8. Introduce dry air through Valve V1, so that the steady state vessel pressure reaches just above atmospheric after closing V1. Record the pressure transducer (P1) reading with a click on "AM to File"
9. Detach the coupling CV4, to isolate the vessel from the gas cylinders. Observe the pressure transducer - P2 reading
10. Place the gas filler pipe to stored position and lock it

Capacitive Discharge system
11. Insert key and turn it to "SAFE", check the interlock display on EHT pulse generator
12. Ensure the gas filler pipe to stored position (locked position)
13. Open the Electronic Component Unit (ECU) and ground "Monitor Point" in ECU with earth probe. If necessary change resistor and discharge capacitor
14. Close and latch the ECU
15. Check interlock shows - CLOSED, Switch key lock to ACTIVE and Ready for Ignition

Ignition

16. Ignite gas mixture from LabVIEW control panel with a click on "Fire & Record" button
17. LabVIEW programme records the pressure (transducer - P2) and temperature (thermocouple T1) rise as function of time in separate files out_press.LMV and out_temp.LMV respectively.
18. Transient values of Voltage and Current will be displayed by oscilloscope. The peak value of voltage (during spark initiation phase) as displayed by oscilloscope is recorded manually
19. If the gas mixture is ignited by spark (confirmed through pressure and temperature rise) then ref. step no. 21, to evacuate the vessel and repeat experiments with fresh gas air mixture
20. If the gas mixture did not ignite, then increase the spark energy using larger capacitor in ECU box as follows:
    - Open ECU, Check interlock show "OPEN". This will operate discharge circuits in the EHT pulse generator
    - Ground "Monitor Point" in ECU with earth probe (this will remove any residual charge in the discharge capacitor). Change the capacitor and resistors as desired
    - Repeat step no. 16. A maximum of four such ignitions attempted before finally evacuating the vessel (step no. 21) and starting with fresh gas air mixture

Exhaust
21. Open valve V5
22. Open valve V6 to atmosphere (to vent excess pressure − if any - to atmosphere)
23. Repeat procedure from step 4 to 20 with different (or same) composition of flammable gas - air mixture and/or Capacitor / Resistor combination

The test runs were repeated using various concentrations of CH_4, H_2 and Air.

Shutdown (end experiments for the day)
For Electrical system

- Open ECU, Check interlocks show "OPEN". This will operate discharge circuits in the EHT pulse generator
- Ground "Monitor Point" in ECU with earth probe (this will remove any residual charge in the discharge capacitor)
- Switch key lock to "SAFE"
- Switch OFF the EHT pulse generator power
- If leaving the equipment unattended, remove key

For Mechanical system
- Ensure all valves (V1, V2, V3, V5 and V6) are closed and the coupling CV4 is attached to the vessel
- Turn off cylinder regulators and depressurise all the lines completely by opening valves V1, V2 and V3 one at a time into the vessel.
- Depressurise vessel by opening valves V5 and V6 to atmosphere, check the pressure transducer readings (P1 & P2) and again close valves V5 and V6 completely

Electrical circuit and Instrumentation

Figure F.1 : Rectifier circuit for the spark generation

Figure F.2 : Block diagram for the LabVIEW programme

Specifications of the Pressure Transducers

Figure G.1: Calibration curve for the Pressure transducer (P1)

Figure G.2 : Calibration curve for the Pressure transducer (P2)

Table H.1 : Experimental runs with methane – air mixture

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
5.60	0	5.60	94.40	3.84	No
5.60	0	5.60	94.40	8.28	No
5.60	0	5.60	94.40	10.73	No
5.60	0	5.60	94.40	13.13	No
5.78	0	5.78	94.22	4.03	No
5.78	0	5.78	94.22	5.69	No
5.78	0	5.78	94.22	10.26	No
5.78	0	5.78	94.22	6.98	No
5.81	0	5.81	94.19	3.92	No
5.81	0	5.81	94.19	3.16	No
5.81	0	5.81	94.19	11.62	No
5.81	0	5.81	94.19	6.28	No
5.82	0	5.82	94.18	3.57	No
5.82	0	5.82	94.18	11.30	No
5.82	0	5.82	94.18	12.25	No
5.82	0	5.82	94.18	6.28	No
6.26	0	6.26	93.74	3.56	No
6.26	0	6.26	93.74	8.24	No
6.26	0	6.26	93.74	7.38	No
6.26	0	6.26	93.74	4.98	No
5.62	0	5.62	94.38	9.07	No
5.62	0	5.62	94.38	8.46	No
5.62	0	5.62	94.38	12.25	No
5.62	0	5.62	94.38	6.74	No
5.78	0	5.78	94.22	3.57	No
5.78	0	5.78	94.22	6.76	No
5.78	0	5.78	94.22	9.69	No
5.78	0	5.78	94.22	6.98	No
5.81	0	5.81	94.19	3.98	No
5.81	0	5.81	94.19	4.69	No
5.81	0	5.81	94.19	13.73	No
5.81	0	5.81	94.19	6.74	No
5.79	0	5.79	94.21	3.56	No
5.79	0	5.79	94.21	11.52	No
5.79	0	5.79	94.21	11.68	No
5.79	0	5.79	94.21	9.59	No
6.01	0	6.01	93.99	4.53	No
6.01	0	6.01	93.99	11.30	No
6.01	0	6.01	93.99	7.84	No
6.01	0	6.01	93.99	7.22	No
6.16	0	6.16	93.84	2.27	No
6.16	0	6.16	93.84	6.75	No
6.16	0	6.16	93.84	5.88	No
6.16	0	6.16	93.84	11.96	Yes
6.21	0	6.21	93.79	2.72	No
6.21	0	6.21	93.79	12.64	No
6.21	0	6.21	93.79	9.79	No
6.21	0	6.21	93.79	9.12	No
6.28	0	6.28	93.72	3.57	No
6.28	0	6.28	93.72	7.46	No

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
6.28	0	6.28	93.72	11.12	No
6.28	0	6.28	93.72	7.47	No
6.03	0	6.03	93.97	1.99	No
6.03	0	6.03	93.97	6.59	No
6.03	0	6.03	93.97	8.28	No
6.03	0	6.03	93.97	13.13	No
6.15	0	6.15	93.85	2.82	No
6.15	0	6.15	93.85	12.88	No
6.15	0	6.15	93.85	10.85	No
6.15	0	6.15	93.85	9.31	No
6.21	0	6.21	93.79	3.56	No
6.21	0	6.21	93.79	7.23	No
6.21	0	6.21	93.79	10.31	No
6.21	0	6.21	93.79	6.74	No
6.21	0	6.21	93.79	2.20	No
6.21	0	6.21	93.79	5.70	No
6.21	0	6.21	93.79	8.62	No
6.21	0	6.21	93.79	10.79	No
6.15	0	6.15	93.85	2.64	No
6.15	0	6.15	93.85	12.42	No
6.15	0	6.15	93.85	10.79	No
6.15	0	6.15	93.85	6.98	No
6.10	0	6.10	93.90	3.30	No
6.10	0	6.10	93.90	7.22	No
6.10	0	6.10	93.90	11.40	No
6.10	0	6.10	93.90	6.98	No .
6.24	0	6.24	93.76	2.27	No
6.24	0	6.24	93.76	5.35	No
6.24	0	6.24	93.76	8.65	No
6.24	0	6.24	93.76	11.68	No
6.00 (average)	0 (average)	6.00 (average)	94.00 (average)	11.96 (Lowest Energy)	
6.97	0	6.97	93.03	3.89	No
6.88	0	6.88	93.12	5.74	Yes
7.23	0	7.23	92.77	4.39	Yes
7.05	0	7.05	92.95	7.73	Yes
6.81	0	6.81	93.19	4.02	Yes
6.64	0	6.64	93.36	12.19	Yes
6.64	0	6.64	93.36	4.55	Yes
6.64	0	6.64	93.36	8.09	Yes
6.25	0	6.25	93.75	12.64	No
6.72	0	6.72	93.28	4.02	No
6.94	0	6.94	93.06	10.87	Yes
6.81	0	6.81	93.19	4.83	Yes
6.59	0	6.59	93.41	3.89	Yes
6.68	0	6.68	93.32	3.95	Yes
6.42	0	6.42	93.58	4.15	Yes
6.27	0	6.27	93.73	6.00	Yes
6.40	0	6.40	93.60	4.78	No
6.22	0	6.22	93.78	4.83	No
6.27	0	6.27	93.73	4.28	Yes
6.99	0	6.99	93.01	4.28	No
6.85	0	6.85	93.15	4.69	Yes
6.52	0	6.52	93.48	6.36	No
6.66	0	6.66	93.34	4.02	Yes
6.51	0	6.51	93.49	5.28	Yes
6.42	0	6.42	93.58	5.43	No
6.44	0	6.44	93.56	6.87	No
6.45	0	6.45	93.55	9.83	Yes
6.65	0	6.65	93.35	13.34	Yes
6.65	0	6.65	93.35	12.42	Yes
6.51	0	6.51	93.49	4.83	Yes

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
6.65	0	6.65	93.35	4.99	Yes
6.27	0	6.27	93.73	12.42	No
6.22	0	6.22	93.78	6.69	No
6.30	0	6.30	93.70	7.57	No
6.51	0	6.51	93.49	9.06	No
6.21	0	6.21	93.79	4.55	Yes
6.37	0	6.37	93.63	3.82	No
6.36	0	6.36	93.64	3.89	No
6.71	0	6.71	93.29	7.37	Yes
6.76	0	6.76	93.24	7.91	No
6.11	0	6.11	93.89	4.98	Yes
6.72	0	6.72	93.28	10.87	Yes
6.39	0	6.39	93.61	12.42	No
6.77	0	6.77	93.23	8.11	No
6.19	0	6.19	93.81	6.52	No
6.38	0	6.38	93.62	10.87	Yes
6.29	0	6.29	93.71	12.64	Yes
6.27	0	6.27	93.73	4.83	Yes
6.49	0	6.49	93.51	6.69	No
6.36	0	6.36	93.64	6.67	Yes
6.49	0	6.49	93.51	10.87	Yes
6.31	0	6.31	93.69	6.36	Yes
6.46	0	6.46	93.54	6.52	Yes
6.63	0	6.63	93.37	7.03	No
6.75	0	6.75	93.25	4.83	Yes
6.49	0	6.49	93.51	6.36	No
6.39	0	6.39	93.61	5.27	No
6.24	0	6.24	93.76	4.57	Yes
6.53	0	6.53	93.47	4.28	No
6.29	0	6.29	93.71	4.69	Yes
6.47	0	6.47	93.53	6.36	No
6.34	0	6.34	93.66	6.20	Yes
6.38	0	6.38	93.62	10.04	No
6.31	0	6.31	93.69	10.87	Yes
6.65	0	6.65	93.35	10.41	No
6.24	0	6.24	93.76	4.02	No
6.72	0	6.72	93.28	4.08	Yes
6.49	0	6.49	93.51	4.28	No
6.87	0	6.87	93.13	3.95	Yes
6.79	0	6.79	93.21	5.27	No
6.65	0	6.65	93.35	4.83	No
6.61	0	6.61	93.39	5.57	No
6.64	0	6.64	93.36	4.98	Yes
7.08	0	7.08	92.92	4.83	Yes
6.49	0	6.49	93.51	4.69	No
6.90	0	6.90	93.10	5.43	Yes
6.47	0	6.47	93.53	5.25	Yes
6.74	0	6.74	93.26	4.83	No
6.68	0	6.68	93.32	4.83	No
7.08	0	7.08	92.92	4.28	No
6.72	0	6.72	93.28	1.66	No
7.37	0	7.37	92.63	1.55	No
6.65	0	6.65	93.35	2.80	No
6.67	0	6.67	93.33	1.31	No
6.87	0	6.87	93.13	3.34	No
6.51	0	6.51	93.49	1.11	No
6.93	0	6.93	93.07	4.34	No
6.86	0	6.86	93.14	2.88	No
6.85	0	6.85	93.15	1.81	Yes
6.31	0	6.31	93.69	3.38	No
6.61	0	6.61	93.39	5.95	Yes
6.80	0	6.80	93.20	4.84	No
6.74	0	6.74	93.26	4.13	Yes
6.38	0	6.38	93.62	2.78	No
6.40	0	6.40	93.60	3.05	No
6.91	0	6.91	93.09	1.80	No
7.09	0	7.09	92.91	4.53	Yes
6.44	0	6.44	93.56	2.28	No
6.20	0	6.20	93.80	5.83	No

