Proceedings Paper:

Imran, FA, Morley, G, Anionwu., VL et al. (3 more authors) (2018) Mist Explosions using the Hartmann Dust Explosion Equipment. In: XII ISHPMIE. 12th International Symposium on Hazards, Prevention and Mitigation of Industrial Explosions – XII ISHPMIE, 12-17 Aug 2018, Kansas City, MO, USA. ISHPMIE .

Mist Explosions using the Hartmann Dust Explosion Equipment

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Abstract

Explosion hazards in the USA and EU include gases, vapors, mists and dusts. There is guidance on the measurement of lean flammability limit and reactivity for gases, vapors and dusts but not for mists explosions. This work explores the use of a modified Hartmann explosion tube for investigating the lean flammability limit and reactivity of mist explosions. Flammable liquids below their flash point can explode if the liquid is atomized and pump diesel fuel was investigated in this work, as the flash point is above 60°C. Each liquid has a critical drop size necessary for it to explode and this is related to the liquid volatility and viscosity. It is about 70µm for kerosene and 10µm for tetralin. For a liquid mist to behave close to a gas in an explosion the drop size has to be small enough to be heated and vaporized in the time it takes the droplet to travel through a laminar flame preheat zone, which is about 1mm at stoichiometric conditions. The use of high pressure air injection in dust explosion test equipment was investigated using the ISO 1m^3 dust explosion vessel, but this could not be made to work, nor could the standard Hartman equipment. However, a simply modification to the Hartman equipment enabled repeatable mist explosions to be generated for diesel. The air deflector was removed from the Hartmann equipment and the liquid injected down the air delivery tube. The reactivity of diesel mist was determined from the rate of pressure rise prior to the explosion tube vent bursting and from the measured initial flame speed. The lean flammability limit of diesel mists was determined as 0.15∅ and no rich limit was found, with tests up to an ∅ of 5. It was concluded that the Hartmann equipment provides a good method for the characterization of mist explosions for diesel like aerosols.

Keywords: mist explosions, atomisation, lean flammability limits, industrial explosions

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Kansas City, USA - August 12-17, 2018

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Introduction

The ATEX Directive [European Commission, 2014] states that an *"explosive atmosphere" means a* mixture, under atmospheric conditions, of air and one or more dangerous substances in the form of gases, vapours, **mists** or dusts in which, after ignition has occurred, combustion spreads to the entire *unburned mixture"*. In the UK this requirement has been part of Process Safety Law since the Factories Act 1930 and 1961 in s.31, where precaution have to be taken for dusts, gases, vapour or substances that if they escape are liable to explode if there is an ignition source. In this case 'substances' would include aerosols or mists. The modern guidance for explosion risks is DSEAR (HSE, 2016) which has the ATEX definition including the requirement to protect against mist explosions. Guidance exists for the measurement of flammability limits and reactivity of gases and vapours (BSI, 2004) and dusts (BSI, 2006) and for what constitutes an explosive atmosphere (BSI, 2009); but there is no Guidance in Europe, the UK or the USA on protection measures for mist or aerosol explosions, in spite of the long history of the law requiring protection against aerosol or mist explosions. The HSE guidance on explosion risks (DSEAR ACOP, paragraph 145) does draw attention to aerosols as an explosion risk: *"Some substances do not form explosive* atmospheres unless they are heated, and some liquids if released under pressure will form a fine mist that can *explode even if there is insufficient vapour."* This makes it clear that it is not necessary to vaporise a flammable liquid for it to explode, all that is needed is for it to be atomised into droplets small enough to propagate a flame through the cold droplets, generally referred to as an aerosol or mist.

 The heat release in the flame front thermally conducts the air ahead of the flame and the droplets vaporise in this preheat zone before they reach the chemical reaction zone. Aerosols are liquid droplets finely dispersed at a drop size that is small enough for the droplets to evaporate and burn when a laminar flame consumes them. For many hydrocarbons this occurs when the aerosol drop size is less than about 70 microns (Zabetakis, 1965). Aerosol explosions are different from vapour explosions that occur when a liquid is heated above its flash point, in that the droplets are initially at the ambient temperature and pressure and the vapour levels are not sufficient to give a flammable mixture e.g. lubricating oil and hydraulic fluids, both of which have no realistic vapour explosion hazard but can explode as aerosols. Laminar flames for the most reactive mixture have a flame thickness of about 1.1mm which increases to about 10mm for near lean limit flames (Andrews and Bradley, 1972a&b). 90% of the flame thickness is the preheat zone, where thermal conduction of heat from the reaction zone occurs. Thus the vaporisation of the fine droplets in aerosols occurs inside the flame preheat zone as the flame propagates into the aerosol, in a distance of the order of 1mm.

Spalding (1951) analysed the time to preheat and burn a droplet of kerosene in an aerosol in a laminar flame. For a laminar propagating flame with a flame speed of 1 m/s and a preheat zone thickness of 1 mm his analysis shows that the residence time is 1ms. For a droplet to vaporise in this time it has to be <30μm. For a near limit flame the flame speed might be 0.5 m/s and the flame thickness 5 mm. The residence time is then 10ms and the drop size needs to be <80μm. Browning and Krall (1955) have shown that a Bunsen burner flame cone with a kerosene aerosol in air of <70µm drop size could be supported and it appeared identical in shape to a propane Bunsen flame at 0.75 equivalence ratio (Ø), with similar burning velocity.

Aerosol explosions have a similarity to dust explosions through the importance of the aerosol drop size and the dust cloud particle size on the lean flammability and flame reactivity (Eckhoff, 2003; Andrews and Phylaktou, 2010, Slatter et al., 2013). Dust explosion hazards are regulated and have recognised experimental equipment (BSI, 2006, Eckhoff, 2003) for lean flammability and reactivity measurements. For a regulated test the dust must be sieved to <63µm and for larger particle sizes the reactivity decreases and the lean limit becomes richer (Slatter et al., 2013). The lean limit for dust

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explosions was shown by Andrews and Phylaktou (2010) to be leaner than for gaseous hydrocarbon explosions with published results at around 0.2Ø compared with 0.45Ø for most hydrocarbons. Typically the volatile fraction of HCO composition dusts, such as wood, is about 70%. If only the volatile fraction burnt then the lean flammability limit would be richer than for hydrocarbon gases and so all the mass that burns must be converted into a gas due to greater volatile release at the higher heating rates in flames. However, this does not explain the leaner burning mixtures if the volatiles were hydrocarbons. Wood has volatiles of CO, H_2 and various hydrocarbons (Irshad, 2014) and this may give a wider lean limit due to the presence of hydrogen. For pure hydrocarbon aerosols it is expected that aerosols with size <70μm would behave as a hydrocarbon gas with a lean limit of 0.45Ø.

Similar effects will occur with aerosols, but currently the droplet size for a mist or aerosol explosions is not specified for testing. However, following the dust explosion experience a droplet size < 70μm is likely to be reasonable with lower values for more viscous higher boiling point liquids. This proposed 70μm critical size is also in agreement with the laminar flame preheat zone vaporisation time, as discussed above, and for the Bunsen burner kerosene aerosol work of Browning and Krall (1955). Also, for kerosene Zabetakis (1965) showed that the critical size for kerosene aerosols was about 70μm and below this size the lean flammability limit agreed with that for vaporised kerosene at 0.58Ø. This kerosene work was based on the original measurements of Anson (1953) and Zabetakis and Rosen (1957). For other liquids Odgers and Kretschmer, (1986) found that the critical drop size was 50μm for iso-octane with a lean limit of 0.42Ø, 75 μm for diesel with a lean limit of 0.5Ø, 60μm for heavy fuel oil at 0.56Ø. Burgoyne and Richardson (1949) using a tube explosion method reported the lean limit for diesel aerosols at 0.56Ø and for cutting oil No. 1 it was 0.49Ø, but the droplet size was not given. Burgoyne and Cohen (1953) reported that Tetralin had a critical drop size of 40μm with a lean flammability limit of 0.54Ø. In contrast Zabetakis (1965) reports a critical drop size of 10μm for tetralin with a lean flammability limit of 0.58Ø.

The distinction between a vapour and aerosol or mist explosion is clear in the regulations on flammable liquids which are concerned with vapour-air flammability and the temperature a liquid has to be heated to, for the vapour to be flammable in air at ambient conditions. The UN Globally Harmonised System (GHS) on the classification and labelling of chemicals has been adopted in Europe and the UK (European Commission, 2008) by the CLP Regulations as well as in the USA by the OSHA Flammable Liquid Classifications Standards (2010) (OSHA CFR 29, Standard No. 1910.106 & 1910.106(a). Flammable liquids are split into four categories governed by the temperature the flammable liquid has to be heated to produce sufficient concentrations of vapor to be flammable (flash point test):
Category 1 – flashpoint < 23 °C and Initial Boiling Point ≤ 35 °C
Category 2 – flashpoint < 23 °C and Initial Boiling Point > 35 °C
Category 3 – 23 °C ≤ flashpoint ≤ 60 °C
Category 4 - 60°C ≤ flashpoint ≤ 93°C
However, what is not made clear is that for categories 3 and 4, fuel temperatures below 60°C and 93°C respectively, where no vapour – air explosion could occur, there is nevertheless an aerosol explosion risk (Puttick, 2008).

Kerosene is a Category 3 flammable liquid and Diesel is Category 4. Diesel is used in the present work to develop experimental methods to investigate aerosol explosion hazard at ambient temperature of the liquid and the air. A further problem is that vapour explosions are limited by the GHS and CLP definition of a flammable atmosphere to air as the oxidant at ambient temperature and pressure conditions. This excludes heated air as the auto-ignition source in aerosol explosions, but does cover hot surface ignition of aerosols at ambient air temperature.

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At the time when the European directives were issued in 2003 there was no guidance on how an area where there is a risk of a mist explosion should be classified (Forsth et al., 2011). Work started to solve the mist problem, which ended up in Annex D of IEC 60079-10-1 (2015). In this Annex some guidance is given on the explosion hazard from flammable mists generated by the release under pressure of high flash point liquids. However, it notes that from currently available regulations there is no standardised test method for aerosol flammability and reactivity**.** This is of particular interest for the Chemical and Process Industries, as the determination and characterisation of hydrocarbon aerosols, which can be formed below a liquid fuel's flash point, play a significant role in determining the safety of such industries. Also, in conducting the Hazardous Area Classification in the Petroleum industry (Energy Institute, 2005) the flammability and reactivity of aerosol explosion risks need to be known.

Table 1: A list of Liquids Considered by HSE that Could Form Mists, Sprays, and Aerosol below their Flashpoints at Ambient Temperature [Puttick,S., 2008]

Lubricating Oil	Vegetable Oil
Hydraulic Oil (both mineral and synthetic types)	Light Fuel Oil
Heavy Fuel Oil (at operating temperature)	Heat Transfer Fluid (at operating temperature)
Jet Fuel / Kerosene	Transformer Oils
Process Fluids	Diesel
Bio-diesel	White spirit

Table 2: Methods of Accidental Production of Flammable Liquid Aerosols (Stanton, 2009; HSE, 2013 and Lefebvre, 1989)

1. Through a high pressure leak (e.g. hydraulic fluid). There are two mechanisms occurring here, the principal one is due to spray break-up in the jet and the secondary mechanism is further fragmentation of the liquid droplets as they hit any neighbouring solid surfaces.
2. Through a high velocity gas interacting with the liquid (e.g. compressed air on liquid)
3. Through condensation (vary rare in incidents, but a common experimental technique). When a liquid comes into contact with a hot surface, a hot vapour may form due to evaporation. Further downstream this hot vapour may cool, producing an aerosol cloud.
4. Through mechanical agitation (e.g.in large marine diesel engine lubricating oil sumps, Freeston et al., 1956)

The HSE (Stanton, 2009; Gant et al., 2012) acknowledges the fact that aerosols can be formed below the flash point of the corresponding liquid fuel, and listed in Table 1 are some of the liquid fuels that could exhibit aerosol explosions. Stanton (2009) also categorized in Table 2 different ways the aerosol might be formed accidentally. Flammable liquids can explode at ambient conditions below the flash point if the liquid is atomized and hence the risk of an aerosol explosion is the same as the risk of atomization of a liquid leak. Table 2 is thus a list of methods by which an aerosol can be formed accidentally and it is also a list of experimental methods by which aerosol explosions can be generated in standard test equipment. In the present work the use of high pressure air jets to atomise a liquid spillage or leak is used as an experimental method.

