

Generation of Flammable Mists from High Flashpoint Fluids: Literature Review

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Abstract

Hazardous Area Classification (HAC) for explosive gas atmospheres is well established, with guidance published in various standards and industry codes of practice. However, the same situation is not currently the case for high flashpoint liquid releases that could give rise to an explosive mist atmosphere. There is a pressing need for clear guidance on mist hazards to allow operators to determine the extent of areas where flammable mists may be present and to select appropriate equipment for use in those areas.

This review paper summarises the findings of a recent literature survey on flammable mists and provides information that will be useful in developing a HAC methodology for explosive mist atmospheres. It focuses on the two fundamental issues: mist flammability and mist generation. The first of these is discussed with reference to five measurable parameters: the Lower Explosive Limit (LEL), Minimum Ignition Energy (MIE), Maximum Experimental Safe Gap (MESG), Minimum Igniting Current (MIC), and Minimum Hot Surface Ignition Temperature (MHSIT). Measurements of these quantities in mists are analysed and models for their prediction are discussed. The second issue of mist generation is examined in the context of mists produced by pressurised sprays and condensation aerosols. Tentative proposals are suggested for developing area classification guidance based on the prediction of the flammable cloud size.

The literature review and the findings emerging from it are currently being used to inform a major Joint Industry Project on mist explosion hazards. The objectives for this project are briefly described.

1. Introduction

The European ATEX 'Workplace' Directive (1999/92/EC), which is implemented in the UK by the DSEAR regulations, requires employers to identify and classify areas of the workplace where explosive atmospheres may occur. They must then ensure that appropriately certified equipment is used in the hazardous zones.

This process of Hazardous Area Classification (HAC) is well established for explosive *gas* atmospheres, with guidance published in various standards and industry codes of practice (BSI, 2010a; IGEM, 2010; EI, 2005). However, the same situation is not currently the case for explosive *mist* atmospheres, produced by releases of high flashpoint liquids. The most recent edition of the relevant standard, BS EN 60079-10-1 (BSI, 2009) contains a new annex which offers some qualitative guidance on mist explosion hazards, but there is little in the way of quantitative methods. The Energy Institute code of practice, IP15 (EI, 2005), also notes that "there is little knowledge on the formation of flammable mists and the appropriate extents of associated hazardous areas ... Further research is needed". Despite this fact, it is clear that

consequences of mist explosions can be very severe. A recent review of reported accidents (Santon, 2009) identified 37 incidents including 20 explosions, of which nine were collectively responsible for a total of 29 fatalities.

The aim of the present paper is to provide a survey the recent literature on flammable mists and summarise information useful for the development of an HAC methodology for explosive mist atmospheres. It is based on the substantial literature survey conducted for the Health and Safety Executive (HSE) recently by Gant (2011).

In the present work, the term “mist” is used to describe any airborne suspension of droplets produced by either pressurized atomisation or condensation of a saturated vapour (sometimes referred to as sprays or aerosols). The review only considers mists of liquids that are below their flashpoint at the ambient temperature. A list of such fluids is given below, indicating the range of materials (and industries) where area classification may be needed:

- | | |
|----------------------------------|-----------------------|
| · Lubricating oil | · Vegetable oil |
| · Hydraulic oil | · Light fuel oil |
| · Heavy fuel oil | · Heat transfer fluid |
| · Jet fuel/kerosene | · Transformer oils |
| · Process fluids (e.g. Solvesso) | · Diesel |
| · Bio-diesel | · White spirit |

Fluids outside the scope of this review include those that boil upon release to ambient pressure (e.g. propane) and low flashpoint liquids such as gasoline.

2. When is a Mist Flammable?

There are a number of potential physical mechanisms that could cause a mist to ignite. Sources of ignition include: heat, electrical, mechanical or chemical energy. For the design of equipment to be used in zoned areas, the main ignition sources are likely to be electrical sparks, hot surfaces, hot gases and flames. The discussion therefore focuses on the following relevant measurable quantities: the Lower Explosive Limit (LEL), Minimum Ignition Energy (MIE), Maximum Experimental Spark Gap (MESG), Minimum Igniting Current (MIC) and Minimum Hot Surface Ignition Temperature (MHSIT). A brief introduction to spark ignition of mists is first provided to put these quantities into context.