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CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
6.23	0	6.23	93.77	5.16	No
6.66	0	6.66	93.34	4.32	No
6.92	0	6.92	93.08	3.56	No
6.72	0	6.72	93.28	4.23	No
6.66	0	6.66	93.34	3.57	No
6.53	0	6.53	93.47	2.27	No
6.80	0	6.80	93.20	2.88	No
6.80	0	6.80	93.20	5.00	Yes
6.70	0	6.70	93.30	2.04	No
6.80	0	6.80	93.20	6.92	No
6.94	0	6.94	93.06	4.53	No
6.38	0	6.38	93.62	4.84	No
6.91	0	6.91	93.09	2.27	Yes
6.88	0	6.88	93.12	2.41	No
6.86	0	6.86	93.14	2.64	No
7.03	0	7.03	92.97	3.21	No
6.35	0	6.35	93.65	2.38	No
6.34	0	6.34	93.66	2.43	No
6.38	0	6.38	93.62	2.20	Yes
6.69	0	6.69	93.31	2.41	No
6.77	0	6.77	93.23	2.57	No
6.85	0	6.85	93.15	1.82	Yes
6.62	0	6.62	93.38	2.13	Yes
6.40	0	6.40	93.60	2.35	No
6.38	0	6.38	93.62	1.80	No
6.61	0	6.61	93.39	2.20	No
6.79	0	6.79	93.21	3.74	Yes
6.69	0	6.69	93.31	3.13	No
6.79	0	6.79	93.21	2.57	No
6.58	0	6.58	93.42	3.29	Yes
6.58	0	6.58	93.42	3.08	No
6.73	0	6.73	93.27	0.40	No
6.89	0	6.89	93.11	0.55	No
6.72	0	6.72	93.28	0.50	No
6.77	0	6.77	93.23	0.78	No
7.01	0	7.01	92.99	0.39	No
6.57	0	6.57	93.43	0.30	No
7.07	0	7.07	92.93	0.94	No
6.96	0	6.96	93.04	1.17	No
6.91	0	6.91	93.09	0.50	No
6.43	0	6.43	93.57	0.87	No
6.85	0	6.85	93.15	0.32	No
6.14	0	6.14	93.86	0.44	No
6.89	0	6.89	93.11	0.75	No
7.01	0	7.01	92.99	0.52	No
7.01	0	7.01	92.99	0.24	No
7.19	0	7.19	92.81	0.99	No
6.47	0	6.47	93.53	1.17	No
6.31	0	6.31	93.69	1.26	No
6.25	0	6.25	93.75	0.44	No
6.95	0	6.95	93.05	0.55	No
6.70	0	6.70	93.30	0.48	No
7.00	0	7.00	93.00	0.80	No
6.81	0	6.81	93.19	0.41	No
6.69	0	6.69	93.31	0.30	No
6.68	0	6.68	93.32	0.87	No
6.60	0	6.60	93.40	0.82	No
6.84	0	6.84	93.16	0.67	No
6.84	0	6.84	93.16	0.43	No
6.72	0	6.72	93.28	0.45	No
6.84	0	6.84	93.16	0.43	No
7.05	0	7.05	92.95	0.27	No
6.46	0	6.46	93.54	0.61	No
6.93	0	6.93	93.07	0.55	No
6.95	0	6.95	93.05	0.87	No
6.35	0	6.35	93.65	0.53	No
6.43	0	6.43	93.57	0.70	No
6.38	0	6.38	93.62	0.32	No
6.35	0	6.35	93.65	0.35	No

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
7.24	0	7.24	92.76	0.78	No
6.74	0	6.74	93.26	0.32	No
6.70	0	6.70	93.30	1.17	No
6.45	0	6.45	93.55	0.23	No
6.42	0	6.42	93.58	0.40	No
6.72	0	6.72	93.28	0.55	No
6.94	0	6.94	93.06	0.50	No
6.80	0	6.80	93.20	0.78	No
6.92	0	6.92	93.08	0.46	No
6.60	0	6.60	93.40	0.94	No
7.02	0	7.02	92.98	0.99	No
6.65	0	6.65	93.35	0.34	No
6.91	0	6.91	93.09	2.32	No
6.91	0	6.91	93.09	4.13	No
6.91	0	6.91	93.09	7.20	No
6.91	0	6.91	93.09	11.26	No
6.63	0	6.63	93.37	2.27	No
6.63	0	6.63	93.37	5.27	No
6.63	0	6.63	93.37	11.19	No
6.63	0	6.63	93.37	11.40	No
6.62	0	6.62	93.38	2.76	No
6.62	0	6.62	93.38	3.93	No
6.62	0	6.62	93.38	9.63	No
6.62	0	6.62	93.38	13.13	Yes
6.77	0	6.77	93.23	0.94	No
6.77	0	6.77	93.23	6.09	No
6.77	0	6.77	93.23	7.91	No
6.77	0	6.77	93.23	10.79	Yes
6.70	0	6.70	93.30	2.37	No
6.70	0	6.70	93.30	6.30	No
6.70	0	6.70	93.30	8.08	Yes
6.84	0	6.84	93.16	0.15	No
6.84	0	6.84	93.16	3.54	No
6.84	0	6.84	93.16	3.39	No
6.84	0	6.84	93.16	10.22	Yes
6.66	0	6.66	93.34	0.52	No
6.66	0	6.66	93.34	2.53	No
6.66	0	6.66	93.34	4.44	Yes
6.67	0	6.67	93.33	6.06	No
6.67	0	6.67	93.33	3.99	No
6.67	0	6.67	93.33	8.28	Yes
6.68	0	6.68	93.32	1.65	No
6.68	0	6.68	93.32	2.34	No
6.68	0	6.68	93.32	3.89	No
6.68	0	6.68	93.32	8.35	Yes
6.85	0	6.85	93.15	0.72	No
6.85	0	6.85	93.15	1.82	Yes
6.80	0	6.80	93.20	0.43	No
6.80	0	6.80	93.20	6.30	No
6.80	0	6.80	93.20	6.04	No
6.80	0	6.80	93.20	8.07	No
6.59	0	6.59	93.41	0.45	No
6.59	0	6.59	93.41	8.09	No
6.59	0	6.59	93.41	6.21	No
6.59	0	6.59	93.41	7.41	No

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
6.58	0	6.58	93.42	1.43	No
6.58	0	6.58	93.42	2.20	No
6.58	0	6.58	93.42	3.64	No
6.58	0	6.58	93.42	8.54	Yes
6.42	0	6.42	93.58	0.34	No
6.42	0	6.42	93.58	1.92	Yes
6.88	0	6.88	93.12	0.32	No
6.88	0	6.88	93.12	1.94	No
6.88	0	6.88	93.12	3.04	No
6.88	0	6.88	93.12	7.41	No
6.55	0	6.55	93.45	0.30	No
6.55	0	6.55	93.45	2.06	No
6.55	0	6.55	93.45	3.76	No
6.55	0	6.55	93.45	8.31	No
6.51	0	6.51	93.49	0.40	No
6.51	0	6.51	93.49	5.95	No
6.51	0	6.51	93.49	3.76	No
6.51	0	6.51	93.49	7.61	No
6.52	0	6.52	93.48	0.28	No
6.52	0	6.52	93.48	1.98	No
6.52	0	6.52	93.48	4.42	No
6.52	0	6.52	93.48	8.54	No
6.92	0	6.92	93.08	0.72	No
6.92	0	6.92	93.08	2.64	No
6.92	0	6.92	93.08	4.53	Yes
6.66	0	6.66	93.34	2.42	No
6.66	0	6.66	93.34	3.48	No
6.66	0	6.66	93.34	4.96	No
6.66	0	6.66	93.34	8.84	Yes
6.69 (average)	0 (average)	6.69 (average)	93.31 (average)	1.82 (Lowest Energy)	
7.71	0	7.71	92.29	0.17	No
7.71	0	7.71	92.29	1.12	No
7.71	0	7.71	92.29	1.45	No
7.71	0	7.71	92.29	3.74	Yes
7.80	0	7.80	92.20	0.34	No
7.80	0	7.80	92.20	2.70	No
7.80	0	7.80	92.20	2.48	Yes
7.80	0	7.80	92.20	0.00	No
7.72	0	7.72	92.28	0.27	No
7.72	0	7.72	92.28	1.47	No
7.72	0	7.72	92.28	3.88	No
7.72	0	7.72	92.28	6.73	No
7.76	0	7.76	92.24	0.35	No
7.76	0	7.76	92.24	2.81	No
7.76	0	7.76	92.24	2.20	Yes
7.76	0	7.76	92.24	0.00	No
7.75	0	7.75	92.25	0.27	No
7.75	0	7.75	92.25	1.78	No
7.75	0	7.75	92.25	11.74	No
7.75	0	7.75	92.25	5.32	No
7.77	0	7.77	92.23	0.30	No
7.77	0	7.77	92.23	0.32	No
7.77	0	7.77	92.23	1.48	Yes
7.95	0	7.95	92.05	0.30	No
7.95	0	7.95	92.05	0.34	No