Ahrens et al. (2002) review NFPA statistics on aerosol explosions where 834 industrial fires were reported to occur due to ignition of class IIIB (now obsolete or non-flammable class, as it is beyond Class 4) combustibles in the U.S.A. during the period 1980-1998 [3]. Class IIIB combustible liquids

include cooking, transformer, lubrication, motor, mineral, and hydraulic oils. They have a flashpoint at or above 93.4°C. The fires on average caused 1 death, 37 injuries, and direct property damage to a value of $24M each year. These fires, where class IIIB liquids were the item first ignited, accounted for 1% of all the fires in these occupancies. The statistics do not reveal whether the liquids were ignited in a spray configuration or a pool configuration. However, 29% of the fires were caused by part failure, leaks or breaks, which could give rise to aerosol formation. The biggest part of the fires, 44% out of 28 listed occupancy types, took place in metal and metal product manufacturing premises. One reason for this is probably that hot surfaces are abundant in those industries. Sprinklers were effective in reducing the loss from this type of fire.

According to statistics from FM Global [Loss Prevention Data, 1993] 134 fires involving hydraulic systems were reported in the period 1981-1990. Half of these fires occurred in the metal industry. A total of 40% of the fires were caused by hydraulic oil being sprayed onto a hot surface. The 30% of fires with no sprinklers accounted for 75% of the property damage. Heat transfer oils (Table 3) were reported by Febo and Valiulis (1995) to be responsible for 54 fires and explosions over a ten year period with a cost of $150M in losses.

Forsth et al. (2011) have studied drop size generated in hydraulic oil leaks with oil pressures of 50, 100 and 150 bar with leaks through 0.25mm and 0.45mm holes and through a short slot 0.23mm wide. It was found that the larger droplets, 50 μm and 100 μm, rain out completely within a time frame of 10-30 s after the end of a 10 s release. Typical drop sizes were 30 - 45µm. The smallest 15 µm droplets carry all airborne liquid after this time. These droplets on the other hand rain out slowly and, if injected in sufficient quantities, remain a hazard for several minutes or more. Oil pressure was the main factor affecting the amount of airborne liquid after 120 s. It is thus clear that hydraulic oil leaks can generate aerosols of a size below the critical level to enable a propagating explosion flame to exits if there is an ignition source. Forsth et al. (2011) also showed that FDS was a good modelling tool for predicting the spray size and penetration.

The drop size necessary to give an aerosol explosion will be shown to depend on the flammable liquid composition, boiling point, density, viscosity and surface tension, as all these factors influence the drop size using standard atomisers (Lefebvre, 1989). However, Spalding's (1951) analysis shows that drop sizes are likely to be <80µm for a propagating aerosol explosion, as shown above. It was shown above that in the literature each flammable liquid has a critical drop size necessary for it to explode at ambient air T & P. This is also the critical (maximum) drop size for burners and engines using liquid fuels. Diesel is the liquid fuel for compression ignition engines and kerosene is used in aero gas turbines, both use spray atomisation to produce droplets below the critical drop size, usually < 70 µm. Most of the work on liquid aerosols for gas turbine applications, with burning below the flash point of the liquid, has been carried out using Method 2 in Table 2 with preheated air to typically 800K by compression. Explosion risks from liquid leaks normally occur in ambient air, apart from in specific process hazards where preheated air is used. In diesel engines the prime method of atomisation is Method 1 in Table 2, high pressure fuel injection through a small hole, with the fuel pressure controlling the drop size for a fixed hole size and up to 3000 bar is used in modern diesels, with hole sizes of about 0.1mm. In gas turbines the prime method of fuel injection is Method 2 in Table 2, using the combustion air flow for atomisation with the combustor pressure loss controlling the air velocity in the range 60 – 100m/s. The present work adopts air injection in dust explosion test equipment for Method 2 atomisation of liquids.

Table 3. Summary of liquids involved in aerosol explosion incidents

		Number of incidents –Total 37
Below the flash point at ambient conditions		
Oils:	Lube oil	5
	Transformer oil	1
	Vegetable oil	3
	Hydraulic oil	1
	Fuel oils	7
	Crude oil	3
	Heat transfer oil	4
Kerosene		7
Naphtha		4
Butyl acrylate		2

Another practical application of liquid fuels below their flashpoint is the use of heavy fuel oils for steam generation and subsequent electric power generation. In this case both the air and the oil are normally heated; typically 600K for the air and 500K for the fuel (Anson, 1953). Atomisation normally uses steam as the atomizing gas for Method 2 in Table 2. High pressure steam is a common heat source in process industries and hence a steam leak in the presence of a liquid fuel leak can give an aerosol explosion risk.

Stanton (2009) and Gant et al. (2012) have reviewed aerosol explosion incidents, as summarized in Table 3. 37 incidents were identified, which was not claimed to list all incidents. In some cases, the incidents may have resulted in large fires following the initial aerosol explosion. In all cases, the incidents arose from the ignition of a mist, in most cases at a temperature near or below the liquid flash point. These incidents are categorised in Table 3. The above incidents exclude large industrial diesel engine lubricating oil sump explosions, which are very common (Freeston et al., 1956). The lube oil in diesel engines is hot ~ 120°C and the agitation of the piston con rods cause splash atomisation of the lube oil. Many diesel engine oil sump explosions have occurred and their sumps always have explosion venting protection, using multiple vents around the periphery of the sump.

An incident not in the above reviews was at British Steel (Redpath, 1995) where a new steel strip rolling mill was destroyed by a hydraulic oil fluid leak due to a hydraulic shock absorber not being bolted down. This fractured the hydraulic connections and led to a large hydraulic oil aerosol explosion through pressure jet atomisation, ignition through hot surface autoignition and a subsequent explosion which blew off the roof of the new rolling mill. The incident also led to a major fire. This was a new rolling mill for which sprinkler protection was advocated but was deemed

Table 4 Sources of ignition of 37 aerosol explosions (Stanton, 2009).

Compression autoignition	1
Electric arc	1
Hot surface	3
Electro Static	6
Combustion chamber/igniter/pilot flame	10
Hot work	1
Engine induction	1
Catalyst	1
Naked light	1
Unknown	12

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to be too expensive. Jagger (2003) has reviewed the fire and explosion hazards of hydraulic fluids. The importance of hydraulic fluid high pressure accidental release aerosol explosions was shown by Willauer et al. (2006) who carried out large-scale explosion tests aboard the USS Shadwell, 2190 TEP hydraulic fluids was heated to 50°C and pressurized to 100 bar. The flashpoint was 246°C and the BP >315°C with a CV of 42.7 MJ/kg. A 65 second continuous spray of 2190 TEP was generated using a single nozzle in the nozzle array with a mist flow rate of approximately 2.45 liters/min. This is approximately a 1.5MW energy flow for 65s or a total energy in the aerosol of 96MJ. The results for three repeat explosions showed that the average overpressures generated were about 5.5 kPa in an enclosure of about $30m^3$ volume.

Stanton (2009) has reviewed the ignition sources in the 37 incidents in Table 3 in Table 4. A large proportion of incidents had no known ignition source and the presence of an ignitor flame on other equipment was the most common ignition source. The next most common ignition source was hot surface auto-ignition. Stanton (2009) comments that the numbers of records (18 out of 27) and incidents (25 out of 37) originating from USA and UK combined are disproportionately high. It is improbable that the incident rates are so much higher in these regions and the numbers of records suggest that the recognition of the hazard and/or the reporting of incidents are simply more thorough, or that the survey was biased towards literature relevant to these countries. Stanton (2009) comments that some very large explosions have arisen from mist ignition and that 9 incidents alone were collectively responsible for a total of 29 fatalities. As shown by the listed records, incidents that are reported in the open literature as of interest are more likely to be severe or major. It must be assumed that a larger number of less serious incidents have also taken place but have not achieved reported prominence in the same way.

2. Aerosol Formation through a High Velocity Gas Interacting with Flammable Liquids

Method 2 in Table 2 has atomisation using a high velocity gas, usually air or steam, with fuel injection through a small hole of diameter, d_o. High air velocity, U_a, over the surface of a liquid jet strips droplets to produce a mist (Lefebvre, 1989; Gant et al., 2012). Lefebvre's (1989) correlation for air blast atomization is given in Eq. 1

$$\frac{SMD}{d_0} = \left\{ A \left(\frac{\sigma_l}{\rho_a \cdot U_a{}^2 \cdot d_0} \right)^{0.5} + B \left(\frac{\mu_l{}^2}{\rho_l \cdot \sigma_l \cdot d_0} \right)^{0.5} \right\} \cdot \left(1 + \frac{F}{A} \right) \tag{1}$$

$$\underbrace{\phantom{A \left(\frac{\sigma_l}{\rho_a \cdot U_a{}^2 \cdot d_0} \right)^{0.5}}}_{\text{1/Weber number}} \quad \underbrace{\phantom{B \left(\frac{\mu_l{}^2}{\rho_l \cdot \sigma_l \cdot d_0} \right)^{0.5}}}_{\substack{\text{Ohnesorge (1936)} \\ \text{number}}}$$

The constant A and B in Eq. 1 depend on the specific geometry of the atomiser. The Weber number term in Eq. 1 includes the velocity term and the Ohnesorge number is the term that includes the viscosity and liquid density terms. The surface tension of the liquid is in both terms. The SMD drop size is proportional to the square root of the liquid jet diameter or fuel hole leak and also inversely proportional to the gas jet velocity. The SMD reduces as the air mass flow increases due to the (1 + F/A) term which shows that a large air flow produces smaller droplets. The SMD increases linearly with liquid viscosity and this is the main liquid property term that controls the Ohnesorge number. The present work is for diesel like liquids and the technique does not result in good atomisation for vegetable oils and lubricating oil due to the high values of the Ohnescorge number, which then dominates the mean drop size.

3. The Measurement of Flammability Limits for Gases and Dusts

Most publications that quote a lean or rich flammability limit for gases quote values from Coward and Jones (1951) or the update of this publication by Zabetakis (1965). The original report was joint

UK/USA research, as Coward was Head of the Safety in Mines Research Establishment at Buxton, which is now the HSE Explosion Research Laboratory, and Jones was Head of the US Bureau of Mines in Pittsburgh, which is now part of NIOSH. The experimental flammability method used was a 50mm ID 1.5m long vertical tube that was ignited at the bottom and the flame had to emerge from the top i.e. a 1.5m propagation distance. Mixtures that only propagated part way along the tube were deemed not to be flammable. The current European standard method for the measurement of gas and vapour flammability limits (BSI, 2003) is based on a German DIN standard and uses an 80mm ID 0.3m long vertical tube with a spark located 60mm from the bottom of the tube and a flammability limit standard based on a minimum flame propagation distance of 100mm from the spark, this is the tube or T method. The vertical tube is enclosed in an oven with up to 70°C heating so that vapours could be measured in the same equipment. In addition the standard has an alternative method, the Bomb method B, which uses either a 5L spherical or cylindrical closed vessel explosion with central ignition and a 50mb pressure rise as the flammability limit criteria. If a cylinder is used then it must have an L/D between 1 and 1.5. This gives a minimum 212mm diameter for a sphere and a minimum tube diameter of 185mm for L/D=1 or 162mm for L/D=1.5 (L=243mm). For central ignition this gives a flame vertical travel distance of between 106mm for the sphere and 92.5 – 120.5 for the range of cylinder L/D, which is a similar flame propagation distance for the tube method. Essentially the EU tube method has deemed that partial flame propagation <1.5m reported by Coward and Jones (1952) to not be flammable are deemed to be flammable in the EU method. Andrews and Bradley (1973) showed that close to the lean and rich flammability limits the hot flame kernel was convected vertically and extinguished either by heat losses on the top of the closed vessel explosion or by the aerodynamically induced vortex ring created by the interaction of the outwards expanding flow from the flame and the vertical convective rise of the low temperature near limit flames.