2.1 Spark Ignition and Flame Propagation

When a spark is generated between two electrodes submerged in a mist of fine droplets, the temperature rapidly increases within a small, roughly spherical, volume of gas and droplets, known as the spark kernel. Within this volume, any fuel droplets rapidly vaporise and the fuel vapour rapidly mixes with the surrounding air. Since the temperature is high, reaction rates tend to be extremely fast and the fuel vapour is almost instantly transformed into combustion products, releasing heat in the process.

If the rate of heat release due to reaction of the evaporated fuel droplets is higher than the rate of heat loss from the spark kernel to the surrounding unburnt gas, high temperatures will be sustained, the spark kernel will grow and a self-sustaining flame will start to propagate through the mist. If, on the other hand, the rate of heat release within the ignition kernel is less than the heat loss to the surroundings, the temperature within the kernel will fall, the rate of fuel evaporation will decrease and the spark kernel will eventually be extinguished (Ballal and Lefebvre, 1978).

2.2 Lower Explosive Limit (LEL)

When the mist droplets are very small (with diameter less than 10 μm), as the flame propagates through the mist, the droplets vaporise ahead of the flame front and the flame travels through essentially a vapour-air mixture. The LEL for the mist in this case is therefore similar to that of the corresponding vapour-air mixture (Burgoyne and Cohen, 1954; Zabetakis, 1965; Faeth and Olson, 1968).

For larger droplets, there is insufficient time for the droplets to vaporise completely before becoming engulfed in the advancing flame front. Each droplet then burns with its own diffusion flame, rather than as a homogeneous gas mixture. For droplets larger than around 40 μm , heat transfer from one burning droplet to its neighbours becomes the principal mechanism for flame propagation through the mist (Burgoyne and Cohen, 1954).

At the LEL, Burgoyne and Cohen (1954) found that the mean distance between droplet centres was around 22 times the droplet diameter for droplets smaller than 10 μm , and around 31 times the droplet diameter for droplets with diameter larger than 40 μm .

For mists with droplets larger than around 20 μm , the LEL is strongly affected by the method of measurement. Many studies have used flame tubes in which the mist is quiescent or moving very slowly when it is ignited. Usually in these experiments, the mist is ignited at the bottom of the tube, and the resulting flame propagates vertically upwards, e.g. Burgoyne and Richardson (1949), Burgoyne and Cohen (1954), Taylor (1957) and Danis *et al.* (1988). A few other measurements have been reported using different experimental arrangements, where the flow has been momentum-dominated and the mist has been produced by a spray, with a range of droplet sizes (Anson, 1953; Rao and Lefebvre, 1976). The measured LEL and MIE in these cases have been quite different from those recorded with nearly-quiescent mists.

In nearly quiescent mists with droplets larger than around 20 μm , the LEL is lower than in mists composed of smaller droplets or just vapour. The measurements of tetralin mists by Burgoyne (1963) showed that the LEL decreased from around 48 g/m^3 to around 5 g/m^3 as the droplet diameter was increased from around 10 μm to 150 μm (Figure 1). To put these concentrations into context, in atmospheric clouds or fog, the water droplet concentration varies between around 0.05 g/m^3 and 1.5 g/m^3 , with droplet diameters of between 1 μm and 30 μm (Middleton, 1952). Flammable mists are therefore likely to be optically dense, i.e. visibility will be severely limited.

Several theories have been put forward to explain the reduction in LEL with increasing droplet size. The most physically consistent appears to be that of Burgoyne (1957; 1963) who related the behaviour to the mist droplets falling downwards under gravity, i.e. the sedimentation effect. For an upward propagating flame, the presence of droplets falling vertically downwards leads to an increase in the relative speed between the flame front and the droplets. This has the effect of increasing the flow rate of fuel into the advancing flame front. Hence the static concentration of fuel can be reduced below the LEL, whilst maintaining the local LEL concentration at the rising flame front. This theory explains the reverse behaviour that is observed for downward propagating flames, where the presence of falling droplets reduces the flow rate of fuel into the advancing flame front and the LEL is increased, or in some cases downward flame propagation is not possible at all.

An additional effect of droplet size relates to the presence of the diffusion flame around individual droplets. As the droplets increase in size, their sedimentation speed increases and the elongated “tails” of the surrounding diffusion flame grow longer. This enhances radiative heat transfer between neighbouring droplets, which enhances flame propagation (Burgoyne, 1963).