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
7.95	0	7.95	92.05	1.43	Yes
7.92	0	7.92	92.08	0.34	No
7.92	0	7.92	92.08	0.66	No
7.92	0	7.92	92.08	3.22	Yes
7.89	0	7.89	92.11	0.30	No
7.89	0	7.89	92.11	0.76	No
7.89	0	7.89	92.11	3.41	Yes
7.96	0	7.96	92.04	0.31	Yes
8.08	0	8.08	91.92	0.31	No
8.08	0	8.08	91.92	0.59	Yes
8.12	0	8.12	91.88	0.28	No
8.12	0	8.12	91.88	0.26	No
8.12	0	8.12	91.88	1.48	Yes
8.12	0	8.12	91.88	0.88	No
8.12	0	8.12	91.88	0.99	No
8.12	0	8.12	91.88	2.64	No
8.12	0	8.12	91.88	3.75	Yes
8.06	0	8.06	91.94	0.56	No
8.06	0	8.06	91.94	0.99	No
8.06	0	8.06	91.94	0.38	Yes
7.84	0	7.84	92.16	0.24	No
7.84	0	7.84	92.16	0.46	Yes
8.12	0	8.12	91.88	0.25	No
8.12	0	8.12	91.88	0.43	Yes
8.19	0	8.19	91.81	0.61	No
8.19	0	8.19	91.81	0.99	No
8.19	0	8.19	91.81	0.24	Yes
8.12	0	8.12	91.88	0.44	Yes
8.12	0	8.12	91.88	0.60	No
8.12	0	8.12	91.88	0.93	No
8.12	0	8.12	91.88	0.29	Yes
8.15	0	8.15	91.85	0.96	Yes
7.96 (average)	0 (average)	7.96 (average)	92.04 (average)	0.24 (Lowest Energy)	
8.63	0	8.63	91.37	0.17	No
8.63	0	8.63	91.37	1.41	Yes
8.66	0	8.66	91.34	0.16	No
8.66	0	8.66	91.34	0.55	Yes
8.64	0	8.64	91.36	0.19	No
8.64	0	8.64	91.36	1.73	No
8.64	0	8.64	91.36	4.54	No
8.64	0	8.64	91.36	5.55	No
8.74	0	8.74	91.26	0.27	No
8.74	0	8.74	91.26	0.55	No
8.74	0	8.74	91.26	2.27	Yes
8.75	0	8.75	91.25	0.93	No
8.75	0	8.75	91.25	0.27	Yes
8.79	0	8.79	91.21	0.42	No
8.79	0	8.79	91.21	0.22	Yes
8.81	0	8.81	91.19	0.32	Yes

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
8.99	0	8.99	91.01	0.45	Yes
8.85	0	8.85	91.15	0.30	Yes
9.07	0	9.07	90.93	0.29	Yes
9.11	0	9.11	90.89	0.34	No
9.11	0	9.11	90.89	0.61	Yes
9.14	0	9.14	90.86	0.53	No
9.14	0	9.14	90.86	1.51	Yes
9.14	0	9.14	90.86	0.43	No
9.14	0	9.14	90.86	1.04	Yes
9.02	0	9.02	90.98	0.27	No
9.02	0	9.02	90.98	0.46	Yes
9.03	0	9.03	90.97	0.41	No
9.03	0	9.03	90.97	0.70	Yes
9.07	0	9.07	90.93	0.44	No
9.07	0	9.07	90.93	0.45	Yes
9.18	0	9.18	90.82	0.33	No
9.18	0	9.18	90.82	0.44	Yes
9.21	0	9.21	90.79	0.70	No
9.21	0	9.21	90.79	0.36	Yes
9.13	0	9.13	90.87	0.16	No
9.13	0	9.13	90.87	0.76	Yes
9.11	0	9.11	90.89	0.49	No
9.11	0	9.11	90.89	0.87	No
9.11	0	9.11	90.89	0.82	Yes
8.95 (average)	0 (average)	8.95 (average)	91.05 (average)	0.22 (Lowest Energy)	
10.38	0	10.38	89.62	0.82	No
10.38	0	10.38	89.62	0.99	Yes
10.65	0	10.65	89.35	0.93	No
10.65	0	10.65	89.35	3.41	Yes
10.57	0	10.57	89.43	0.68	No
10.57	0	10.57	89.43	5.02	Yes
10.57	0	10.57	89.43	0.22	No
10.57	0	10.57	89.43	2.06	No
10.57	0	10.57	89.43	5.16	Yes
10.78	0	10.78	89.22	1.04	No
10.78	0	10.78	89.22	0.83	Yes
10.71	0	10.71	89.29	0.70	No
10.71	0	10.71	89.29	1.28	No
10.71	0	10.71	89.29	3.04	Yes
10.60	0	10.60	89.40	1.17	No
10.60	0	10.60	89.40	2.32	No
10.60	0	10.60	89.40	7.99	No
10.60	0	10.60	89.40	6.20	Yes
10.63	0	10.63	89.37	0.23	No
10.63	0	10.63	89.37	2.42	No
10.63	0	10.63	89.37	3.64	No
10.63	0	10.63	89.37	7.84	No
10.64	0	10.64	89.36	0.60	No

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
10.64	0	10.64	89.36	1.06	No
10.64	0	10.64	89.36	1.52	Yes
10.62	0	10.62	89.38	0.91	No
10.62	0	10.62	89.38	7.67	No
10.62	0	10.62	89.38	6.36	Yes
10.57	0	10.57	89.43	0.93	No
10.57	0	10.57	89.43	1.15	Yes
10.57	0	10.57	89.43	0.00	No
10.62	0	10.62	89.38	0.85	No
10.62	0	10.62	89.38	5.33	No
10.62	0	10.62	89.38	6.52	Yes
10.75	0	10.75	89.25	0.62	No
10.75	0	10.75	89.25	3.75	No
10.75	0	10.75	89.25	6.52	No
10.75	0	10.75	89.25	6.07	No
10.62	0	10.62	89.38	0.42	No
10.62	0	10.62	89.38	2.53	No
10.62	0	10.62	89.38	10.04	No
10.53	0	10.53	89.47	0.44	No
10.53	0	10.53	89.47	0.55	No
10.53	0	10.53	89.47	2.93	Yes
10.58	0	10.58	89.42	0.80	No
10.58	0	10.58	89.42	4.50	No
10.58	0	10.58	89.42	12.64	No
10.58	0	10.58	89.42	10.85	Yes
10.69	0	10.69	89.31	0.76	No
10.69	0	10.69	89.31	8.37	No
10.69	0	10.69	89.31	8.28	Yes
10.63	0	10.63	89.37	0.83	No
10.63	0	10.63	89.37	4.00	No
10.63	0	10.63	89.37	6.46	Yes
10.71	0	10.71	89.29	0.55	No
10.71	0	10.71	89.29	2.20	No
10.71	0	10.71	89.29	9.79	Yes
10.82	0	10.82	89.18	0.50	No
10.82	0	10.82	89.18	2.57	No
10.82	0	10.82	89.18	9.79	No
10.82	0	10.82	89.18	7.98	No
10.63 (average)	0 (average)	10.63 (average)	89.37 (average)	0.83 (Lowest Energy)	

Table H.2 : Experimental runs with hydrogen– air mixture

H₂ conc. (%)	CH₄ conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
5.48	0	5.48	94.52	0.73	No
5.48	0	5.48	94.52	0.51	No
5.48	0	5.48	94.52	1.56	No
5.48	0	5.48	94.52	6.36	No
5.76	0	5.76	94.24	0.51	No
5.76	0	5.76	94.24	4.00	No
5.76	0	5.76	94.24	8.76	No
5.76	0	5.76	94.24	10.08	No
5.77	0	5.77	94.23	1.39	No
5.77	0	5.77	94.23	7.91	No
5.77	0	5.77	94.23	6.10	No
5.77	0	5.77	94.23	12.54	No
5.92	0	5.92	94.08	1.18	No
5.92	0	5.92	94.08	2.87	No
5.92	0	5.92	94.08	6.69	No
5.92	0	5.92	94.08	9.79	No
6.00	0	6.00	94.00	0.79	No
6.00	0	6.00	94.00	2.32	No
6.00	0	6.00	94.00	12.42	No
6.00	0	6.00	94.00	8.76	No
6.12	0	6.12	93.88	0.69	No
6.12	0	6.12	93.88	2.37	No
6.12	0	6.12	93.88	12.64	No
6.12	0	6.12	93.88	9.53	No
6.17	0	6.17	93.83	0.58	No
6.17	0	6.17	93.83	3.41	No
6.17	0	6.17	93.83	3.50	No
6.17	0	6.17	93.83	13.76	No
6.16	0	6.16	93.84	1.39	No
6.16	0	6.16	93.84	7.91	No
6.16	0	6.16	93.84	6.10	No
6.16	0	6.16	93.84	11.96	Yes
6.19	0	6.19	93.81	0.37	No
6.19	0	6.19	93.81	2.23	No
6.19	0	6.19	93.81	4.06	No
6.19	0	6.19	93.81	4.00	No
6.22	0	6.22	93.78	0.44	No
6.22	0	6.22	93.78	2.53	No
6.22	0	6.22	93.78	11.52	No
6.22	0	6.22	93.78	9.28	No
6.25	0	6.25	93.75	0.44	No
6.25	0	6.25	93.75	2.53	No
6.25	0	6.25	93.75	11.52	No
6.25	0	6.25	93.75	9.28	No
6.30	0	6.30	93.70	1.06	No
6.30	0	6.30	93.70	0.76	Yes
6.40	0	6.40	93.60	0.18	No
6.40	0	6.40	93.60	1.65	No
6.40	0	6.40	93.60	6.07	No
6.40	0	6.40	93.60	6.10	No