Table 5: Flammability Limits by the EU and US BM Tube Methods in Equivalence Ratio, Ø

Gas	Lean EU (BSI 2003)	Rich EU (BSI 2003)	Zabetakis US BM 627 1965 Lean	Zabetakis US BM 6271965 Rich
Methane	0.45	1.90	0.50	1.68
Ethylene	0.38	6.58	0.41	8.04
n-hexane	0.46	3.70	0.56	2.85
1,3,5 trimethyl benzene (1.59% Ø=1)	0.50 70°C	4.31		

Table 5 compares the EU (BSI, 2003) and US BM lean and rich flammability limits converted to equivalence ratio, Ø. For lean mixtures the EU tube method has lower limits than the US BN method, 10% lower for methane, 9% lower for ethylene and 18% lower for n-hexane. For rich mixtures the agreement of the two methods is poorer and inconsistent. The EU method has a 13% richer limit for methane, a 18% leaner limit for ethylene and a 31% richer limit for n-hexane. However, the EU method (BSI, 2003) only requires 10% agreement with the values in Table 5 for a measurement laboratory to be certified, so that the two methods agree for some lean limit gases as the differences in Table 5 are <10% for methane and ethylene.

Table 5 shows that the lean flammability limit for hydrocarbons is to within 10% 0.5Ø for all hydrocarbons, apart from ethylene. Acetylene and hydrogen are the other more reactive gases that have leaner limits than 0.5Ø. The vapour of flammable liquid aerosols would be the type of

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hydrocarbons with a lean limit of 0.5Ø and the published lean limits for liquid aerosols were reviewed above and most had a lean limit close to 0.5Ø with a range from 0.42 to 0.58. This indicates that the cold liquid aerosols have a lean limit that is close to that of the fully vaporised fuels. This is because the drop sizes tested were sufficiently small to vaporise the droplets in the 1mm thick preheat zone of the flame. In the review of Bowen and Sherville (1994) it was recommended that in explosion hazard assessment, the simplistic current practice of modelling condensate fuels as propane vapor be continued, until further research provides us with the information required to model aerosol effects within hydrocarbon explosions more rigorously. 24 years later we are in no better position in aerosol explosion hazard assessment and this recommendation in terms of the lean limit is supported by the above evidence. However, the present results show that leaner mixtures than in the literature can propagate an explosion and this is in agreement with dust explosion data.

It has been shown above that aerosol explosions have many characteristics that are similar to dust explosions, most importantly the dependence of the mixture reactivity and LFL on the particle or droplet size. The experimental methods used to investigate dust explosions are the 1 m^3 and 20L spheres and the Hartman vertical tube (Eckhoff, 2003; BSI, 2002 and 2006). The two closed vessel spherical explosion techniques use 10 kJ chemical ignitors which gives a 50mb pressure rise in the 1m^3 sphere and 1 bar in the 20L sphere. This large pressure rise in the 20L sphere from the ignitor makes it unsuitable for lean limit measurement as the pressure rise increased the air temperature and this results in leaner limits (NFPA68, 2013). The EU standard on the lean flammability and reactivity measurements of dusts (BSI, 2006) gives the ISO standard 1 m^3 spherical closed vessel method as the prime measurement method, but allows the Hartman tube method to be used for flammability measurements if it can be shown to give the same results as for the 1 m^3 method. Andrews and Phylaktou (2010) have reviewed the literature and shown that the two methods do give similar results for the lean flammability limit or minimum explosion concentration (MEC). This is why the Hartmann equipment has been used in the present work.

The Hartmann vertical tube is 69mm ID and 335mm long with a 4J spark 120mm from the bottom, where the dust is placed and dispersed with a blast of compressed air. Comparison with the EU (BSI, 2006) tube method for gas flammability limit determination shows that these two pieces of equipment are very similar and this was why this equipment was used to investigate aerosol explosions in the present work. Also as an existing standard piece of explosion test equipment it could easily be incorporated into a future aerosol explosion testing standard.

4. Laminar Burning Velocity and Flame Speed Measurements for Aerosol Explosions

The laminar burning velocity or spherical explosion flame speed are measurement of the reactivity of a mixture and for explosion protection it is not sufficient to know the flammability limits, the mixture reactivity must be known for vent design and explosion suppression protection design. In NFPA68 (2013) the statement is made that there is no information that relates the reactivity of aerosol (0 - 30µm) explosions to vent area requirements. In many cases the same equipment that has been used to determine the lean and rich flammability limits for aerosols has also been used to measure the flame speed and then derive the burning velocity. Andrews and Bradley (1972a) have reviewed methods of measuring the laminar burning velocity and shown that most methods have systematic errors related to the finite thickness of the flame front and flame curvature (or flame stretch) effects. Typical many measurement methods had burning velocity values 20% to low because of systematic errors in the measurement method. Unfortunately, all published flame speed and burning velocity data for aerosols also has systematic errors that were there when the techniques were used for gases. The same problem exists for dusts and there are few published measurements of dust burning velocity (Eckhoff, 2003). The limited flame speed data in the literature,uses the tube

method, spraying the aerosol from the closed top of a vertical tube which is ignited at the open bottom using a flame, so the test method is close to that for aerosol flammability. In the work of Burgoyne and Richardson (1949), Burgoyne and Cohen (1953) and Burgoyne (1957 and 1963) the tube method was extensively used with tube sizes from 17-70mm diameter. Only the largest of these will give flame speeds independent of the tube diameter. However, although the flame speed in a tube is related to the mixture reactivity, Andrews and Bradley (1972a) showed that it could not be easily related to the fundamental laminar burning velocity or to the spherical flame speed.

Bowen and Cameron (1999) have reviewed the methods for measuring the flame speed in aerosols. They highlighted the widespread use of aerosol formation by condensation, Method 3 in Table 2. This can be done by either vaporising the liquid and injecting the hot vapor into cool air to produce condensation, as first used by Browning and Krall (1955), who produced a 1µm spray or kerosene, which behaved as a gas in flame propagation. Similar condensation aerosols can be generated in closed vessels by changing the pressure (Lawes and Saat, 2010). Burgoyne and Cohen (1954) created monodisperse tetralin aerosols using this technique in the droplet size range 8±38 pm. They found that a transition in the character of the flame took place at about 10 pm, below which the appearance of the flame was that of a pre-mixed gaseous flame, as discussed above in relation to the flammability limits. For larger drop sizes, the flame took the form of a brush-like spray of discrete burning drops, where drops burned diffusionally in their own envelope of air and flame, one burning drop igniting adjacent ones, spreading combustion in a `network' fashion. However, this mode of burning narrowed the flammable range. It was concluded that the burning velocity in an aerosol of `large' drops is significantly higher than for a comparable aerosol of `small' drops. NFPA68 (2013) is based on venting design using the laminar burning velocity as the reactivity parameter and it discusses this influence of burning velocity dependence on drop size in some detail. However, there is no application of the venting guidance to aerosols, in spite of the long discussion on the burning velocity of aerosols. Lawes and Saat (2010) have undertaken similar work with iso-octane over the size range 5 – 20 µm. The spherical flame speed at the maximum reactivity mixture varied from 2.5 to 2.9 m/s at 1.3Ø. For lean mixtures size had no influence on the flame speed, but for mixtures at 2Ø, the spherical flame speed increased with droplet size. The explanation given was partial evaporation of the droplets, but this is unlikely in this size range as Spalding's (1951) droplet flame evaporation theory shows.

The largest explosions of aerosols in the literature were undertaken by Kopyt et al. (1989) who created 18—28 µm spray droplet size using a superheated fuel atomizer with a $600m^3$ aerosol fuel cloud (10.5m diameter) dispersed in the open atmosphere and ignited centrally. The flame speeds increased with radius as shown in Fig. 1. At a critical radius the flame acceleration ceased and propagation was at a high velocity which was a function of the fuel type. Surprisingly, the highest flame speed was for the least volatile fuel (fuel oil) and the lowest flame speed for the most volatile fuel (gasoline). At smaller radius the reduced flame speeds were in agreement with those of Bowen and Shervil (1994) for kerosene and hexane. Incidents such as that at British Steel, discussed above, are likely to have had aerosol cloud sizes of the order in this work, but this does not make a good test method and a way of getting realistic aerosols on a laboratory scale is required for practical regulations.

The problem with the superheated fuel and condensation method of investigating aerosol explosions is that the drop sizes are too small to be representative of real accidental aerosol explosion incidents. They are also too small relative to practical gas turbine and diesel fuel injector (Lefebvre, 1989) spray sizes. Abdul Hussain and Andrews (1990) showed that with air blast atomization using high air flow rates and low F/A ratios in Eq. 1 gave the lowest SMD in the literature for the same velocity. Drop sizes varied from 20 to 100µm depending on the air velocity, but drop sizes <20µm were unlikely at practical air velocities. In dust explosion test methods the dust is dispersed with a blast of air from an

external high pressure reservoir, the principle investigated here is to use this air flow to atomise liquid fuels and then explode in the same way as for dust explosions.

Willauer et al. (2006) have developed a new method for investigating aerosol flame propagation which used rotating disc atomization of liquid fuels and was demonstrated with JP5. An aluminium rotary disk, 76 mm in diameter had a cavity 25 mm in diameter and approximately 6.3 mm deep in the center with four radial holes, spaced 90 degrees apart. Fluid is delivered to the center of the disk and a motor creates centrifugal forces that push the fluid through the radial holes, producing an aerosol. A propane fed Bunsen burner, located 130 mm from the edge of the disc, was the aerosol ignition source. Each aerosol system was studied over a range of rotational speeds from 1000 – 16,000 rpm and liquid flow rates to the disk of 50 – 200 mL/min. The SMD for JP5 was from 20 to 70 µm over the range of rotational speeds. Flame speeds were measured as in the present work from the time to propagate between thermocouples in an array. A propane flame was used to ignite the spray. The equipment was operated in ambient are and there was no control of the equivalence ratio of the flame. This equipment is useful for demonstrating whether a fluid was flammable as an aerosol, but the lack of control of equivalence ratio means that it is not suitable for MEC and flame reactivity quantitative measurements.
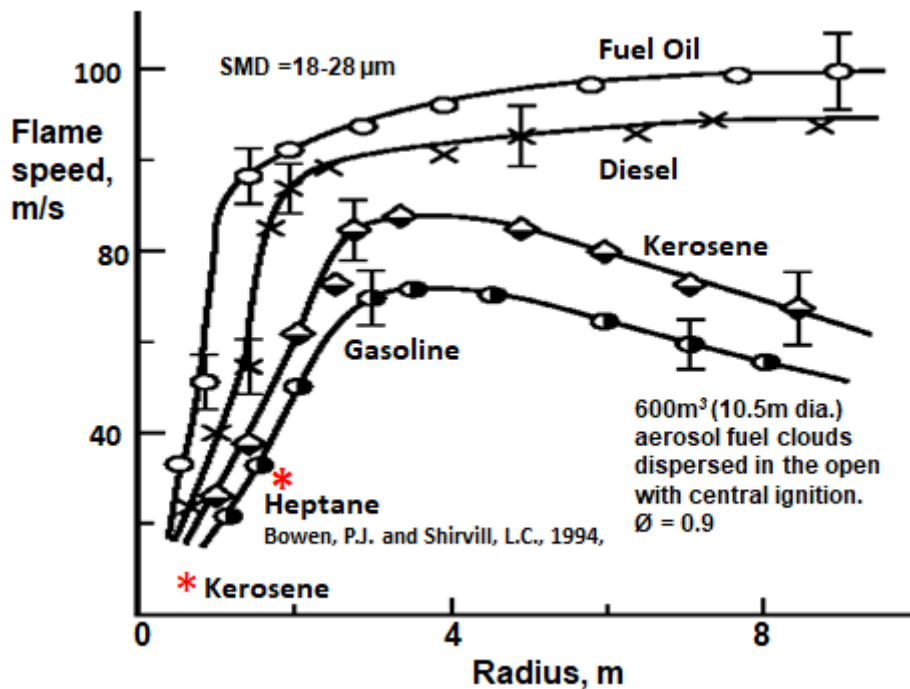