Burgoyne (1963) developed a semi-empirical correlation to predict the LEL of monodisperse¹ quiescent tetralin mists, shown as the solid line in Figure 1. This related the static LEL concentration, C_s , to the “kinetic” concentration of mist at the moving flame front, C_k , the particle sedimentation speed, V_s , and the flame speed, V_f , as follows.

$$C_k = C_s \left(\frac{V_s + V_f}{V_f} \right) \quad (1)$$

where C_k was found to decrease linearly with increasing droplet size. Apart from this correlation, no general-purpose model appears to have been developed to predict the LEL of mists.

In momentum-dominated sprays, the LEL has been found to exhibit altogether different behaviour with increasing droplet size as compared to quiescent mists. In the experiments undertaken by Rao and Lefebvre (1976), a horizontally-directed kerosene spray was ignited in a co-flowing air stream moving at between 19 m/s and 50 m/s. The results showed that the LEL increased as the droplet size increased. Similar trends were obtained in kerosene spray experiments by Anson (1953), who measured the size of droplets that caused combustion to become unstable in a flame tube, where the flow speed was around 15 m/s and the flame was stabilized in the wake of a flat annular baffle. In these flows, the sedimentation speed may be small in comparison to the droplet velocity and there may be other effects related to droplet breakup, the entrainment of fresh air and turbulence.

2.3 Minimum Ignition Energy (MIE)

The MIE necessary to produce a self-propagating flame in a mist is affected by a number of factors. The most significant of these is the droplet size, but the fuel concentration, volatility, air velocity and the presence of any fuel vapour (in addition to the droplets) are also important. These factors were studied in detail by Ballal and Lefebvre (1978; 1979; 1981b) and Peters and Mellor (1980), who developed models for the ignition energy and quenching distance that were shown to be good agreement with experimental data for a range of mists and sprays of various substances.

For a given mist concentration (in terms of mass of liquid within a given volume of air), as the droplet size decreases, the effective surface area available for evaporation increases and therefore the MIE decreases. For monodisperse mists, the optimum droplet diameter which requires the minimum MIE appears to be between 10 μm and 30 μm (Singh and Polymeropoulos, 1985; Danis *et al.*, 1988). Within this size range, the droplets seem to be sufficiently large to still create some wrinkling of the flame front (and enhance combustion), whilst being sufficiently small to evaporate quickly. It has been thought that flame speeds in mists with the optimum droplet size may exceed those of the equivalent vapour mixture, for the same overall fuel concentration (Polymeropoulos and Das, 1975; Hayashi *et al.*, 1976; Ballal and Lefebvre, 1981a; Danis *et al.*, 1988; Bowen and Cameron, 1999). However, laboratory-scale experiments have not yet confirmed this (Lawes *et al.*, 2006). For further information, see Sulaiman and Lawes (2010) and Lawes and Saat (2011).

The ignition energy of mists decreases monotonically with increasing concentration, up to at least an equivalence ratio of 2.2 (Dietrich *et al.*, 1990). Results presented by Ballal and Lefebvre (1978; 1979; 1981b) and Danis *et al.* (1988) suggest that the MIE required to ignite

¹ *i.e. composed of droplets with uniform size*

a mist is probably higher than the MIE required to ignite a vapour-air mixture of the same substance at its optimum concentration for ignition. However the data are limited and the modelling results are not conclusive. The experiments conducted by Ballal and Lefebvre (1978; 1979; 1981b) were all limited to concentrations at or below stoichiometric, whilst those conducted by Danis *et al.* (1988) were for equivalence ratios up to 1.8 and droplet sizes greater than or equal to 30 μm .

For fuel concentrations close to stoichiometric, the ignition energy of a mist is usually greater than that of the equivalent vapour-air mixture, due to the need for the spark to first provide sufficient energy to vaporise the liquid droplets and then raise the vapour temperature sufficiently for ignition to occur. However, at much lower concentrations, approaching the vapour LEL, the ignition energy necessary to ignite a fuel mist can be lower than that required to ignite the equivalent vapour mixture (Danis *et al.*, 1988). This follows from the fact that mists composed of droplets larger than around 20 μm can be ignited at concentrations below the vapour LEL, whereas (by definition) vapour cannot be ignited at this concentration.