H₂ conc. (%)	CH₄ conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
6.54	0	6.54	93.46	0.50	No
6.54	0	6.54	93.46	2.37	No
6.54	0	6.54	93.46	11.09	No
6.54	0	6.54	93.46	9.28	No
6.78	0	6.78	93.22	1.06	No
6.78	0	6.78	93.22	6.30	No
6.78	0	6.78	93.22	8.99	No
6.78	0	6.78	93.22	10.22	No
6.82	0	6.82	93.18	0.18	No
6.82	0	6.82	93.18	1.65	No
6.82	0	6.82	93.18	6.07	No
6.82	0	6.82	93.18	6.10	No
6.90	0	6.90	93.10	1.67	No
6.90	0	6.90	93.10	4.69	No
6.90	0	6.90	93.10	8.62	No
6.90	0	6.90	93.10	9.04	No
6.92	0	6.92	93.08	0.33	No
6.92	0	6.92	93.08	2.83	No
6.92	0	6.92	93.08	2.36	No
6.92	0	6.92	93.08	10.87	No
6.99	0	6.99	93.01	1.18	No
6.99	0	6.99	93.01	2.87	No
6.99	0	6.99	93.01	5.25	No
6.99	0	6.99	93.01	5.60	Yes
6.98	0	6.98	93.02	1.87	No
6.98	0	6.98	93.02	4.43	No
6.98	0	6.98	93.02	7.20	No
6.98	0	6.98	93.02	10.88	No
6.32 (average)	0 (average)	6.32 (average)	93.67 (average)	0.76 (Lowest Energy)	
9.44	0	9.44	90.56	0.28	No
9.44	0	9.44	90.56	1.31	No
9.44	0	9.44	90.56	3.14	No
9.44	0	9.44	90.56	9.01	No
9.59	0	9.59	90.41	0.26	No
9.59	0	9.59	90.41	0.22	No
9.59	0	9.59	90.41	1.05	Yes
9.61	0	9.61	90.39	0.25	No
9.61	0	9.61	90.39	1.08	Yes
9.74	0	9.74	90.26	0.10	Yes
9.58	0	9.58	90.42	0.37	No
9.58	0	9.58	90.42	0.34	No
9.58	0	9.58	90.42	1.02	Yes
9.64	0	9.64	90.36	0.30	No
9.64	0	9.64	90.36	0.34	No
9.64	0	9.64	90.36	1.48	Yes
9.75	0	9.75	90.25	0.17	No
9.75	0	9.75	90.25	0.70	No
9.75	0	9.75	90.25	2.27	No
9.75	0	9.75	90.25	2.32	Yes
9.94	0	9.94	90.06	0.41	No
9.94	0	9.94	90.06	0.31	No
9.94	0	9.94	90.06	2.11	No
9.94	0	9.94	90.06	0.62	Yes
10.10	0	10.10	89.90	0.38	No

H$_2$ conc. (%)	CH$_4$ conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
10.10	0	10.10	89.90	0.72	No
10.10	0	10.10	89.90	1.28	No
10.10	0	10.10	89.90	1.56	Yes
10.27	0	10.27	89.73	1.08	No
10.27	0	10.27	89.73	2.06	No
10.27	0	10.27	89.73	2.34	No
10.27	0	10.27	89.73	4.63	Yes
10.39	0	10.39	89.61	0.57	No
10.39	0	10.39	89.61	0.72	Yes
10.70	0	10.70	89.30	0.30	No
10.70	0	10.70	89.30	1.59	No
10.70	0	10.70	89.30	3.76	No
10.70	0	10.70	89.30	3.91	Yes
11.04	0	11.04	88.96	0.13	No
11.04	0	11.04	88.96	1.27	No
11.04	0	11.04	88.96	1.33	No
11.04	0	11.04	88.96	4.39	No
11.22	0	11.22	88.78	1.18	No
11.22	0	11.22	88.78	0.68	No
11.22	0	11.22	88.78	1.24	No
11.22	0	11.22	88.78	2.81	Yes
11.15	0	11.15	88.85	0.24	No
11.15	0	11.15	88.85	0.27	Yes
11.64	0	11.64	88.36	0.15	No
11.64	0	11.64	88.36	0.13	No
11.64	0	11.64	88.36	0.38	Yes
11.50	0	11.50	88.50	0.29	No
11.50	0	11.50	88.50	0.39	Yes
11.68	0	11.68	88.32	0.16	No
11.68	0	11.68	88.32	0.19	No
11.68	0	11.68	88.32	0.22	No
11.68	0	11.68	88.32	0.15	Yes
12.02	0	12.02	87.98	0.30	No
12.02	0	12.02	87.98	0.38	No
12.02	0	12.02	87.98	0.27	No
12.02	0	12.02	87.98	3.67	Yes
12.73	0	12.73	87.27	0.55	No
12.73	0	12.73	87.27	0.66	Yes
10.65 (average)	0 (average)	10.65 (average)	89.35 (average)	0.10 (Lowest Energy)	
19.51	0	19.51	80.49	0.37	Yes
19.67	0	19.67	80.33	0.33	Yes
19.78	0	19.78	80.22	0.15	Yes
20.13	0	20.13	79.87	0.13	Yes
20.31	0	20.31	79.69	0.20	No
20.31	0	20.31	79.69	0.14	Yes
20.46	0	20.46	79.54	0.20	Yes
20.95	0	20.95	79.05	0.16	Yes
20.30	0	20.30	79.70	0.11	No
20.30	0	20.30	79.70	0.07	Yes

H₂ conc. (%)	CH₄ conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
21.10	0	21.10	78.90	0.27	No
21.10	0	21.10	78.90	0.14	Yes
21.70	0	21.70	78.30	0.15	No
21.70	0	21.70	78.30	0.12	Yes
21.87	0	21.87	78.13	0.20	No
21.87	0	21.87	78.13	0.14	Yes
21.18	0	21.18	78.82	0.13	Yes
21.19	0	21.19	78.81	0.04	No
21.19	0	21.19	78.81	0.04	Yes
21.25	0	21.25	78.75	0.87	Yes
21.25	0	21.25	78.75	0.55	Yes
21.28	0	21.28	78.72	0.08	Yes
22.51	0	22.51	77.49	0.36	Yes
21.56	0	21.56	78.44	0.20	No
21.56	0	21.56	78.44	0.14	Yes
21.60	0	21.60	78.40	0.15	Yes
21.72	0	21.72	78.28	0.09	Yes
20.96 (average)	0 (average)	20.96 (average)	79.04 (average)	0.04 (Lowest Energy)	
24.09	0	24.09	75.91	0.05	No
24.09	0	24.09	75.91	0.36	Yes
24.38	0	24.38	75.62	0.09	No
24.38	0	24.38	75.62	0.03	Yes
24.41	0	24.41	75.59	0.15	Yes
24.53	0	24.53	75.47	0.08	Yes
25.51	0	25.51	74.49	0.10	No
25.51	0	25.51	74.49	0.10	Yes
25.51	0	25.51	74.49	0.05	Yes
25.72	0	25.72	74.28	0.08	Yes
25.74	0	25.74	74.26	0.10	Yes
25.77	0	25.77	74.23	0.12	Yes
25.80	0	25.80	74.20	0.17	Yes
25.65	0	25.65	74.35	0.16	Yes
26.11	0	26.11	73.89	0.03	Yes
26.19	0	26.19	73.81	0.13	No
26.19	0	26.19	73.81	0.26	Yes
26.24	0	26.24	73.76	0.11	Yes
25.60	0	25.60	74.40	0.12	Yes
26.07	0	26.07	73.93	0.11	Yes
25.60	0	25.60	74.40	0.11	Yes
26.22	0	26.22	73.78	0.12	No

H₂ conc. (%)	CH₄ conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
26.22	0	26.22	73.78	0.12	Yes
25.76	0	25.76	74.24	0.08	Yes
25.87	0	25.87	74.13	0.12	Yes
25.47 (average)	**0 (average)**	**25.47 (average)**	**74.53 (average)**	**0.03 (Lowest Energy)**	
29.86	0	29.86	70.14	0.03	Yes
29.11	0	29.11	70.89	0.03	Yes
29.37	0	29.37	70.63	0.06	Yes
29.54	0	29.54	70.46	0.06	Yes
29.21	0	29.21	70.79	0.06	No
29.21	0	29.21	70.79	0.02	Yes
29.26	0	29.26	70.74	0.04	No
29.26	0	29.26	70.74	0.05	Yes
29.45	0	29.45	70.55	0.05	Yes
29.70	0	29.70	70.30	0.04	No
29.70	0	29.70	70.30	0.03	Yes
30.07	0	30.07	69.93	0.06	Yes
30.05	0	30.05	69.95	0.07	Yes
30.10	0	30.10	69.90	0.06	Yes
31.07	0	31.07	68.93	0.06	Yes
31.03	0	31.03	68.97	0.16	No
31.03	0	31.03	68.97	0.10	Yes
31.96	0	31.96	68.04	0.07	No
31.96	0	31.96	68.04	0.11	Yes
30.62	0	30.62	69.38	0.24	Yes
31.38	0	31.38	68.62	0.08	Yes
30.87	0	30.87	69.13	0.05	No
30.87	0	30.87	69.13	0.07	Yes
31.50	0	31.50	68.50	0.19	Yes
31.34	0	31.34	68.66	0.08	Yes
31.31	0	31.31	68.69	0.07	Yes
30.38 (average)	**0 (average)**	**30.38 (average)**	**69.62 (average)**	**0.02 (Lowest Energy)**	
43.73	0	43.73	56.27	0.06	No
43.73	0	43.73	56.27	0.43	Yes
44.26	0	44.26	55.74	0.19	Yes
44.70	0	44.70	55.30	0.07	No
44.70	0	44.70	55.30	0.06	Yes
44.18	0	44.18	55.82	0.09	Yes
46.09	0	46.09	53.91	0.15	No
46.09	0	46.09	53.91	0.14	Yes
45.00	0	45.00	55.00	0.12	No

H$_2$ conc. (%)	CH$_4$ conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
45.00	0	45.00	55.00	0.15	Yes
44.99	0	44.99	55.01	0.22	Yes
45.73	0	45.73	54.27	0.19	No
45.73	0	45.73	54.27	0.21	Yes
46.34	0	46.34	53.66	0.17	No
46.34	0	46.34	53.66	0.23	Yes
46.49	0	46.49	53.51	0.26	No
46.49	0	46.49	53.51	0.29	Yes
47.00	0	47.00	53.00	0.32	No
47.00	0	47.00	53.00	0.28	Yes
47.36	0	47.36	52.64	0.12	No
47.36	0	47.36	52.64	0.08	Yes
45.41	0	45.41	54.59	0.56	Yes
43.32	0	43.32	56.68	0.35	Yes
45.09	0	45.09	54.91	0.31	Yes
46.51	0	46.51	53.49	0.19	No
46.51	0	46.51	53.49	0.22	Yes
46.78	0	46.78	53.22	0.32	Yes
46.68	0	46.68	53.32	0.19	No
46.68	0	46.68	53.32	0.26	Yes
47.68	0	47.68	52.32	0.21	Yes
47.48	0	47.48	52.52	0.19	No
47.48	0	47.48	52.52	0.37	Yes
45.68 (average)	0 (average)	45.68 (average)	54.32 (average)	0.06 (Lowest Energy)	
59.24	0	59.24	40.76	0.45	No
59.24	0	59.24	40.76	0.44	No
59.24	0	59.24	40.76	1.47	No
59.24	0	59.24	40.76	2.64	Yes
58.02	0	58.02	41.98	0.19	No
58.02	0	58.02	41.98	0.30	No
58.02	0	58.02	41.98	0.59	No
58.02	0	58.02	41.98	0.95	Yes
59.99	0	59.99	40.01	0.21	No
59.99	0	59.99	40.01	0.22	No
59.99	0	59.99	40.01	0.22	Yes
59.71	0	59.71	40.29	0.17	No
59.71	0	59.71	40.29	0.20	No
59.71	0	59.71	40.29	0.75	No
59.71	0	59.71	40.29	1.09	Yes
60.27	0	60.27	39.73	0.27	No
60.27	0	60.27	39.73	0.26	Yes
61.16	0	61.16	38.84	0.18	No
61.16	0	61.16	38.84	0.17	No
61.16	0	61.16	38.84	0.73	Yes
62.30	0	62.30	37.70	0.18	No
62.30	0	62.30	37.70	0.21	No
62.30	0	62.30	37.70	0.25	Yes