Figure 1: Flame speed as a function of radius for large aerosol clouds (Kopyt et al., 1989)

5. Investigation of the Use of the ISO 1 m³ Dust Explosion Vessel for Aerosol Explosions

The 1 m³ ISO dust explosion vessel, which is used in international dust explosion characterisation standards (BSI, 2006; ISO, 1985), has a 5L external air vessel that is filled with the dust and then compressed with air to 20bar. The dust is injected after opening a solenoid valve and the air pushes the dust down into the 1m³ vessel via a 'C' ring with the dust distributed through typically 24 4mm diameter holes. The application of this method of fuel injection was applied to diesel fuel which was placed in the external 5L chamber and dispersed with the 20 bar compressed air. This gives sonic flow at the C ring holes and should have given good atomisation. However, with the standard 10KJ chemical ignitor and standard 0.6s ignition delay no explosion occurred for 0.75 – 2.5Ø. After the test the diesel fuel was found coating the walls. Clearly this method of liquid fuel injection did not

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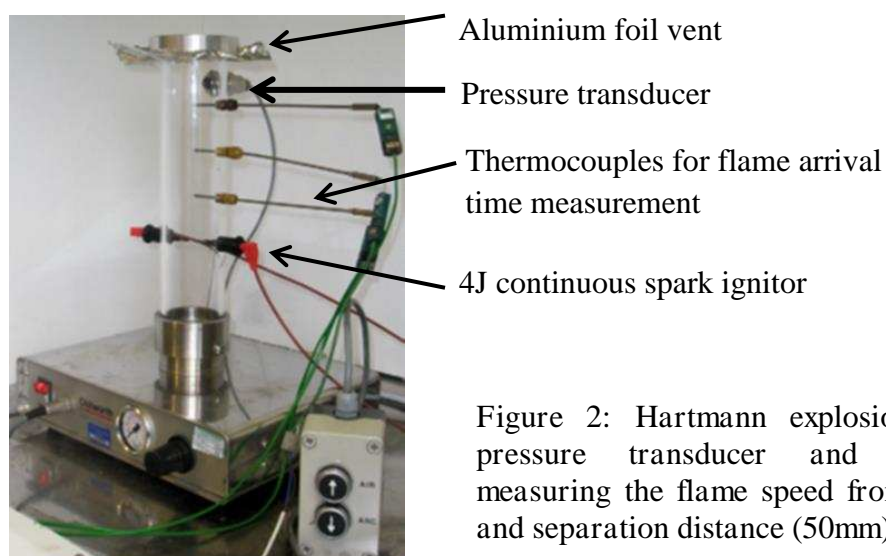
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cause atomisation as there was no high velocity air surrounding the fuel jet. Instead it appears the air pushed the diesel fuel as a slug of fuel through the C rings ahead of the air and simply forced it onto the walls of the test vessel.

For dust explosions with coarse powders Satter et al. (2016) and Saeed et al (2017) have developed a method of fuel injection based on the coarse powders being placed inside the vessel in a 0.4m diameter hemisphere in the bottom of the vessel. The external compressed air was dispersed though a tube inserted into the dust in the hemispherical chamber. This gave good dispersion of the dust and enabled dust explosions to be undertaken with very coarse biomass. This injection system was investigated with diesel fuel placed in the hemispherical vessel at the base of the 1 m^3 explosion vessel. The liquid was then atomised with compressed air injected into the diesel pool from the external 5L 20 bar air vessel. Unfortunately this atomisation and dispersion system did not work, even with a 10 kJ central ignitor. Again after the explosion the vessel was opened and the diesel fuel had been deposited unburnt on the vessel walls. This led the abandonment of using the ISO 1 m^3 explosion vessel for aerosol explosion characterisation.

6. Development of the Hartmann Explosion Tube for Aerosol Explosions.

6.1 Use of the Standard Hartmann System

Huescar (2013) and Saeed (2016) have described the development of the Hartmann equipment to enable flame speed and initial rate of pressure rise determined as measurements of the mixture reactivity, which correlates with the MEC and rate of pressure rise measured in the ISO 1 m^3 dust explosion vessel. This modified Hartmann equipment was used in the present work with the 4J continuous spark on, before the air injection occurred, as shown in Fig. 2. The standard Hartmann tube has a dust containment volume at the base of the tube that is dispersed with compressed air from a 60mL external volume pressurised to 6barg. The present Hartmann equipment, was that used for minimum ignition energy studies (BSI, 2002), used a Perspex tube with an aluminium foil vent at the top that burst at about 0.5 barg. The compressed air injection raised the pressure in the tube to 0.3barg. This enabled the pressure rise between the start of combustion and the bursting of the vent, to be determined as the initial rate of pressure rise. This is proportional the mixture reactivity. In addition the mixture reactivity was measure by determining the vertical flame speed on the centreline of the tube, using the time of arrival at three exposed junction Type K thermocouples at 50mm from the spark and then at 50mm intervals.

Aluminium foil vent

Pressure transducer

Thermocouples for flame arrival time measurement

4J continuous spark ignitor

Figure 2: Hartmann explosion tube with added pressure transducer and thermocouples for measuring the flame speed from the time of arrival and separation distance (50mm).

The Hartmann explosion tube was use with diesel fuel by placing the liquid in the container at the base of the tube where the dust is normally put. With the external air compressed to 6 barg the liquid was expected to be atomised. Instead there was no explosion and the diesel was deposited on the inside surface of the tube. This occurred for a range of Ø and it was clear that this was another method of aerosol generation that did not work.

6.2 Modification of the Hartmann to Enable High Velocity Air to Flow Over the Liquid
Air atomisation of liquids requires a relative velocity between the air and the liquid. Both the ISO 1m³ ISO dust explosion vessel and the initial Hartmann explosion tube methods had a high velocity air jet but did not have the liquid fuel injected into the air, but the air either pushed the fuel ahead of the air as in the ISO vessel C ring injector with liquid fuels or impinged onto a wall fuel film which resulted in the impingement air jet wall flow simply driving the liquid fuel ahead of it onto the tube walls, as in the standard Hartmann equipment. The ISO 1 m³ vessel with the liquid in a hemisphere on the bottom of the vessel also had the air jet impinge on the stationary liquid, which simply pushed the liquid onto the adjacent walls and did not produce an atomised spray that could be ignited.

The Hartmann equipment has a high velocity air jet that impinges on a deflector at the end of the air delivery tube, which is then deflected down onto the dust in the bottom of the explosion tube. The modification made in the present work was to remove the deflector and place the liquid fuel in the air supply tube using a syringe. The internal mechanism of the Hartmann equipment (manufactured by Chilworth Technology) is viewed from the bottom in Fig. 3. The ignition delay was a modification we made but its effect is not part of the present work, which was all carried out with the standard 4J continuous spark on before the air flow was initiated. Fig. 4 shows a close up, from the bottom, of the air passage from the control solenoid that releases the compressed air from the 60mL storage volume. The injected diesel fuel will flow under gravity down the delivery tube leaving some diesel fuel attached to the wall due to surface tension, but some will also accumulate in the bottom of the U bend. When air flow is initiated it will flow over the liquid on the floor of the tube or attach to the wall of the tube giving crossflow atomisation. This relatively crude liquid fuel injection system gave an aerosol explosion the first time it was used and gives sensible results as shown below.

The quantity of diesel was determined prior to the test to achieve a particular equivalence ratio. The total mass of air in the Hartmann is fixed at the end of the air delivery and this does not burst the aluminium foil vent. The mass injected can thus be calculated from the pressure in the explosion tube at the end of injection. The pressure transducer is flush with the wall of the tube just upstream of the vent. The volume of the Hartmann tube is 1L and after injection the tube pressure is 1.3bara so the total mass of air is 1.56g. For diesel with a H/C ratio of 1.9 the stoichiometric A/F is 14.3. For a stoichiometric mixture in the aerosol explosion this required 0.11 g of diesel, which at a density of 840 kg/m³ is an injected volume of 0.13 mL. The amount of liquid injected was varied around this amount to achieve different equivalence ratios.

110mg or 0.13 mL of liquid fuel is a very small quantity of diesel to inject and a mass weigh scale accurate to 0.1mg was used, so that the mass accuracy for a stoichiometric mixture was <1%. The technique to control the amount of fuel injected accurately was to put the approximate amount of fuel in a calibrated syringe, weigh the syringe and fuel, inject the diesel down the air delivery tube and then re-weigh the syringe to measure the diesel fuel still attached to the glass surface by surface tension after injection. The difference in weights was the mass of diesel injected. It was also found that for diesel fuel, after a successful ignition, there was evidence of liquid fuel impinged on the tube wall in the region near the air jet outlet. Tissue paper was weighed, the diesel wiped off the wall with the tissue and then the tissue paper was reweighed and this enabled the unburnt diesel to be reasonably estimated and the amount of diesel fuel burnt in the spray determined. The amount deposited as a % of the amount injected varied from 3 to 72% and increased as the amount of fuel

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injection increased which caused richer mixtures. Two equivalence ratios are reported in this work, the nominal injected fuel equivalence ratio and the corrected equivalence ratio to account for the fuel deposited on the tube walls. In recent work with kerosene there was no wall deposition of fuel that could be measured or seen.

After a test the compressed air was injected several times to clear any remaining diesel from the delivery pipes. However, any residual fuel was not measured in this work but occasionally some was visible, particularly for very rich mixtures. In future this residual fuel will be captured and measured so that the equivalence ratio can be corrected for this fuel that does not participate in the explosion. A simple tissue over the end of the tube would suffice to collect this fuel.
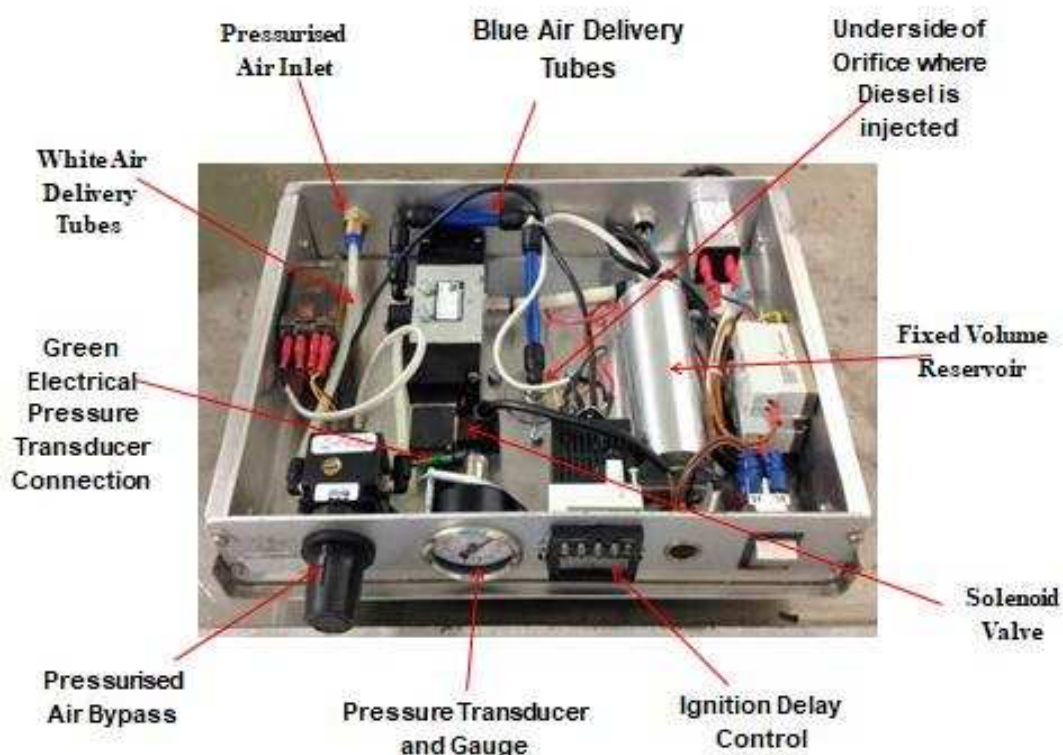
Figure 3: Air flow path in the Hartmann equipment with the liquid fuel injected into the blue air delivery tube so it will sit in the bottom of the U.