The presence of any vapour reduces the MIE of a mist (Ballal and Lefebvre, 1981b). As the fuel volatility is increased, the amount of energy required to vaporise the droplets is reduced and therefore the ignition requires less energy (Peters and Mellor, 1980). The results presented by Ballal and Lefebvre (1978; 1979; 1981b) and Peters and Mellor (1980) showed that the influence of the fuel volatility appeared to be well-approximated using the Spalding mass transfer number.

A unified set of models for the MIE of mists was presented by Ballal and Lefebvre (1981b) that accounted for fuel evaporation, reaction kinetics, fluid dynamics effects (including turbulence), different ambient pressures, fuel-air ratio, droplet size, the presence of polydisperse droplets, and fuel volatility. The results were shown to be in very good agreement with measurement data, although these were limited to concentrations at or below stoichiometric. Danis *et al.* (1988) subsequently showed that their models gave good predictions at concentrations up to an equivalence ratio of 1.8.

2.4 Maximum Experimental Safe Gap (MESG)

Flameproof electrical equipment is designed such that the enclosure is sufficiently strong to withstand the maximum overpressure from an internal explosion. Any holes, slits or joints in the enclosure walls are also sufficiently narrow that they will prevent transmission of a flame from the inside to the outside of the enclosure.

The key parameter which indicates whether or not a flame will be transmitted through the narrow gaps in the enclosure walls is the MESG. This is measured for gases according to the BS EN 60079-20 standard (BSI, 2010b) using apparatus consisting of two chambers, one inside the other, that are both filled with a mixture of flammable gas and air. The inner spherical chamber is linked to the outer chamber by a slot-shaped gap around its circumference, and the wall thickness around this gap is such that the path length between inner and outer chambers is 25 mm. The mixture is ignited in the inner chamber and the flame propagation into the outer chamber is assessed. When the gap width is sufficiently small, the flame is quenched as it passes from the inner to the outer chamber, and the flammable mixture in the outer chamber is not ignited. The MESG is defined as the maximum gap between the two parts of an explosion chamber that prevents ignition of the external gas mixture, for all concentrations of the tested substance in air.

For gases, the MESG and/or the Minimum Igniting Current (MIC) is used to classify the gas into one of four “gas groups” (I, IIA, IIB and IIC). Suitable flameproof equipment is then

selected for use in potentially flammable atmospheres on the basis of this gas group classification.

The only study which has aimed to measure the MESG of mists appears to be the work of Capp (1988), who used an 8 litre internal explosion sphere filled with a flammable mixture of propane and air, and a 90 litre outer cylindrical volume filled with mist. The mist was generated by an air-driven spray gun. Four different liquids were tested: butanol, kerosene, hexanol and ethylene glycol.

The MESG was found to be between 15% and 23% higher for mists of butanol, kerosene and hexanol, than the MESG of their equivalent vapours, which were measured at a higher temperature necessary to vaporise the liquid. The ethylene glycol could not be ignited at the highest available gap width of 2.6 mm.

One of the significant limitations of the study by Capp (1988) is that the mist occupied only the outer chamber of the measurement apparatus, whilst the initiating flammable substance inside the inner chamber was propane gas, not mist. The measurements therefore provided an indication of whether the propane-air flame was quenched as it passed through the narrow slot, or whether it had sufficient remaining energy to initiate combustion of the mist in the outer chamber.

In principle, it may be possible to gain further useful insight into the MESG of mists from measurements and mathematical models for the ignition behaviour of droplets in a high-temperature gas environment. There is a significant body of literature that examines this scenario, due to its importance in internal combustion engines and gas turbines. Reviews are provided by, for example, Law (1982), Babrauskas (2003), and Sirignano (2005). However, no attempt appears to have been made yet to predict the MESG directly.

2.5 Minimum Igniting Current (MIC)

Low voltage equipment for use in flammable atmospheres is selected on the basis of “intrinsic safety” according to the Minimum Igniting Current (MIC), rather than the Minimum Ignition Energy (MIE). The MIE is measured by applying a sufficiently large voltage between two electrodes held a fixed distance apart, whereas the MIC is measured using a break-spark apparatus in which ignition is produced by mechanically breaking current-carrying wires. If the same classification system for the selection of intrinsically safe equipment was to be used for mists as it is currently for gases, this means that the MIE measurements for mists discussed above are not directly applicable to the choice of equipment.