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H₂ conc. (%)	CH₄ conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
61.21	0	61.21	38.79	0.25	No
61.21	0	61.21	38.79	0.17	No
61.21	0	61.21	38.79	0.27	No
61.21	0	61.21	38.79	0.28	Yes
61.31	0	61.31	38.69	0.18	No
61.31	0	61.31	38.69	1.07	No
61.31	0	61.31	38.69	2.91	No
61.31	0	61.31	38.69	4.55	Yes
63.00	0	63.00	37.00	0.17	No
63.00	0	63.00	37.00	0.98	No
63.00	0	63.00	37.00	3.13	No
63.00	0	63.00	37.00	4.28	Yes
63.10	0	63.10	36.90	0.65	No
63.10	0	63.10	36.90	1.39	No
63.10	0	63.10	36.90	1.86	Yes
63.24	0	63.24	36.76	0.17	No
63.24	0	63.24	36.76	1.06	No
63.24	0	63.24	36.76	3.34	Yes
63.54	0	63.54	36.46	0.17	No
63.54	0	63.54	36.46	1.05	No
63.54	0	63.54	36.46	2.93	Yes
63.62	0	63.62	36.38	0.16	No
63.62	0	63.62	36.38	1.02	No
63.62	0	63.62	36.38	2.08	No
63.62	0	63.62	36.38	4.96	Yes
63.68	0	63.68	36.32	0.25	No
63.68	0	63.68	36.32	1.07	No
63.68	0	63.68	36.32	3.23	Yes
64.04	0	64.04	35.96	0.29	No
64.04	0	64.04	35.96	1.27	No
64.04	0	64.04	35.96	3.21	Yes
64.75	0	64.75	35.25	0.75	No
64.75	0	64.75	35.25	2.64	No
64.75	0	64.75	35.25	3.05	No
64.75	0	64.75	35.25	3.13	Yes
63.17	0	63.17	36.83	1.08	No
63.17	0	63.17	36.83	4.94	No
63.17	0	63.17	36.83	3.34	Yes
62.56	0	62.56	37.44	0.25	No
62.56	0	62.56	37.44	1.12	No
62.56	0	62.56	37.44	3.34	Yes
63.64	0	63.64	36.36	0.21	No
63.64	0	63.64	36.36	1.16	No
63.64	0	63.64	36.36	3.34	No
63.64	0	63.64	36.36	4.83	No
62.11 (average)	0 (average)	62.11 (average)	37.89 (average)	0.22 (Lowest Energy)	

Table H.3 : Experimental runs with 75 % methane + 25 % Hydrogen with air

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
6.18	1.99	8.17	91.83	0.19	No
6.18	1.99	8.17	91.83	1.27	Yes
6.30	2.29	8.60	91.40	0.31	No
6.30	2.29	8.60	91.40	4.35	Yes
6.34	2.31	8.65	91.35	0.70	Yes
6.05	1.79	7.84	92.16	0.25	No
6.05	1.79	7.84	92.16	0.98	No
6.05	1.79	7.84	92.16	3.23	No
6.05	1.79	7.84	92.16	9.63	No
5.99	2.02	8.00	92.00	0.30	No
5.99	2.02	8.00	92.00	3.10	Yes
6.28	1.77	8.05	91.95	0.76	No
6.28	1.77	8.05	91.95	3.87	Yes
6.21	2.00	8.22	91.78	0.25	No
6.21	2.00	8.22	91.78	1.20	Yes
6.21	1.77	7.98	92.02	0.19	No
6.21	1.77	7.98	92.02	1.39	Yes
5.81	2.25	8.06	91.94	0.58	Yes
6.12	1.79	7.92	92.08	0.61	Yes
5.91	2.07	7.98	92.02	0.58	Yes
5.61	2.06	7.67	92.33	0.62	Yes
6.02	2.25	8.27	91.73	0.62	Yes
5.72	1.75	7.47	92.53	0.36	No
5.72	1.75	7.47	92.53	1.56	No
5.72	1.75	7.47	92.53	5.05	Yes
6.21	2.35	8.56	91.44	0.61	Yes
5.81	1.85	7.65	92.35	0.65	Yes
5.95	1.75	7.70	92.30	0.42	No
5.95	1.75	7.70	92.30	2.32	Yes
5.97	1.89	7.86	92.14	0.73	Yes
5.56	2.04	7.60	92.40	0.24	No
5.56	2.04	7.60	92.40	1.27	Yes
5.90	2.09	7.99	92.01	0.53	No
5.90	2.09	7.99	92.01	1.02	Yes
6.01 (average)	2.01 (average)	8.02 (average)	91.98 (average)	0.27 (Lowest Energy)	
8.44	2.61	11.05	88.95	0.11	Yes
8.35	2.20	10.55	89.45	0.13	Yes
7.91	2.17	10.07	89.93	0.10	Yes
8.23	2.74	10.97	89.03	0.11	Yes

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
7.81	2.39	10.20	89.80	0.32	No
7.81	2.39	10.20	89.80	0.49	Yes
8.23	2.96	11.19	88.81	0.12	Yes
7.60	2.98	10.58	89.42	0.11	Yes
8.46	2.58	11.04	88.96	0.10	Yes
7.97	2.59	10.56	89.44	0.16	Yes
8.18	2.61	10.79	89.21	0.11	Yes
7.54	2.87	10.41	89.59	0.12	Yes
8.09	2.25	10.35	89.65	0.28	No
8.09	2.25	10.35	89.65	0.21	No
8.09	2.25	10.35	89.65	1.78	Yes
7.58	2.39	9.97	90.03	0.10	Yes
8.27	2.83	11.10	88.90	0.15	Yes
8.05	2.44	10.49	89.51	0.11	Yes
8.08	2.81	10.89	89.11	0.21	Yes
8.33	2.91	11.24	88.76	0.39	No
8.33	2.91	11.24	88.76	0.42	No
8.33	2.91	11.24	88.76	2.32	Yes
8.05	2.75	10.81	89.19	0.33	No
8.05	2.75	10.81	89.19	0.27	No
8.05	2.75	10.81	89.19	1.24	Yes
7.39	2.68	10.07	89.93	1.06	Yes
8.00	2.98	10.97	89.03	0.16	Yes
8.03 (average)	2.64 (average)	10.67 (average)	89.33 (average)	0.10 (Lowest Energy)	
9.70	3.17	12.87	87.13	0.31	No
9.70	3.17	12.87	87.13	0.32	No
9.70	3.17	12.87	87.13	0.52	Yes
9.77	3.22	12.98	87.02	0.25	No
9.77	3.22	12.98	87.02	0.40	No
9.77	3.22	12.98	87.02	2.32	Yes
10.26	3.24	13.50	86.50	0.42	No
10.26	3.24	13.50	86.50	1.69	No
10.26	3.24	13.50	86.50	4.34	Yes
9.79	3.38	13.17	86.83	0.42	No
9.79	3.38	13.17	86.83	0.49	No
9.79	3.38	13.17	86.83	1.27	Yes
10.50	3.10	13.60	86.40	0.52	No
10.50	3.10	13.60	86.40	3.94	Yes
9.66	3.18	12.84	87.16	0.39	No
9.66	3.18	12.84	87.16	3.22	No
9.66	3.18	12.84	87.16	7.83	Yes
10.32	3.15	13.47	86.53	0.42	No
10.32	3.15	13.47	86.53	2.27	Yes
10.54	3.87	14.42	85.58	0.36	No
10.54	3.87	14.42	85.58	1.35	Yes

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
10.02	3.46	13.48	86.52	0.20	No
10.02	3.46	13.48	86.52	1.16	No
10.02	3.46	13.48	86.52	2.93	Yes
10.08	3.53	13.61	86.39	0.36	No
10.08	3.53	13.61	86.39	1.65	No
10.08	3.53	13.61	86.39	3.88	Yes
9.71	3.20	12.91	87.09	0.26	No
9.71	3.20	12.91	87.09	1.56	No
9.71	3.20	12.91	87.09	3.99	Yes
10.47	3.07	13.55	86.45	0.65	No
10.47	3.07	13.55	86.45	1.97	No
10.47	3.07	13.55	86.45	2.20	Yes
10.03	3.67	13.70	86.30	0.25	No
10.03	3.67	13.70	86.30	2.75	No
10.03	3.67	13.70	86.30	5.61	Yes
10.17	3.04	13.21	86.79	0.88	No
10.17	3.04	13.21	86.79	4.57	No
10.17	3.04	13.21	86.79	5.48	Yes
10.57	3.68	14.25	85.75	0.43	No
10.57	3.68	14.25	85.75	2.02	No
10.57	3.68	14.25	85.75	3.55	No
10.57	3.68	14.25	85.75	9.63	Yes
9.93	3.73	13.66	86.34	0.73	No
9.93	3.73	13.66	86.34	4.35	No
9.93	3.73	13.66	86.34	5.89	Yes
10.35	3.55	13.90	86.10	0.43	No
10.35	3.55	13.90	86.10	3.67	No
10.35	3.55	13.90	86.10	5.75	No
10.35	3.55	13.90	86.10	10.04	No
9.67	3.65	13.32	86.68	0.55	No
9.67	3.65	13.32	86.68	3.60	No
9.67	3.65	13.32	86.68	4.94	Yes
9.78	3.78	13.55	86.45	0.44	No
9.78	3.78	13.55	86.45	3.87	No
9.78	3.78	13.55	86.45	8.99	Yes
10.59	3.19	13.78	86.22	0.61	No
10.59	3.19	13.78	86.22	2.54	No
10.59	3.19	13.78	86.22	5.35	No
10.59	3.19	13.78	86.22	7.55	No
10.1 (average)	3.39 (average)	13.49 (average)	86.51 (average)	0.98 (Lowest Energy)	
11.90	4.32	16.22	83.78	0.58	No
11.90	4.32	16.22	83.78	2.16	No
11.90	4.32	16.22	83.78	4.71	No
11.90	4.32	16.22	83.78	10.41	Yes
12.70	4.21	16.91	83.09	0.39	No
12.70	4.21	16.91	83.09	2.06	No
12.70	4.21	16.91	83.09	4.98	No
12.70	4.21	16.91	83.09	6.94	Yes
12.33	4.19	16.52	83.48	0.99	No
12.33	4.19	16.52	83.48	4.94	No
12.33	4.19	16.52	83.48	8.15	No
12.33	4.19	16.52	83.48	10.04	No
12.89	3.99	16.88	83.12	0.56	No
12.89	3.99	16.88	83.12	2.64	No