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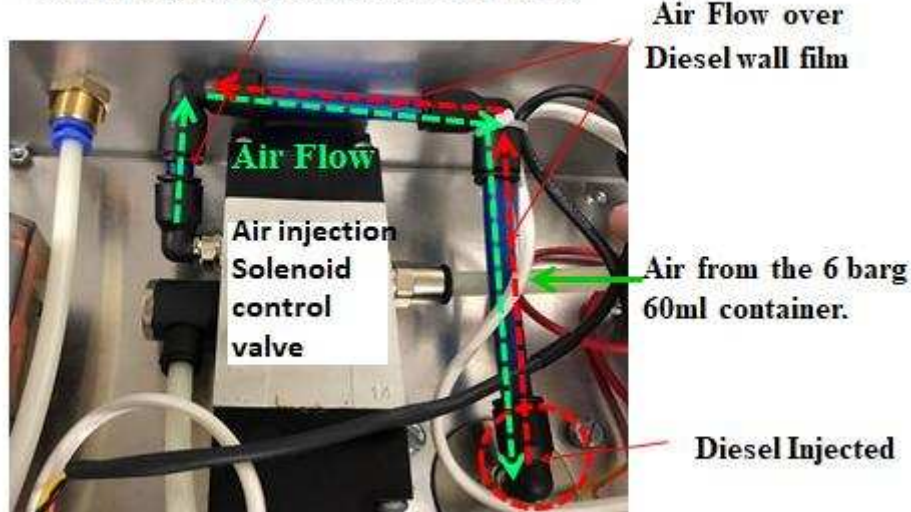XII ISHPMIE
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Figure 4: Close up of the Hartmann air flow pathway and the location of the diesel fuel that is injected into the tube and flow down under gravity into the top U bend (this picture is taken from the bottom of the equipment).
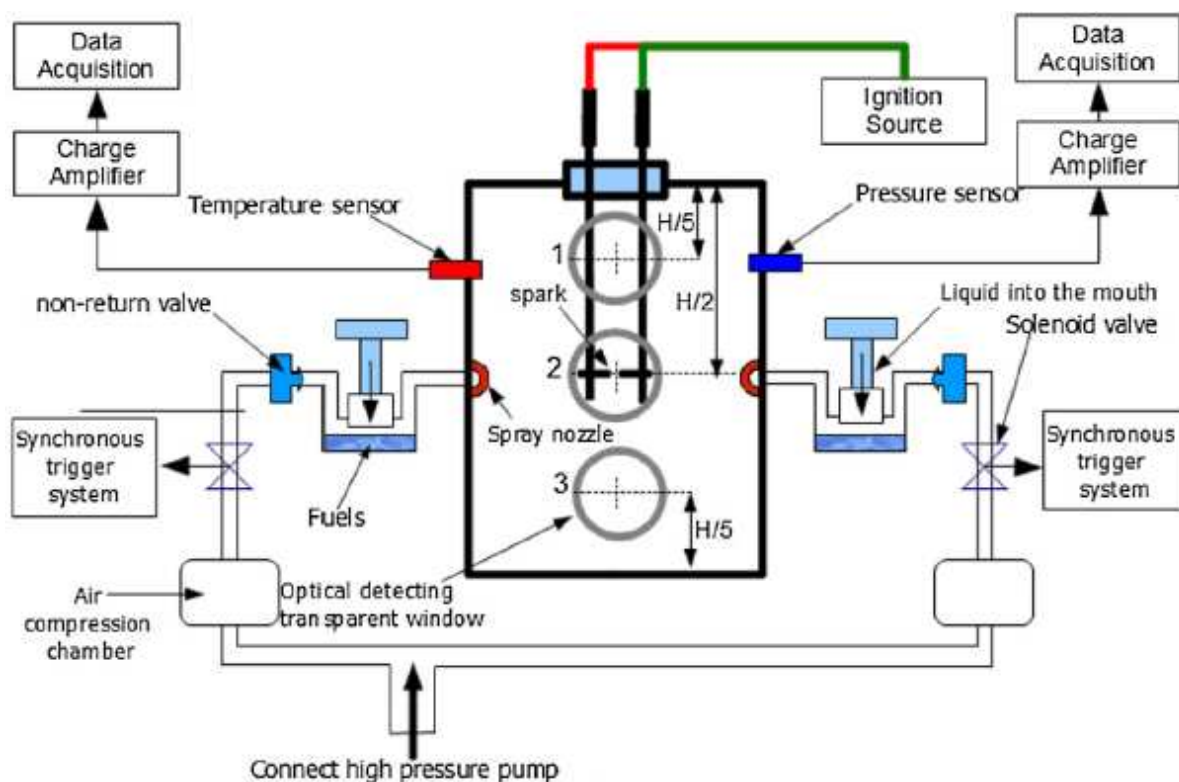

Figure 5 Method of liquid atomization used by Liu et al. (2016) in a closed vessel using high pressure air across the surface on the liquid as the atomization mechanism. Two opposed flow injectors used to achieve a stationary aerosol cloud.

Liu et al. (2015) have developed a similar method of liquid aerosol atomisation for aerosol flammability and reactivity determination, this is shown in Fig.5. It uses the same principle as in the present work, the crossflow of compressed air (5-8 barg) across a liquid surface. However, in their design two liquid injection systems are used in an opposed jet configuration, as shown in Fig. 5. Also closed vessel combustion of the aerosol occurs so that the maximum rate of pressure rise and K_G ($dp/dt_{max}/V^{0.33}$) can be determined. It is not clear from their work how the actual amount of liquid fuel that participated in an explosion was determined. However, as they studied JP-10 and dodecane, which are relatively volatile, they probably had no wall deposition losses. They found that very rich aerosols could burn and there was no rich flammability limit, which is also found in the present work. However, they could not explode lean mixtures and report a lean limit that converts to between 4 and 10Ø, which are extremely rich mixtures. This is difficult to understand as their spray size measurements indicate a size range from 10 - 100µm and SMD 20 - 35µm. These are completely at variance with the literature reviewed above and hence this method does not present a viable test method for aerosol flammability studies. The present work using a similar technique has no problem in exploding lean mixtures and finds lean limits lower than for gaseous hydrocarbons.

7. Results for Flammability and Reactivity for Diesel Aerosols using the Leeds Hartmann Method for Aerosol Explosions.

7.1 Characteristics of Diesel Aerosol Explosions
Figure 6 shows high speed videos of a near stoichiometric diesel aerosol explosion with a flame speed of about 2.7 m/s. The flame spans the tube diameter and is highly visible with radiation in the burnt gases, indicating heat release after the main flame front has propagated. This explosion was more violent than any dust explosion for coal or biomass or food products carried out in the Leeds work on dust explosions, as will be shown later. A near lean limit flame is shown in Fig. 7 with an
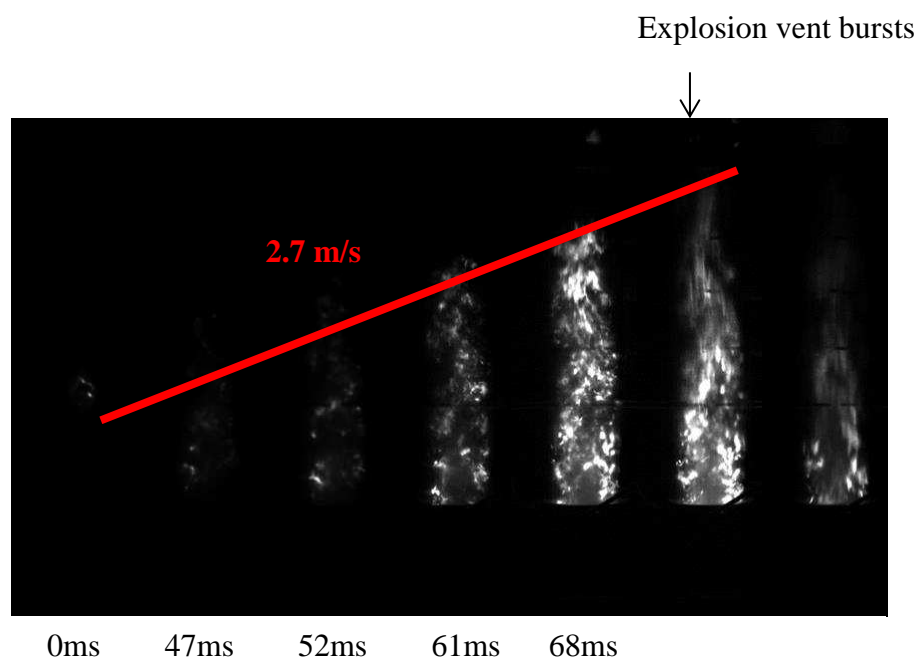

Figure 6: High speed videos of a near stoichiometric diesel aerosol explosion

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Explosion
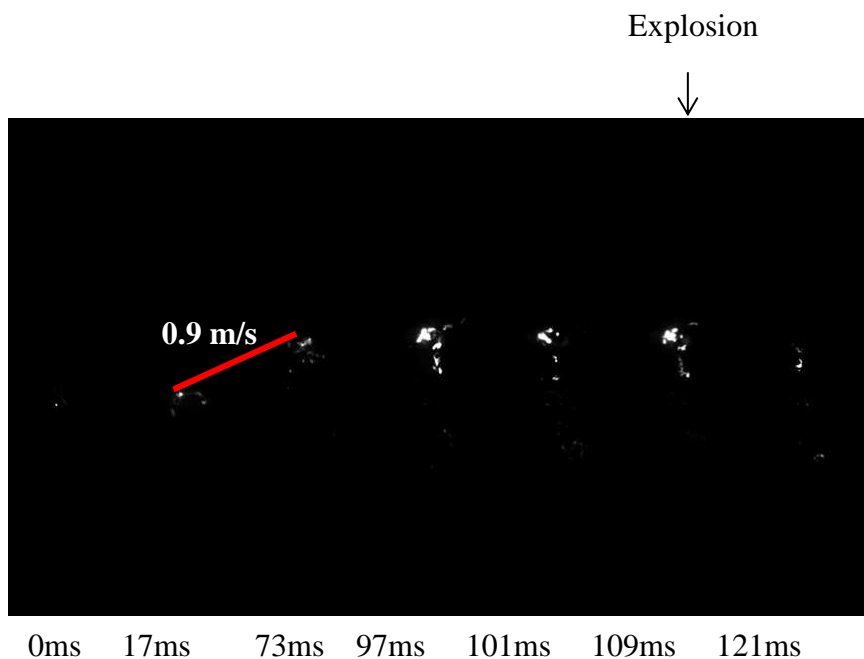↓

0ms   17ms   73ms 97ms 101ms 109ms 121ms

Figure 7: High speed video of a near lean limit diesel aerosol flame propagation

initial flame speed of 0.9 m/s which then has little further flame movement. This still had sufficient pressure rise to burst the vent cover and thus be counted as a flammable mixture.