Unfortunately, there appear to be no measurements of the MIC of flammable mists reported in the literature. However, the MIE, MIC and MESG for flammable vapours appear to be correlated with one another, as shown in Figures 2 and 3. It is unclear whether the MIE, MIC and MESG would be similarly correlated for flammable mists. Some factors affecting the MIE are known to be different from those affecting the MIC. For example, Zborovszky (1976) showed that the fuel concentration had relatively little effect on the MIC provided that fuel concentration was in the flammable range, whereas the MIE is known to be strongly affected by the fuel concentration.

In terms of selecting electrical equipment for use in explosive mist atmospheres, it would be useful in further work to establish whether, for instance, the MIC of a mist is greater than the MIC of its equivalent vapour. Intrinsically safe equipment that is rated for a flammable gas atmosphere of a particular substance might then be used safely in a flammable mist of the same substance.

2.6 Minimum Hot Surface Ignition Temperature (MHSIT)

To ignite a spray or mist using a hot surface, it is necessary for the hot surface to first vaporise a sufficient quantity of fuel to produce flammable concentrations of vapour and, secondly, for the temperature of the flammable vapour to be sustained for a period longer than the chemical ignition delay time. The effectiveness of hot surface ignition therefore depends on many factors, including the physical properties of the liquid, the concentration of fuel in the air, the droplet size, and the shape and extent of the heated surface. Ignition becomes more likely as the temperature or the extent of the hot surface is increased. The contact time and the concentration and temperature gradients near the surface, which are affected by the local ventilation flow, are also important parameters. The type of material used to make the surface and any surface treatments can have an effect, due to the formation of either film or nucleate boiling, and its effect on heat transfer rates. Some materials may also produce catalytic reactions or form an insulating oxidation layer, which can also affect the heat transfer rates. Reviews of the physical mechanisms involved in hot surface ignition are provided by Bennett (2001) and Babrauskas (2003).

One of the earliest comprehensive studies of lubricating oil mist ignition by hot surfaces was conducted by Freeston *et al.* (1956), for application to crankcase explosions. Small-scale tests were conducted using a heated ignition tube, 305 mm long and 38 mm in diameter, through which known concentrations of oil mist were passed. The mists were generated by condensation of a saturated oil vapour, and progressively leaner or richer mixtures were tested to establish the flammability limits. Various different metals were used for the ignition tube, to match those materials commonly found in marine diesel engines. Using this apparatus, Freeston *et al.* (1956) showed that there were two separate temperature regions in which hot surface ignition of oil mists was possible: a low-temperature region from 270 °C to 350 °C, which was thought to be related to the occurrence of cool flames, and a high temperature region which extended upwards from 400 °C. In between these two temperature regions, ignition was not possible. The oil tested was a typical engine oil of SAE 30 viscosity and it was considered that similar results would be obtained with any other engine oil. No measurements were made of the mist droplet size, although it is likely to have been relatively small, since it was formed by condensation.

Further full-scale tests were reported by Freeston *et al.* (1956) which involved the ignition of a flammable mist within the crankcase of a 1,100 h.p. Sulzer diesel engine. The force of the resulting explosion was sufficient to damage the building in which the engine was housed. The tests showed that explosion venting of marine diesels was in itself insufficient to limit the maximum overpressures, and that some measures to prevent a flame from spreading throughout the crankcase were necessary.

A wide-ranging review of the ignition of droplets, sprays and spills by hot surfaces was undertaken by Babrauskas (2003), which considered a variety of fuels (lubricating oils, gasoline, kerosene, hydraulic oils etc.) and both open and semi-confined geometries. It found that there was considerable scatter in the measured MHSIT for the same fuel, depending upon the particular experimental arrangement. For unconfined flat plate surfaces, ignition was found to require surface temperatures more than 200 °C higher than the AIT. As the geometry became progressively more confined, the MHSIT decreased until it was equal to the AIT in the fully-confined case.

Babrauskas (2003) offered practical guidance on the MHSIT, whereby if the space was approximated as a cube and the hot surfaces occupied more than one face of the cube, the substance should be assumed to ignite at the AIT. For unconfined release scenarios with just one heated surface, provided that the substance was not prone to cool-flame ignition, Babrauskas (2003) suggested following the American Petroleum Institute guidance (API,

1991) which states that hot surfaces are liable to ignite fuel vapour if the surface temperature exceeds the AIT by 200 °C.