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
12.89	3.99	16.88	83.12	7.05	No
12.89	3.99	16.88	83.12	9.23	Yes
12.16	4.65	16.82	83.18	0.34	No
12.16	4.65	16.82	83.18	2.31	No
12.16	4.65	16.82	83.18	5.19	Yes
12.69	4.05	16.74	83.26	0.46	No
12.69	4.05	16.74	83.26	3.29	No
12.69	4.05	16.74	83.26	4.71	No
12.69	4.05	16.74	83.26	7.55	No
12.27	4.67	16.94	83.06	0.72	No
12.27	4.67	16.94	83.06	3.80	No
12.27	4.67	16.94	83.06	7.83	No
12.27	4.67	16.94	83.06	6.65	No
12.88	4.20	17.09	82.91	0.53	No
12.88	4.20	17.09	82.91	2.81	No
12.88	4.20	17.09	82.91	6.18	No
12.88	4.20	17.09	82.91	11.74	Yes
12.37	4.30	16.67	83.33	0.60	No
12.37	4.30	16.67	83.33	1.78	No
12.37	4.30	16.67	83.33	13.58	No
12.37	4.30	16.67	83.33	9.79	No
12.32	4.41	16.72	83.28	0.75	No
12.32	4.41	16.72	83.28	2.98	No
12.32	4.41	16.72	83.28	7.83	No
12.32	4.41	16.72	83.28	11.74	No
12.41	4.60	17.01	82.99	0.48	No
12.41	4.60	17.01	82.99	4.33	No
12.41	4.60	17.01	82.99	9.43	Yes
12.76	3.96	16.73	83.27	0.41	No
12.76	3.96	16.73	83.27	2.32	No
12.76	3.96	16.73	83.27	5.89	No
12.76	3.96	16.73	83.27	6.36	No
12.60	4.12	16.71	83.29	0.44	No
12.60	4.12	16.71	83.29	2.81	No
12.60	4.12	16.71	83.29	6.17	No
12.60	4.12	16.71	83.29	10.04	No
12.47	4.72	17.19	82.81	0.78	No
12.47	4.72	17.19	82.81	4.50	No
12.47	4.72	17.19	82.81	9.16	No
12.47	4.72	17.19	82.81	8.45	No
12.30	4.12	16.42	83.58	0.34	No
12.30	4.12	16.42	83.58	2.37	No
12.30	4.12	16.42	83.58	5.75	No
12.30	4.12	16.42	83.58	6.04	No
12.07	3.96	16.03	83.97	0.20	No
12.07	3.96	16.03	83.97	2.32	No
12.07	3.96	16.03	83.97	5.89	No
12.07	3.96	16.03	83.97	6.36	No
12.00	4.34	16.33	83.67	0.42	No
12.00	4.34	16.33	83.67	4.13	No
12.00	4.34	16.33	83.67	10.87	No
12.28	3.96	16.23	83.77	1.33	No
12.28	3.96	16.23	83.77	4.43	No
12.28	3.96	16.23	83.77	8.82	No
12.28	3.96	16.23	83.77	10.49	No

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CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
12.01	4.11	16.11	83.89	0.33	No
12.01	4.11	16.11	83.89	1.78	No
12.01	4.11	16.11	83.89	5.89	No
12.01	4.11	16.11	83.89	4.62	No
13.02	4.66	17.67	82.33	0.73	No
13.02	4.66	17.67	82.33	3.35	No
13.02	4.66	17.67	82.33	6.17	No
13.02	4.66	17.67	82.33	9.23	No
12.42 (average)	4.28 (average)	16.70 (average)	83.30 (average)	5.19 (Lowest Energy)	

Table H.4 : Experimental runs with 50% methane +50% Hydrogen with air

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
2.80	2.83	5.64	94.36	0.40	No
2.80	2.83	5.64	94.36	4.43	No
2.80	2.83	5.64	94.36	11.74	Yes
3.06	3.17	6.23	93.77	0.44	No
3.06	3.17	6.23	93.77	4.18	No
3.06	3.17	6.23	93.77	12.42	Yes
3.00	3.48	6.48	93.52	0.36	No
3.00	3.48	6.48	93.52	3.67	No
3.00	3.48	6.48	93.52	9.23	Yes
2.81	2.83	5.64	94.36	0.42	No
2.81	2.83	5.64	94.36	2.47	No
2.81	2.83	5.64	94.36	10.41	Yes
3.15	3.44	6.59	93.41	0.94	No
3.15	3.44	6.59	93.41	5.95	No
3.15	3.44	6.59	93.41	8.48	No
3.15	3.44	6.59	93.41	8.54	No
2.71	3.20	5.91	94.09	0.08	No
2.71	3.20	5.91	94.09	2.54	No
2.71	3.20	5.91	94.09	5.09	Yes
2.71	3.20	5.91	94.09	0.00	No
3.40	3.21	6.61	93.39	0.87	No
3.40	3.21	6.61	93.39	3.53	No
3.40	3.21	6.61	93.39	7.83	No
3.40	3.21	6.61	93.39	11.26	No
2.81	3.24	6.05	93.95	0.20	No
2.81	3.24	6.05	93.95	0.82	No
2.81	3.24	6.05	93.95	2.72	Yes
3.37	3.38	6.75	93.25	0.72	No
3.37	3.38	6.75	93.25	6.92	No
3.37	3.38	6.75	93.25	6.57	No
3.37	3.38	6.75	93.25	12.25	No
3.30	2.79	6.09	93.91	0.87	No
3.30	2.79	6.09	93.91	3.80	No
3.30	2.79	6.09	93.91	8.31	No
3.30	2.79	6.09	93.91	10.58	No
2.76	3.11	5.87	94.13	0.72	No
2.76	3.11	5.87	94.13	5.72	No
2.76	3.11	5.87	94.13	7.51	No
2.76	3.11	5.87	94.13	11.40	No
2.76	3.40	6.17	93.83	0.42	No
2.76	3.40	6.17	93.83	1.65	No
2.76	3.40	6.17	93.83	13.34	Yes
3.32	3.31	6.63	93.37	0.50	No
3.32	3.31	6.63	93.37	1.83	No
3.32	3.31	6.63	93.37	8.90	Yes
2.68	2.80	5.48	94.52	0.99	No
2.68	2.80	5.48	94.52	4.07	No
2.68	2.80	5.48	94.52	8.15	No
2.68	2.80	5.48	94.52	8.54	No

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
3.18	3.16	6.35	93.65	0.45	No
3.18	3.16	6.35	93.65	1.60	No
3.18	3.16	6.35	93.65	12.64	No
3.18	3.16	6.35	93.65	9.41	No
3.07	3.39	6.46	93.54	0.33	No
3.07	3.39	6.46	93.54	2.71	No
3.07	3.39	6.46	93.54	9.43	Yes
3.30	2.70	5.99	94.01	0.34	No
3.30	2.70	5.99	94.01	3.30	No
3.30	2.70	5.99	94.01	8.46	Yes
3.30	3.29	6.59	93.41	0.50	No
3.30	3.29	6.59	93.41	1.65	No
3.30	3.29	6.59	93.41	11.52	No
3.30	3.29	6.59	93.41	13.13	No
2.83	3.02	5.85	94.15	0.41	No
2.83	3.02	5.85	94.15	2.53	No
2.83	3.02	5.85	94.15	10.87	Yes
2.64	2.82	5.46	94.54	0.28	No
2.64	2.82	5.46	94.54	1.83	No
2.64	2.82	5.46	94.54	6.79	Yes
3.01 (average)	3.13 (average)	6.14 (average)	93.86 (average)	2.72 (Lowest Energy)	
5.38	4.85	10.23	89.77	0.22	No
5.38	4.85	10.23	89.77	1.69	No
5.38	4.85	10.23	89.77	2.88	Yes
4.90	4.68	9.58	90:42	0.31	Yes
5.17	5.24	10.41	89.59	0.25	Yes
4.76	5.05	9.82	90.18	0.18	Yes
4.94	5.34	10.28	89.72	0.55	No
4.94	5.34	10.28	89.72	1.92	No
4.94	5.34	10.28	89.72	2.34	Yes
5.29	5.38	10.67	89.33	0.19	Yes
5.10	5.36	10.46	89.54	0.17	Yes
4.62	5.10	9.72	90.28	0.43	Yes
4.22	5.31	9.53	90.47	0.49	Yes
4.89	5.07	9.96	90.04	0.70	No
4.89	5.07	9.96	90.04	3.16	Yes
4.72	5.13	9.85	90.15	0.35	No
4.72	5.13	9.85	90.15	1.27	Yes
4.80	5.19	9.99	90.01	0.22	Yes
4.73	4.73	9.46	90.54	0.34	Yes
5.31	5.11	10.41	89.59	1.01	Yes
4.87	4.79	9.67	90.33	1.01	No
4.87	4.79	9.67	90.33	1.39	Yes
4.89	5.09	9.98	90.02	0.17	Yes
5.36	5.24	10.60	89.40	0.16	Yes

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
5.54	4.51	10.05	89.95	0.72	No
5.54	4.51	10.05	89.95	2.81	Yes
5.19	4.71	9.90	90.10	0.24	Yes
5.12	4.84	9.97	90.03	0.17	Yes
4.99 (average)	5.04 (average)	10.03 (average)	89.97 (average)	0.16 (Lowest Energy)	
6.68	6.92	13.60	86.40	0.09	Yes
7.00	6.94	13.93	86.07	0.12	Yes
6.83	6.47	13.30	86.70	0.09	Yes
7.07	6.63	13.70	86.30	0.12	Yes
6.70	6.88	13.58	86.42	0.41	No
6.70	6.88	13.58	86.42	1.04	Yes
7.31	6.94	14.25	85.75	0.10	Yes
6.74	6.85	13.58	86.42	0.17	Yes
6.66	6.67	13.33	86.67	0.11	Yes
6.39	6.62	13.01	86.99	0.10	Yes
7.03	6.77	13.79	86.21	0.39	No
7.03	6.77	13.79	86.21	0.32	Yes
6.49	6.35	12.84	87.16	0.11	Yes
6.79	6.36	13.15	86.85	0.12	Yes
6.68	6.45	13.13	86.87	0.11	Yes
6.84	6.68	13.52	86.48	0.27	No
6.84	6.68	13.52	86.48	0.28	Yes
6.72	6.63	13.35	86.65	0.12	Yes
6.62	6.57	13.19	86.81	0.10	Yes
6.65	6.49	13.15	86.85	0.15	Yes
7.09	6.84	13.93	86.07	0.10	Yes
6.79	6.70	13.49	86.51	0.40	No
6.79	6.70	13.49	86.51	0.55	No
6.79	6.70	13.49	86.51	2.16	Yes
6.84	6.85	13.69	86.31	0.35	No
6.84	6.85	13.69	86.31	0.66	No
6.84	6.85	13.69	86.31	2.31	Yes
6.80 (average)	6.68 (average)	13.48 (average)	86.52 (average)	0.09 (Lowest Energy)	
7.88	8.21	16.09	83.91	0.14	No
7.88	8.21	16.09	83.91	0.31	No
7.88	8.21	16.09	83.91	2.36	Yes
8.12	8.01	16.12	83.88	0.13	Yes
8.17	8.24	16.41	83.59	0.10	Yes
8.06	8.17	16.22	83.78	0.13	Yes
7.84	8.30	16.14	83.86	0.41	No
7.84	8.30	16.14	83.86	1.04	Yes