An example of the pressure rise during a diesel aerosol explosion is shown in Fig. 8 and compared with an air only injection and no explosion. The increase in pressure above the air injection only test is proof that flame propagation has occurred. In the bomb method for gaseous flammability a pressure rise of 50mb is deemed to be a flammable mixture and the same criteria will be used in this
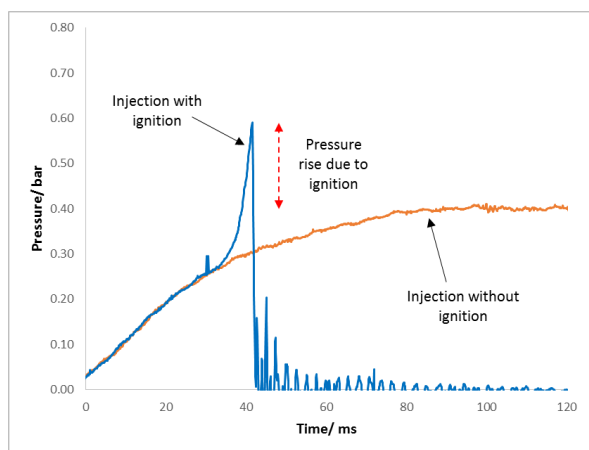

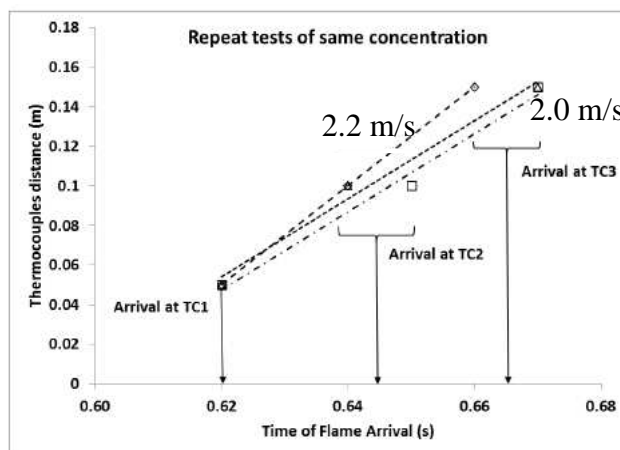Figure 8 Pressure time record for diesel aerosol   Figure 9: Flame arrival time v. TC position
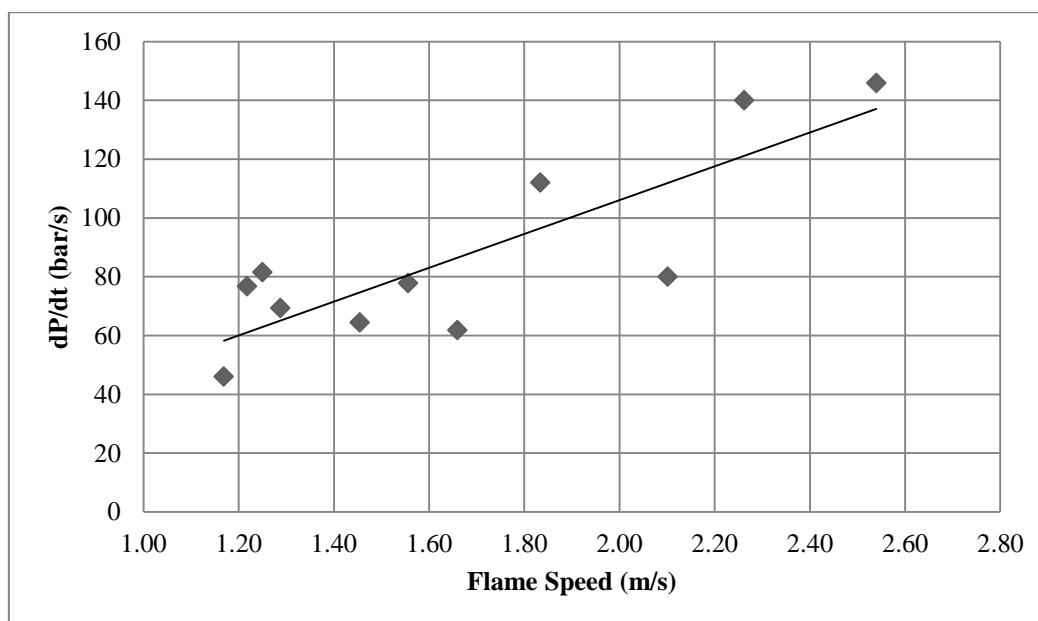
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Figure 10: Correlation of dp/dt$_{max}$ and flame speed for a range of diesel aerosols mixtures.
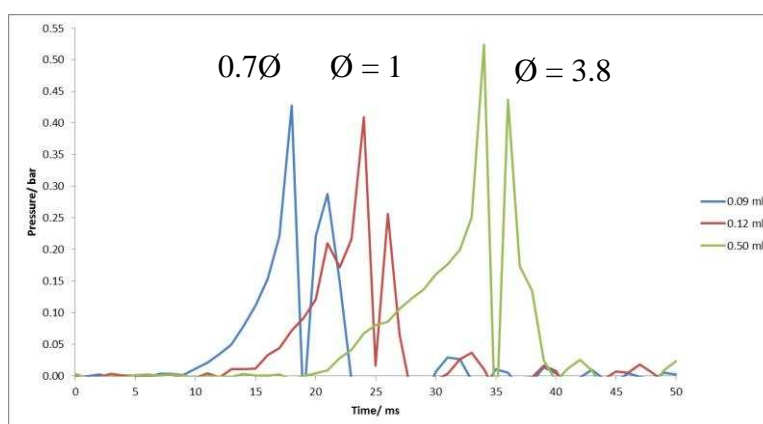

Figure 11. Pressure time records showing a double explosion with the second explosion occurring after the vent burst and more air entered the tube.

work. In Fig. 8 there is 200mb pressure rise above that for air only and would be greater if the vent had not burst. However, the most practical determination of flammability is that the vent bursts and this occurred in most and the magnitude of this is proportional diesel aerosol explosions. The rate of pressure rise just before the vent cover fails was measured to the mixture reactivity. The rate of pressure rise is proportional to the mass burning rate and most of the results will be shown in terms of this parameter as the reactivity term. Fig. 9 shows a typical vertical flame speed record for the time of flame arrival at the three thermocouples. Three repeat tests for the same concentration are shown with a flame speed of between 2.2 and 2.0 m/s. The peak rate of pressure rise and the flame speed are correlated flame reactivity terms, as shown in Fig. 10.

Figure 11 shows the pressure time records for lean, stoichiometric and rich mixtures. These all exhibit a double pressure peak and this has been seen with rich biomass dust explosions (Slatter et al., 2016). The first peak is the pressure rise from the initial flame propagation, and then the pressure falls as the vent bursts. Once the tube is open to external air the products of the initial combustion may include unburnt fuel and for rich mixtures the products will include CO and H_2, react with the air and there is a second flame propagation in the tube and a second pressure rise. The surprising

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feature was that this second explosion occurred for lean mixtures and indicates that the combustion efficiency was poor, leaving fuel unburnt to react with external air.

7.2 Lean mixtures reactivity and the lean flammability limit

Figure 12 shows the maximum rate of pressure rise just before the vent bursts as a function of the equivalence ratio, Ø. Lean mixtures were investigated and the lean limit was determined as 0.2Ø. This is leaner than a gas flame would give, which for hydrocarbons is usually 0.45Ø. This is very similar to dust explosions where literature values for several dusts have a lean limit of 0.2Ø (Andrews and Phylaktou, 2010). It is possible that the very lean limits are influenced by incomplete fuel and air mixing. The spray of diesel fuel from the central tube may not mix with all the air before it is ignited. This could give local rich mixture ignition in an overall lean mixture. This stratification of combustion is possible with the fuel injection method used. However, the agreement with 20L sphere and 1 m^3 MEC data for the Hartmann equipment (Andrews and Phylaktou, 2010), indicates that very lean flammability limits may be a feature of aerosol and dust explosions. Future work will investigate the use of an ignition delay between the aerosol injection and the spark occurring. The aim will be to give the aerosol more time to mix.

7.3 Repeat Tests after Several Months

It was found that the Hartmann equipment we were using, after a period of not testing, did not repeat the air injection process, as shown in Fig. 13. Why this occurred is not known, but the effect appeared to be a reduced storage volume of air. However, the Hartmann equipment still gave good explosions with diesel aerosols. The maximum rate of pressure rise just before the vent burst is shown in Fig. 14 and a lean limit of about 0.2Ø is found again. However, the magnitude of the maximum rate of pressure rise was much lower than for the results in Fig. 12 with the peak dP/dt reduced for stoichiometric mixtures from 400 in Fig. 12 to 150 bar/s in Fig. 14. However, these lower dp/dt for diesel aerosols are still much higher than for biomass dust explosions as shown in Fig. 15 for bagasse (Saeed et al., 2016b), where maximum rates of pressure rise are 30 for the finest particle size and as low as 5 bar/s for coarse bagasse.

7.4 Rich Mixtures and the Absence of a Rich Flammability Limit

Experiments with rich mixtures are shown as a function of the maximum rate of pressure rise in Fig. 16. In terms of the injected Ø the rich burning range was very wide and there appeared to be no rich limit. However, when the unburnt fuel deposited on the Hartman tube walls was corrected for the
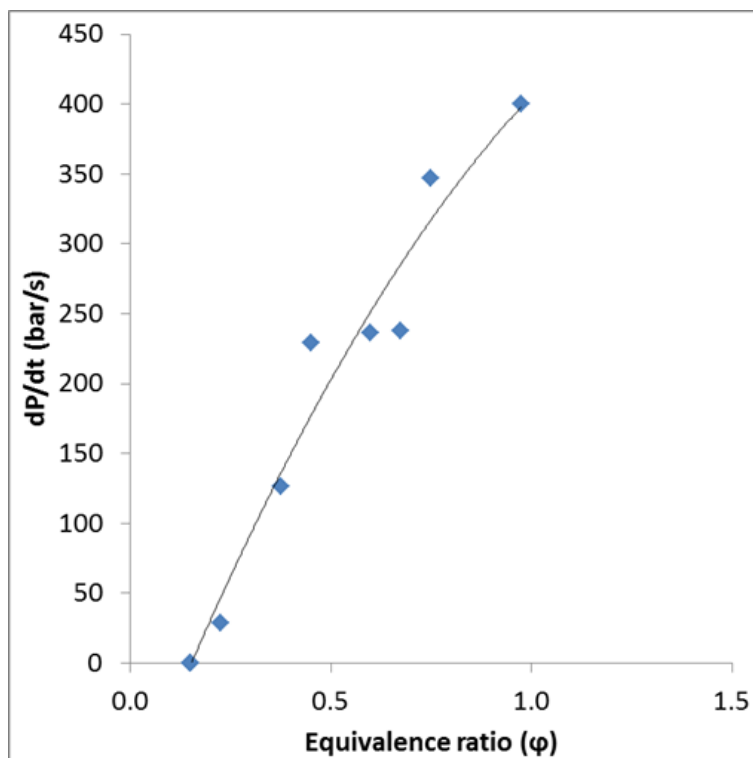
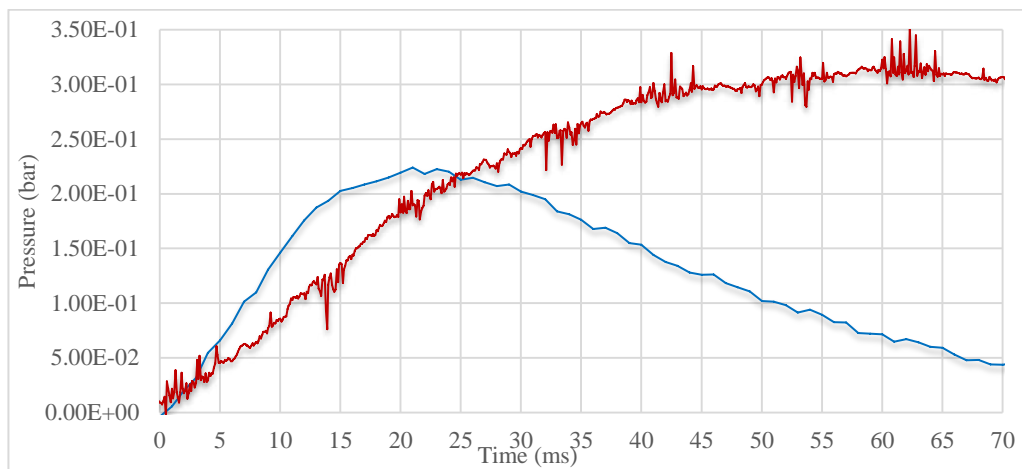Figure 12: Maximum rate of pressure rise just before the vent burst as a function of Ø

Figure 13: Repeat tests of the air injection system after a long break in testing.

corrected Ø only went up to Ø = 5 and the trend would be for a rich limit around Ø=6. About 50% of the fuel injected for rich mixtures was deposited on the Hartmann tube walls. If the corrected rich limit was in the range 5 - 6Ø, it would not be greatly different from the high MW gaseous hydrocarbons in Table 1 for the EU flammability method. More work is required with more liquid fuel injected to determine if there is a rich limit for diesel aerosols. For dusts there is the same problem as the literature shows no rich limits have been achieved (Eckhoff, 2003) and the results of Saeed et al (2016a,b, 2017) for biomass dust show that no rich limit had been found.

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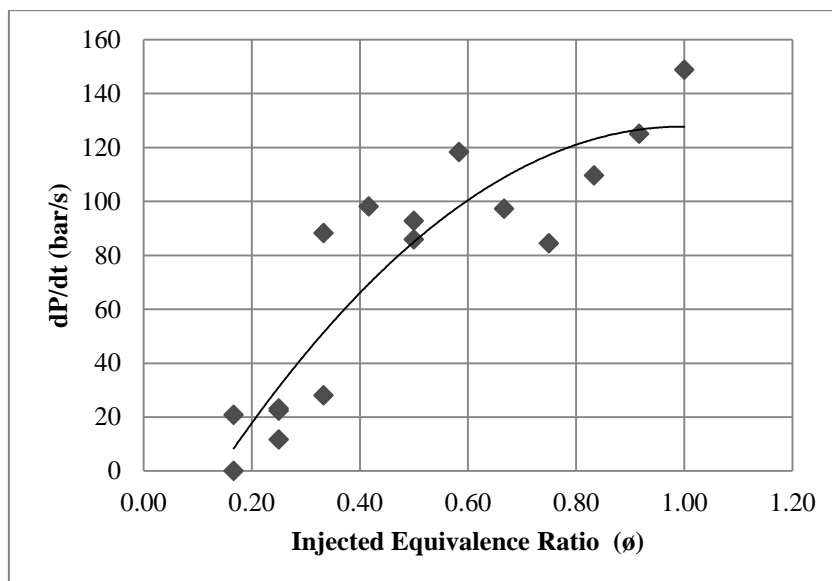XII ISHPMIE
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Figure 14: Repeat tests of the maximum rate of pressure rise just before the vent burst as a function of the equivalence ratio in the lean range.
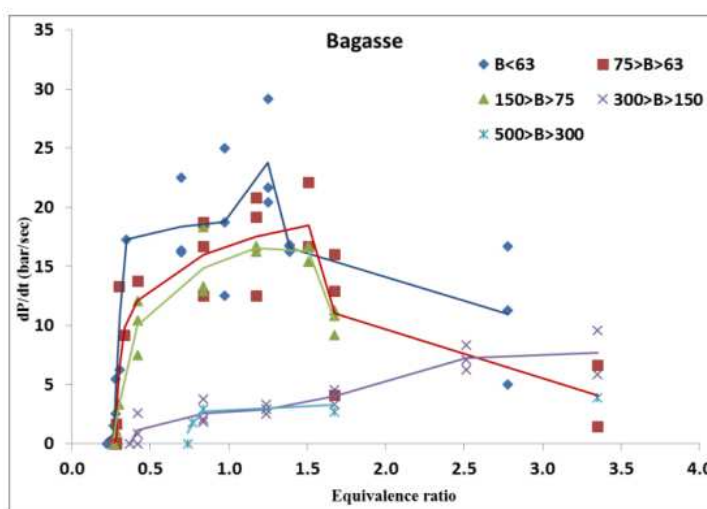

Figure 15: Typical results for the Hartmann with pulverised biomass, Bagasse, for various particle sizes, which shows much lower dp/dt than for aerosols in Fig. 14. (Saeed et al., 2016b)
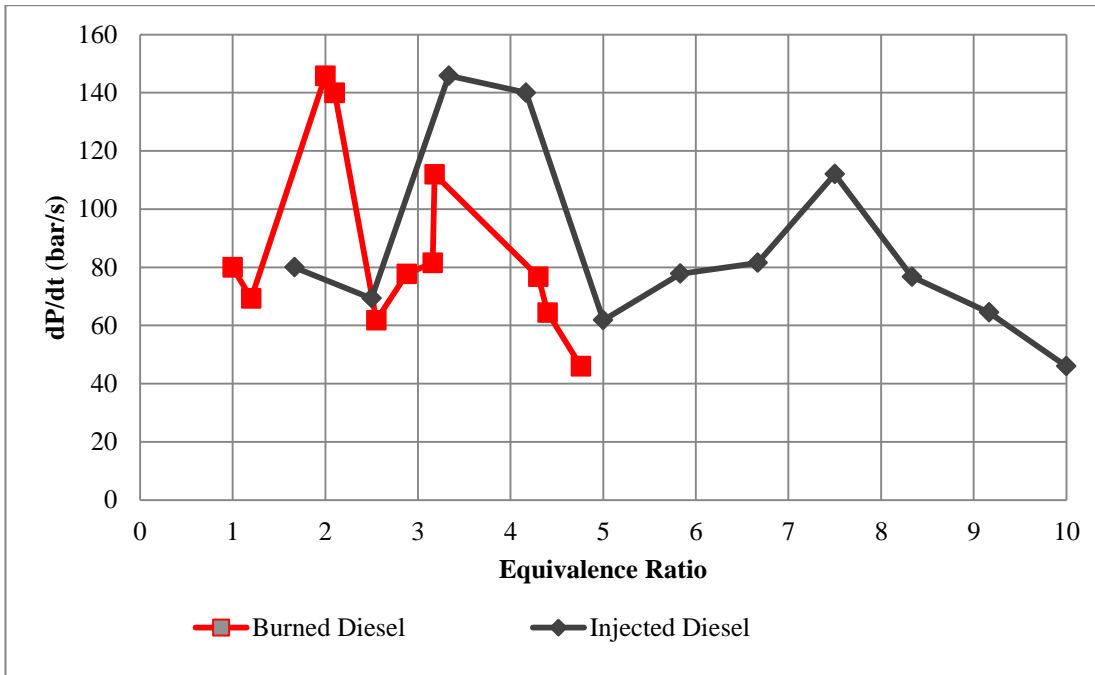
Figure 16: Maximum rates of pressure rise as a function of the rich equivalence ratios. Comparison of injected Ø and Ø corrected for tube wetting.

7.5 Repeatability of Aerosol Explosions

For an experimental method to be used as a reference method it has to be repeatable. For the reactivity in terms of the maximum pressure rise when the vent burst Fig. 17 shows the results of ten repeat tests for injected stoichiometric mixtures. Comparison is made with the air injection pressure and it is clear that one of the repeat stoichiometric explosions had no explosion. If this is excluded then the other nine repeats vary the peak pressure rise above the air only pressure from 0.2 to 0.38 bar with an average of 0.27, which is a variation of up to 40% from the mean. Some of this variation will come from variation in the burst pressure of the vent, as aluminium foil is not always consistent in its bursting pressure. Also some variation will come from the variation in the burnt equivalence ratio as only the injected equivalence ratio is controlled. However, most of this will come from variation in the atomisation process in the aerosol flame propagation rate. Fig. 11 shows that part of the variability is the time from the start of the air injection to the pressure rising and this is related to the difficulties of the initial flame propagation from the spark. This is also shown in Figs. 6 and 7, where the greatest time in the flame propagation is the time from the spark to the first visible propagating flame. The continuous spark in the Hartmann helps in this ignition process as the spark is always present and creates a hot ball of gas to vaporise the initial fuel droplets. It is the variability in this process that controls the variability in the final flame pressure rise. Fig. 18 shows three repeat flame speed measurements for injected stoichiometric mixtures, which have fairly repeatable flame speeds from 2.0 to 2.6. However, Fig. 18 shows a very large variation in the time to the start of flame propagation. Time zero in Fig. 18 is the time of initiating the flow of compressed air. Fig. 18 shows that the time from the spark to the flame to arrive at the first thermocouple 50mm above the spark is variable by 80ms, which is longer than the flame propagation time over the 150mm measurement length, which is 47, 45 and 42ms in Fig. 18. It is clear that diesel aerosol explosions with a 4J continuous spark struggle to develop a stable propagating flame and that the source of the variability is in this initial flame development period. The long development time also results in mixing of the aerosol continuing and the air jet turbulence decaying, both of which influence the subsequent flame speed and flammability.
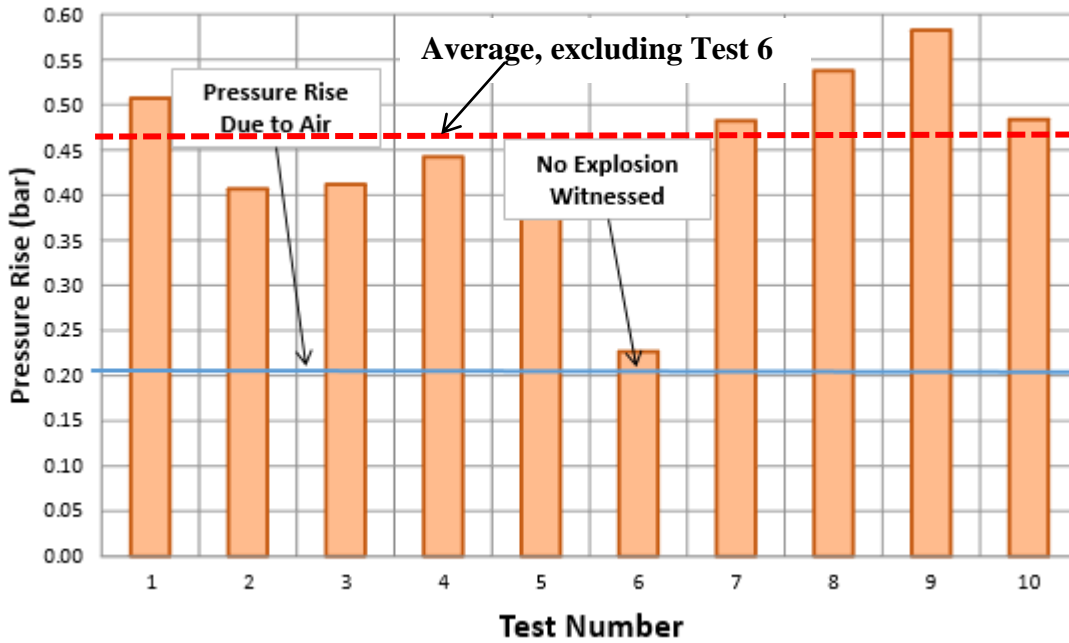
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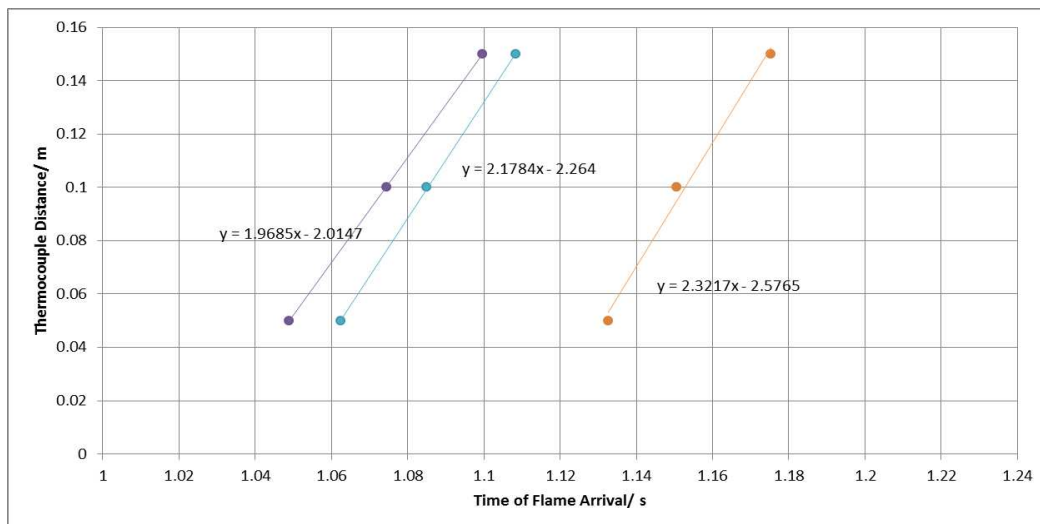Figure **17:** Stoichiometric Pressure Rise from Several Repeat Tests,

Figure 18: Flame speed measurements for 3 repeat stoichiometric diesel aerosol explosions.