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
8.24	7.95	16.19	83.81	0.11	Yes
8.41	7.74	16.15	83.85	0.10	Yes
8.53	8.37	16.91	83.09	0.12	Yes
7.88	7.92	15.80	84.20	0.11	Yes
7.73	7.84	15.57	84.43	0.42	No
7.73	7.84	15.57	84.43	0.31	Yes
8.37	7.88	16.25	83.75	0.11	Yes
8.07	8.04	16.12	83.88	0.11	Yes
8.06	8.24	16.30	83.70	0.11	Yes
8.19	8.08	16.27	83.73	0.25	No
8.19	8.08	16.27	83.73	0.26	Yes
8.29	8.16	16.44	83.56	0.13	Yes
7.83	8.35	16.18	83.82	0.10	Yes
8.39	8.12	16.51	83.49	0.14	Yes
8.02	8.14	16.16	83.84	0.10	Yes
7.82	7.70	15.52	84.48	0.22	No
7.82	7.70	15.52	84.48	0.62	No
7.82	7.70	15.52	84.48	2.98	Yes
7.83	7.93	15.76	84.24	0.36	No
7.83	7.93	15.76	84.24	0.62	No
7.83	7.93	15.76	84.24	2.81	Yes
8.09 (average)	8.07 (average)	16.16 (average)	83.84 (average)	0.10 (Lowest Energy)	
10.36	9.63	19.99	80.01	0.27	No
10.36	9.63	19.99	80.01	4.53	No
10.36	9.63	19.99	80.01	3.77	No
10.36	9.63	19.99	80.01	7.73	No
10.81	10.10	20.91	79.09	1.06	No
10.81	10.10	20.91	79.09	3.87	No
10.81	10.10	20.91	79.09	4.11	Yes
9.49	10.01	19.50	80.50	2.04	No
9.49	10.01	19.50	80.50	2.37	No
9.49	10.01	19.50	80.50	4.83	No
9.49	10.01	19.50	80.50	11.82	No
10.26	10.53	20.79	79.21	0.99	No
10.26	10.53	20.79	79.21	0.41	No
10.26	10.53	20.79	79.21	1.24	No
10.26	10.53	20.79	79.21	6.65	No
10.40	10.04	20.44	79.56	0.72	No
10.40	10.04	20.44	79.56	6.18	No
10.40	10.04	20.44	79.56	6.70	No
10.40	10.04	20.44	79.56	9.53	No
9.51	10.39	19.89	80.11	0.36	No
9.51	10.39	19.89	80.11	2.42	No
9.51	10.39	19.89	80.11	4.73	No
9.51	10.39	19.89	80.11	5.12	No
10.11	9.98	20.09	79.91	0.88	No
10.11	9.98	20.09	79.91	1.87	No
10.11	9.98	20.09	79.91	4.23	No

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)		Energy (mJ)	Success of Ignition
10.11	9.98	20.09	79.91		7.16	No
9.71	9.67	19.38	80.62		0.32	No
9.71	9.67	19.38	80.62		2.82	No
9.71	9.67	19.38	80.62		5.28	No
9.71	9.67	19.38	80.62		11.62	No
9.96	10.05	20.01	79.99		0.34	No
9.96	10.05	20.01	79.99		3.30	No
9.96	10.05	20.01	79.99		8.65	Yes
10.02	10.57	20.60	79.40		0.19	No
10.02	10.57	20.60	79.40		6.18	No
10.02	10.57	20.60	79.40		3.05	No
10.02	10.57	20.60	79.40		6.54	No
10.10	10.26	20.36	79.64		0.69	No
10.10	10.26	20.36	79.64		2.01	No
10.10	10.26	20.36	79.64		6.87	No
10.10	10.26	20.36	79.64		7.16	Yes
10.28	9.66	19.94	80.06		0.32	No
10.28	9.66	19.94	80.06		1.43	No
10.28	9.66	19.94	80.06		6.52	No
10.28	9.66	19.94	80.06		11.96	No
10.48	10.04	20.51	79.49		0.41	No
10.48	10.04	20.51	79.49		1.92	No
10.48	10.04	20.51	79.49		3.77	No
10.48	10.04	20.51	79.49		5.42	No
9.46	9.88	19.34	80.66		0.29	No
9.46	9.88	19.34	80.66		1.87	No
9.46	9.88	19.34	80.66		4.58	No
9.46	9.88	19.34	80.66		10.05	No
9.94	9.73	19.67	80.33		0.99	No
9.94	9.73	19.67	80.33		3.80	No
9.94	9.73	19.67	80.33		9.88	No
9.94	9.73	19.67	80.33		11.40	Yes
9.45	10.15	19.60	80.40		0.45	No
9.45	10.15	19.60	80.40		1.60	No
9.45	10.15	19.60	80.40		12.64	No
9.45	10.15	19.60	80.40		9.41	No
10.04	9.53	19.57	80.43		0.44	No
10.04	9.53	19.57	80.43		3.75	No
10.04	9.53	19.57	80.43		8.09	No
10.43	9.66	20.09	79.91		0.34	No
10.43	9.66	20.09	79.91		2.72	No
10.43	9.66	20.09	79.91		11.30	Yes
10.38	9.59	19.97	80.03		0.15	No
10.38	9.59	19.97	80.03		1.92	No
10.38	9.59	19.97	80.03		3.61	No
10.38	9.59	19.97	80.03		8.54	No
10.76	10.57	21.32	78.68		0.76	No
10.76	10.57	21.32	78.68		3.35	No
10.76	10.57	21.32	78.68		2.74	No
10.76	10.57	21.32	78.68		10.66	No
12.00	12.10	24.10	75.90		0.99	No
12.00	12.10	24.10	75.90		2.52	No
12.00	12.10	24.10	75.90		3.57	No
12.00	12.10	24.10	75.90		6.04	No
12.00	12.10	24.10	75.90		7.56	No

CH4 conc. (%)	H2 conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
15.07	16.42	31.49	68.51	0.34	No
15.07	16.42	31.49	68.51	2.27	No
15.07	16.42	31.49	68.51	3.41	No
15.07	16.42	31.49	68.51	8.81	No
18.95	19.63	38.59	61.41	0.51	No
18.95	19.63	38.59	61.41	2.11	No
18.95	19.63	38.59	61.41	6.36	No
18.95	19.63	38.59	61.41	6.52	No
10.10 (average)	10.00 (average)	20.10 (average)	79.90 (average)	4.11 (Lowest Energy)	

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Table H.5 : Experimental runs with 25% methane + 75% Hydrogen with air

H₂ conc. (%)	CH₄ conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
5.97	2.40	8.37	91.63	0.72	No
5.97	2.40	8.37	91.63	6.92	No
5.97	2.40	8.37	91.63	9.68	No
5.97	2.40	8.37	91.63	8.76	No
6.01	2.63	8.64	91.36	0.56	No
6.01	2.63	8.64	91.36	6.30	No
6.01	2.63	8.64	91.36	9.16	No
6.01	2.63	8.64	91.36	10.31	No
6.07	2.29	8.36	91.64	0.94	No
6.07	2.29	8.36	91.64	4.14	Yes
6.46	1.96	8.42	91.58	0.36	No
6.46	1.96	8.42	91.58	2.98	No
6.46	1.96	8.42	91.58	5.75	No
6.46	1.96	8.42	91.58	9.53	Yes
6.22	2.67	8.89	91.11	0.25	No
6.22	2.67	8.89	91.11	2.75	No
6.22	2.67	8.89	91.11	5.75	No
6.22	2.67	8.89	91.11	7.38	Yes
6.17	2.29	8.45	91.55	0.61	No
6.17	2.29	8.45	91.55	5.38	No
6.17	2.29	8.45	91.55	8.65	Yes
5.87	1.92	7.79	92.21	0.65	No
5.87	1.92	7.79	92.21	2.06	No
5.87	1.92	7.79	92.21	9.68	Yes
6.09	2.20	8.30	91.70	0.87	No
6.09	2.20	8.30	91.70	3.80	No
6.09	2.20	8.30	91.70	12.73	No
6.09	2.20	8.30	91.70	9.28	No
5.75	2.00	7.75	92.25	0.42	No
5.75	2.00	7.75	92.25	2.47	No
5.75	2.00	7.75	92.25	9.06	Yes
6.31	1.88	8.20	91.80	0.73	No
6.31	1.88	8.20	91.80	5.95	No
6.31	1.88	8.20	91.80	12.73	No
6.31	1.88	8.20	91.80	10.58	No
6.22	2.39	8.61	91.39	0.42	No
6.22	2.39	8.61	91.39	1.65	No
6.22	2.39	8.61	91.39	10.41	Yes
6.45	2.57	9.02	90.98	0.40	No
6.45	2.57	9.02	90.98	3.48	No
6.45	2.57	9.02	90.98	11.74	Yes
6.42	2.40	8.81	91.19	0.76	No
6.42	2.40	8.81	91.19	3.48	No
6.42	2.40	8.81	91.19	10.97	No
6.42	2.40	8.81	91.19	11.96	No
6.26	2.16	8.41	91.59	0.20	No
6.26	2.16	8.41	91.59	0.82	No
6.26	2.16	8.41	91.59	2.72	No
6.26	2.16	8.41	91.59	6.44	Yes