7.6 Repeatability of the Lean Flammability Limit

The lean limits in Figs. 12 and 14 have only a few data points to support the lean limit of 0.2Ø and Fig. 14 indicates that it could be 0.17Ø. The present methodology does not allow a precise Ø to be set up as the syringe injector has to be weighed before and after the test to determine the amount injected, this is the injected Ø. However, for diesel some of the spray hits the wall and does not participate in the flame propagation. This was determined after the test, as discussed earlier. The Ø with this unburnt diesel correction is the corrected Ø. It is thus difficult to set the test Ø in advance and the only practical way the test Ø can be varied is to control the initial amount in the syringe injector and then correct the Ø after the test. 40 repeat tests were carried out around the lean limit from the initial tests in Figs. 12 and 14 and the results are shown in Fig. 18. The number for each bar on the probability plots in Fig. 18 show the number of tests that were at the corrected Ø. Unfortunately the distribution of Ø was wide and although 40 tests is many more than anyone else in
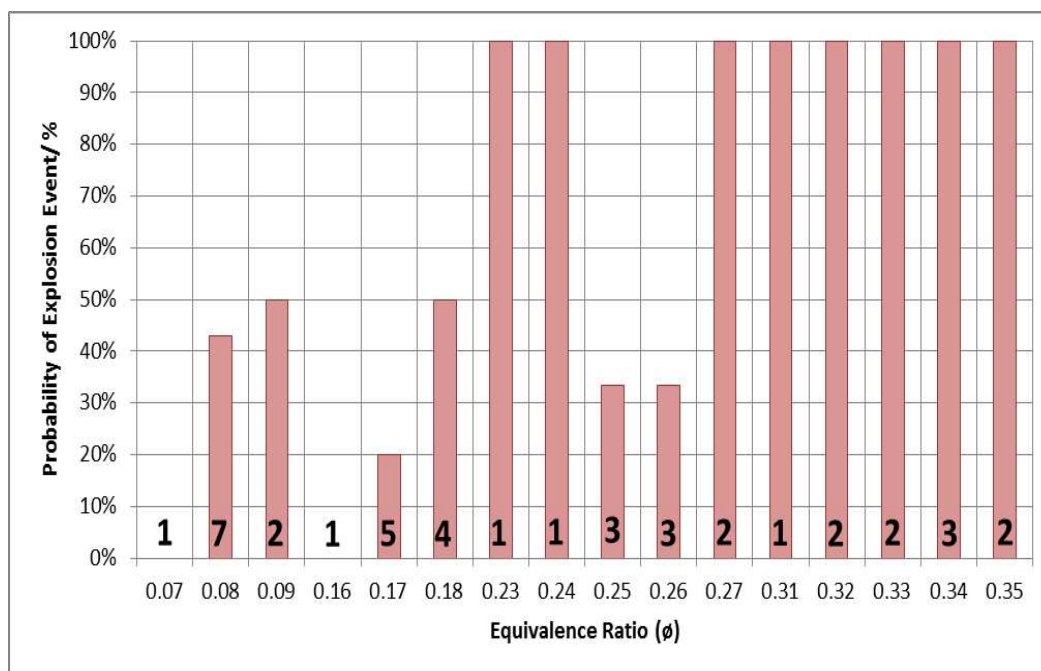
Figure 18: Explosion probability at various equivalence ratios close to the lean limit with the corresponding number of repeat tests at each equivalence ratio.
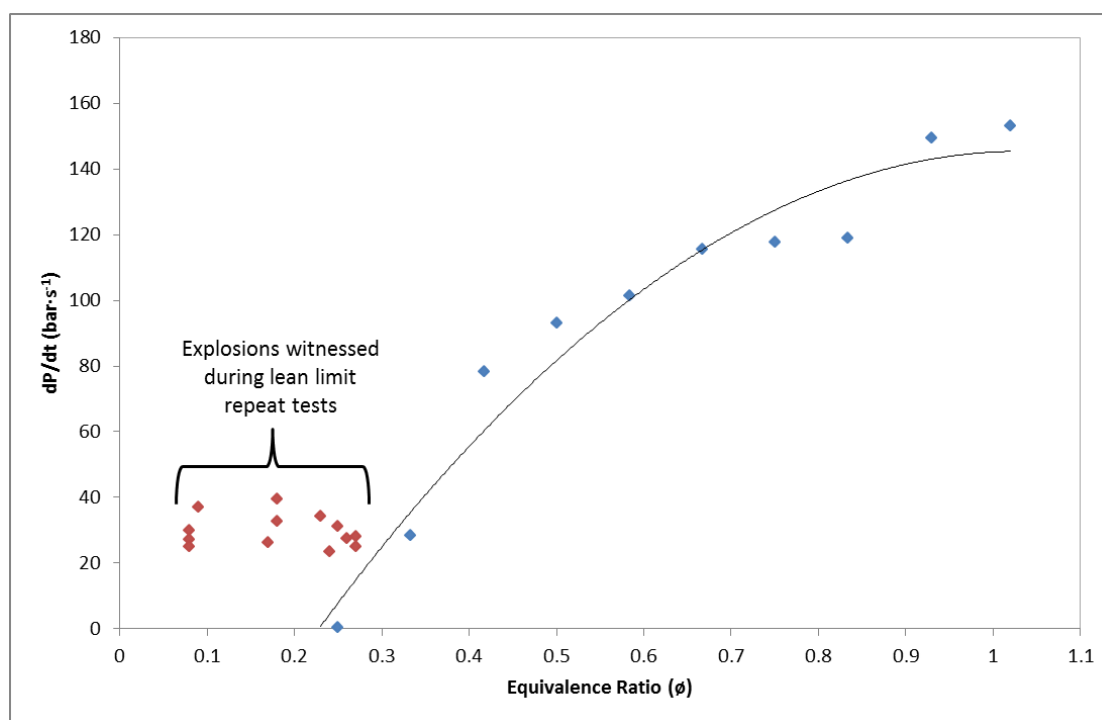
Figure 19: Rate of pressure rise just before the vent burst as a function of the corrected equivalence ratio (blue) with repeat tests (red) for mixtures outside the lean limit in the initial (blue) aerosol flammability investigations.

the literature has undertaken, it is clear that it is insufficient to be precise about the lean limit for diesel aerosols, as the number of repeats at the same Ø (to 0.01Ø resolution) was low. The maximum

number of repeat explosion with 7 repeats was at 0.08Ø, with 42% probability (i.e. 3 out of 7 explosions). Fig. 18 shows that the decision on a precise value for the lean limit is difficult and the flammability standards are of no help as they give minimal guidance.

Standards (BSI, 2003 and 2006) for gas and dust lean flammability limits refer to the limits as the mixture that does not burn, rather than the leanest mixture that does burn, which used to be in older standards. However, they do not detail how this is determined. In the gas and vapour standard (BSI 2003) for the EU the lean limit has to reproducible to the values in Table 5 to 10%, which for standard hydrocarbons is effectively +/- 0.05Ø if the lean limit is 0.5Ø. However, if the lean limit is 0.2Ø for aerosols or 0.1Ø for hydrogen on a mass basis then the accuracy required for 10% of the LFL is 0.02Ø for diesel aerosol and 0.01Ø for hydrogen. If the lean limit for a hydrocarbon gas is 0.45 then you would only have to test 0.4Ø and 0.5Ø either side this limit, so there is no regulatory reason to determine lean limits for gases and vapours to better than 10%, which is rather a lax standard. It is even worse for dusts as the mixtures to be tested are defined in the legislation (BSI 2006) as 500, 250, 125, 60, 30, 15, 7 g/m^3 which in terms of equivalence ratio for a hydrocarbon dust (80 g/m^3 for Ø=1) is a Ø of 6.3, 3.1, 1.6, 0.75, 0.38, 0.19 and 0.09 respectively. 30 and 60 g/m^3 are the most common MEC for dusts at hydrocarbon dust equivalence ratios of 0.75 and 0.38Ø. Clearly this is a worse resolution of the lean limit than for gases. It means that there is no resolution of the lean limit between 0.75 and 0.38Ø. It is simply stated in the legislation that if 60 g/m^3 explodes and 30 g/m^3 does not then the lean limit is 30 g/m^3 and there is no interest in the fact that the actual lean limit may be 50 g/m^3 (Saeed et al., 2016a). It is obviously a safe assumption for the MEC for dusts, but can lead to more expense in the dust protection equipment as more ventilation will be required to dust concentrations to be safe.

Huescar et al. (2013) introduced an improved method for dust MEC using the Hartmann explosion tube equipment, based on repeat tests and ignition probability with 10 repeats at each concentration. If a condition is that none of the 10 repeat tests explodes then very lean limits would result. It was proposed in this work at 50% probability should be the lean limit condition. The present work tried to apply this strategy to aerosol explosions, but the difficulty in setting a precise Ø made the application difficult, as shown in Fig. 18. If 50% probability of ignition is accepted than Fig. 18 would place the lean limit for diesel aerosols at Ø=0.18 or 0.27, as the 40 repeat tests had too great a variation in the corrected Ø to have sufficient repeats at each Ø. However, this is still a better resolution of the lean limit than is required for dust MEC evaluation. Fig. 19 shows the measured peak rates of pressure rise for mixtures between 0.08 – 0.27Ø. They show that all the mixtures had a significant rate of pressure rise, but that it was low. This indicates partial flame propagation, which returns to the definition of flammability. Is 100mm flame movement sufficient for aerosols, in this work Fig. 7 indicates that 50mm flame movement sometimes occurs, but sufficient pressure rise has occurred for this to be deemed flammable.

In spite of these concerns over the precise value of the lean flammability limit for diesel aerosols, it is clear that we have produced repeatable explosions that show the lean limit is 0.2Ø +/- ~0.1Ø. This is clearly leaner than for hydrocarbons other than ethylene and acetylene where the lean limit is 0.45 +/-0.05Ø in Table 5 with the stated 10% repeatability. This shows that diesel aerosol explosion lean flammability limit is leaner than for gaseous or vapour hydrocarbons. Why this occurs is for future research, but one mechanism is local mixture fluctuations along the flame front. A small droplet vaporises in the flame and the flame reaction zone only sees the vapour and not the droplet, if the drop size is below the critical for full evaporation in the flame front. However, transversely in the flame front the local mixture could be richer where the droplet was and leaner in the gap between droplets. This rich/lean local variation in the flame front could enhance the flame stability and extend the lean flammability limit.

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8. Conclusions

There are no standards for the determination of the flammability and reactivity of liquid hydrocarbon aerosols, in spite of the long standing legal requirement in the EU and USA for protection against aerosol explosion hazards.

The number of industrial incidents of aerosol explosions and the associated damage and lives lost, warrant the development of suitable test procedures for determining the flammability limits and reactivity of hydrocarbon aerosols. Such standard should apply to liquids with flash points above those in the current flammability standards with a maximum of $93^{\circ}C$. However, the standards should also apply for any liquid with a flashpoint above ambient temperature and the liquid used below its flashpoint, as they are all an aerosol explosion hazard.

There are similarities between dust and aerosol explosions, particularly the importance of the drop or particle size and this makes existing dust explosion standard test equipment a possible route for aerosol testing.

The ISO 1 m^3 dust explosion vessel with diesel liquid placed in the same location as for dusts and operated in the same way, could not be made to explode for two methods of fuel injection. The Hartmann equipment in its standard form could not be made to give an aerosol explosion. In all these cases the air injection system sprayed the liquid onto the vessel walls and did not form an aerosol that was flammable.

A modification to the Hartmann equipment that removed the dust disperser and injected the liquid fuel down the air delivery pipe gave strong diesel aerosol explosions at the first attempt and is recommended as a technique worth developing into a standard for aerosol explosion testing for diesel like liquids.

Experimental results using this modified Hartmann equipment showed with many repeat tests that the lean limit is 0.2Ø +/- ~0.1Ø. This is clearly leaner than for hydrocarbons other than ethylene and acetylene where the lean limit is 0.45 +/-0.05Ø by the EU standard tube method. This shows that diesel aerosol explosion lean flammability limit is leaner than for gaseous or vapour hydrocarbons and this should be taken into account in hazard evaluations. Also the use of the initial rate of pressure rise prior to the vent bursting in the Hartmann explosion tube, shows that the reactivity of diesel aerosols is substantially higher than for biomass dust explosions in the same equipment.

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