H₂ conc. (%)	CH₄ conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
6.23	2.27	8.50	91.50	0.87	No
6.23	2.27	8.50	91.50	3.53	No
6.23	2.27	8.50	91.50	12.73	No
6.23	2.27	8.50	91.50	13.43	No
6.04	2.18	8.23	91.77	0.44	No
6.04	2.18	8.23	91.77	4.07	No
6.04	2.18	8.23	91.77	12.42	Yes
5.82	2.32	8.14	91.86	0.36	No
5.82	2.32	8.14	91.86	3.67	No
5.82	2.32	8.14	91.86	12.19	No
5.90	2.51	8.41	91.59	0.94	No
5.90	2.51	8.41	91.59	6.42	No
5.90	2.51	8.41	91.59	13.56	No
5.90	2.51	8.41	91.59	11.37	Yes
6.30	2.23	8.52	91.48	0.08	No
6.30	2.23	8.52	91.48	2.06	Yes
6.61	2.16	8.77	91.23	0.42	No
6.61	2.16	8.77	91.23	3.84	No
6.61	2.16	8.77	91.23	4.71	Yes
6.16 (average)	2.27 (average)	8.43 (average)	91.57 (average)	2.06 (Lowest Energy)	
10.24	3.09	13.32	86.68	0.12	Yes
10.13	3.32	13.45	86.55	0.16	Yes
9.86	3.59	13.44	86.56	0.10	Yes
9.93	3.41	13.34	86.66	0.11	Yes
10.18	3.84	14.02	85.98	0.32	Yes
9.77	3.31	13.08	86.92	0.12	Yes
9.98	3.60	13.58	86.42	0.11	Yes
10.14	3.62	13.77	86.23	0.10	No
10.14	3.62	13.77	86.23	1.09	Yes
10.07	3.03	13.09	86.91	0.16	Yes
10.02	3.54	13.56	86.44	0.11	Yes
10.15	3.80	13.94	86.06	0.39	Yes
10.57	3.60	14.16	85.84	0.50	No
10.57	3.60	14.16	85.84	1.13	No
10.57	3.60	14.16	85.84	2.48	Yes
9.78	3.71	13.49	86.51	0.32	No
9.78	3.71	13.49	86.51	1.73	Yes
9.72	3.62	13.34	86.66	0.12	Yes
10.20	3.87	14.07	85.93	0.16	Yes
10.38	3.29	13.67	86.33	0.42	No
10.38	3.29	13.67	86.33	0.56	No
10.38	3.29	13.67	86.33	2.58	Yes
9.83	3.03	12.87	87.13	0.11	Yes
10.89	3.28	14.18	85.82	0.76	No
10.89	3.28	14.18	85.82	0.88	Yes

H₂ conc. (%)	CH₄ conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
10.41	3.49	13.91	86.09	0.12	Yes
10.50	3.90	14.40	85.60	0.48	No
10.50	3.90	14.40	85.60	0.75	Yes
10.14 (average)	3.50 (average)	13.64 (average)	86.36 (average)	0.10 (Lowest Energy)	
13.61	5.03	18.64	81.36	0.11	Yes
13.40	4.73	18.13	81.87	0.10	Yes
13.60	4.52	18.12	81.88	0.08	Yes
13.27	4.71	17.98	82.02	0.08	Yes
12.87	4.37	17.23	82.77	0.18	Yes
12.91	4.33	17.24	82.76	0.11	Yes
13.44	4.22	17.66	82.34	0.10	Yes
13.53	4.60	18.13	81.87	0.08	Yes
13.36	4.64	18.00	82.00	0.08	Yes
12.82	4.29	17.11	82.89	0.11	Yes
13.33	4.76	18.08	81.92	0.17	Yes
13.14	4.30	17.45	82.55	0.51	No
13.14	4.30	17.45	82.55	0.56	No
13.14	4.30	17.45	82.55	1.73	Yes
13.47	4.65	18.12	81.88	0.19	Yes
13.25	4.50	17.76	82.24	0.12	Yes
13.42	4.45	17.88	82.12	0.09	Yes
12.80	4.27	17.07	82.93	0.19	No
12.80	4.27	17.07	82.93	0.24	No
12.80	4.27	17.07	82.93	0.67	Yes
13.69	4.74	18.43	81.57	0.07	Yes
13.51	5.08	18.59	81.41	0.52	No
13.51	5.08	18.59	81.41	0.76	Yes
13.67	4.67	18.34	81.66	0.11	Yes
13.75	4.29	18.04	81.96	0.45	No
13.75	4.29	18.04	81.96	0.46	No
13.75	4.29	18.04	81.96	1.47	Yes
13.34 (average)	4.56 (average)	17.90 (average)	82.10 (average)	0.07 (Lowest Energy)	
16.00	5.84	21.85	78.15	0.18	Yes
16.33	5.90	22.23	77.77	0.17	Yes
15.48	5.78	21.26	78.74	0.14	Yes
15.32	5.08	20.40	79.60	0.42	No
15.32	5.08	20.40	79.60	0.46	No
15.32	5.08	20.40	79.60	2.64	Yes
16.10	5.75	21.86	78.14	0.35	Yes
15.62	5.53	21.15	78.85	0.14	Yes

H₂ conc. (%)	CH₄ conc. (%)	Total flammables conc. (%)	Air conc. (%)		Energy (mJ)	Success of Ignition
16.19	5.03	21.22	78.78		0.12	Yes
15.58	5.72	21.30	78.70		0.12	Yes
16.24	5.59	21.83	78.17		0.17	Yes
15.44	5.28	20.72	79.28		0.11	Yes
15.81	6.04	21.86	78.14		0.19	Yes
16.22	5.02	21.24	78.76		0.49	No
16.22	5.02	21.24	78.76		0.61	No
16.22	5.02	21.24	78.76		2.42	Yes
15.58	5.88	21.46	78.54		0.31	No
15.58	5.88	21.46	78.54		0.40	No
15.58	5.88	21.46	78.54		2.06	Yes
15.87	5.82	21.69	78.31		0.14	Yes
15.73	5.65	21.38	78.62		0.17	Yes
15.48	5.19	20.67	79.33		0.21	Yes
15.60	5.50	21.09	78.91		0.12	Yes
16.14	5.53	21.67	78.33		0.70	No
16.14	5.53	21.67	78.33		0.58	Yes
15.53	5.86	21.40	78.60		0.38	No
15.53	5.86	21.40	78.60		0.33	No
15.53	5.86	21.40	78.60		0.73	Yes
16.42	5.86	22.28	77.72		0.11	Yes
15.84 (average)	5.59 (average)	21.43 (average)	78.57 (average)		0.11 (Lowest Energy)	
20.64	6.51	27.15	72.85		0.22	No
20.64	6.51	27.15	72.85		0.31	No
20.64	6.51	27.15	72.85		1.24	Yes
20.37	7.23	27.59	72.41		0.17	No
20.37	7.23	27.59	72.41		0.42	No
20.37	7.23	27.59	72.41		1.39	No
20.37	7.23	27.59	72.41		5.89	Yes
20.61	7.15	27.76	72.24		0.22	No
20.61	7.15	27.76	72.24		2.37	No
20.61	7.15	27.76	72.24		5.48	No
20.61	7.15	27.76	72.24		6.20	Yes
19.62	6.59	26.21	73.79		0.28	No
19.62	6.59	26.21	73.79		2.37	No
19.62	6.59	26.21	73.79		5.89	No
19.62	6.59	26.21	73.79		7.20	Yes
20.61	7.02	27.63	72.37		0.36	No
20.61	7.02	27.63	72.37		0.42	No
20.61	7.02	27.63	72.37		1.39	No
20.61	7.02	27.63	72.37		3.52	Yes
20.19	6.41	26.60	73.40		0.33	No
20.19	6.41	26.60	73.40		0.42	No
20.19	6.41	26.60	73.40		1.12	No
20.19	6.41	26.60	73.40		8.93	Yes
20.00	6.96	26.96	73.04		0.42	No
20.00	6.96	26.96	73.04		0.42	No
20.00	6.96	26.96	73.04		1.39	No
20.00	6.96	26.96	73.04		5.60	Yes

H₂ conc. (%)	CH₄ conc. (%)	Total flammables conc. (%)	Air conc. (%)	Energy (mJ)	Success of Ignition
20.31	7.10	27.41	72.59	0.30	No
20.31	7.10	27.41	72.59	0.42	No
20.31	7.10	27.41	72.59	1.39	Yes
20.29	7.33	27.63	72.37	0.31	No
20.29	7.33	27.63	72.37	0.42	No
20.29	7.33	27.63	72.37	1.39	No
20.29	7.33	27.63	72.37	11.74	Yes
20.66	6.89	27.55	72.45	0.41	No
20.66	6.89	27.55	72.45	2.81	No
20.66	6.89	27.55	72.45	6.42	No
20.66	6.89	27.55	72.45	10.04	No
21.12	6.64	27.76	72.24	0.30	No
21.12	6.64	27.76	72.24	2.81	No
21.12	6.64	27.76	72.24	6.67	No
21.12	6.64	27.76	72.24	7.55	No
20.20	6.79	27.00	73.00	0.30	No
20.20	6.79	27.00	73.00	1.05	No
20.20	6.79	27.00	73.00	8.79	No
20.20	6.79	27.00	73.00	10.04	Yes
19.58	6.43	26.01	73.99	0.32	No
19.58	6.43	26.01	73.99	0.33	No
19.58	6.43	26.01	73.99	1.27	No
19.58	6.43	26.01	73.99	6.46	Yes
20.18	6.74	26.92	73.08	0.24	No
20.18	6.74	26.92	73.08	2.54	No
20.18	6.74	26.92	73.08	3.34	No
20.18	6.74	26.92	73.08	4.69	No
20.20	6.65	26.85	73.15	0.23	No
20.20	6.65	26.85	73.15	0.42	No
20.20	6.65	26.85	73.15	1.39	No
20.20	6.65	26.85	73.15	11.16	Yes
20.45	7.27	27.72	72.28	0.22	No
20.45	7.27	27.72	72.28	0.42	No
20.45	7.27	27.72	72.28	1.39	No
20.45	7.27	27.72	72.28	3.34	Yes
20.62	6.57	27.19	72.81	0.31	No
20.62	6.57	27.19	72.81	2.54	No
20.62	6.57	27.19	72.81	3.34	No
20.62	6.57	27.19	72.81	11.09	No
20.42	6.65	27.07	72.93	0.76	No
20.42	6.65	27.07	72.93	2.75	No
20.42	6.65	27.07	72.93	9.51	Yes
20.42	6.65	27.07	72.93	0.00	No
20.57	6.83	27.40	72.60	0.33	No
20.57	6.83	27.40	72.60	1.39	No
20.57	6.83	27.40	72.60	3.34	No
20.57	6.83	27.40	72.60	8.71	No
21.08	7.01	28.10	71.90	0.48	No
21.08	7.01	28.10	71.90	0.98	No
21.08	7.01	28.10	71.90	4.58	No
21.08	7.01	28.10	71.90	5.27	No
20.39 (average)	6.38 (average)	27.22 (average)	72.78 (average)	1.24 (Lowest Energy)	