Further examination of the MHSIT of sprays was undertaken by Hawksworth *et al.* (2001) and Jagger *et al.* (2003). In the latter study, tests were carried out using a large stainless steel heated plate positioned vertically either perpendicular or at an angle to the spray axis. Mineral oil with an AIT of 341 °C was found to ignite in 5 out of 7 tests with a plate temperature of only 400 °C. For a polyol ester with an AIT of 400 °C, the measured MHSIT was 490 °C. There was no observed dependence on the angle of the spray impact. The results from the tests are summarised in Table 1 and they demonstrate clearly that the API guidance (API, 1991; 2003) is not sufficiently conservative. Instead, the AIT appears to provide a more suitable, conservative estimate of the MHSIT for sprays or liquid streams.

Although not related directly to MHSIT, it was demonstrated by Jagger *et al.* (2003) that the temperature at which a substance can ignite may be considerably lower than the AIT if the liquid is soaked into porous insulating material, such as pipe lagging. This phenomenon was examined by soaking a small cube of mineral silicate insulation in the test fluid and placing it inside a supporting stainless steel gauze container, which was then put into an oven. The oven temperature was increased in stages from 110 °C to 210 °C and self-heating was judged to occur if the sample temperature exceeded that of the oven by 60 °C. The onset temperatures for runaway self-heating are given in Table 1. For most of the hydraulic fluids tested, the runaway temperature was several hundred degrees Celsius lower than the AIT. Whilst the AIT appears to provide a conservative upper limit for surface temperatures to avoid hot surface ignition of sprays or mists, this work indicated that if the liquid released happened to soak into insulation it could ignite at lower temperatures, confirming the phenomenon which has often been observed in practice. Practical guidance on measures to reduce the likelihood of lagging fires, such as use of closed cell insulation near leak points, was provided by Fuhr (1992).

Various studies have shown that the flashpoint does not provide a useful measure of the MHSIT. In the work of Yuan (2006), the MHSIT was found to increase as the flashpoint of the material was increased, whilst in one of the studies reviewed by Babrauskas (2003), the reverse trend was produced. Hawksworth *et al.* (2001) also found that the MHSIT either increased or decreased with the flashpoint, depending upon whether the hot surface was vertical or horizontal. Babrauskas (2003) suggested that the reason for this behaviour was that the MHSIT is a function of both the volatility and reactivity. For unconfined releases, both of these factors affect the MHSIT in roughly equal proportions, whilst in confined releases the MHSIT is dominated by the reactivity of the substance. The flashpoint, on the other hand, is primarily a function of the volatility of the substance, and to a lesser extent the reactivity.

Burgoyne (1957) noted that for spark ignition in the vicinity of hot surfaces, prolonged contact between mists of liquid hydrocarbons and hot surfaces could result in cracking and the formation of vapours which do not share the combustion properties of the original hydrocarbons. The generation of hydrogen or acetylene, for instance, could significantly lower the flammability limits. It was noted that this would need to be taken into account if the mist explosion hazard in a hot environment was to be controlled using either inert gases or suitably-rated flameproof or intrinsically-safe equipment.

Guidance on the ignition of mists by hot surfaces is provided in the Code of Practice for Atmospheric Oil Mist Detectors, issued by the International Maritime Organisation (IMO, 2003). This notes that temperatures as low as 150 °C may be sufficient to cause ignition.

3. How is a Flammable Mist Produced?

3.1 Pressurized Sprays

In terms of practical guidance for HAC of mists, it would be beneficial to define an operating pressure below which a liquid release into an unconfined space would not give rise to a mist. This may enable certain low-pressure systems to be classified as non-hazardous. To investigate this matter requires many factors to be taken into account, but some insight can be obtained from simple empirical correlations that determine when atomization takes place, based on the reservoir pressure, orifice diameter and liquid properties.

Relevant analysis can be found in the early work of Ohnesorge (1936), who characterised the behaviour of liquid jets using the liquid Reynolds number, Re_l :

$$Re_l = \frac{\rho_l U_l D}{\mu_l} \quad (1)$$

and the Ohnesorge number², Oh :

$$Oh = \frac{\mu_l}{\sqrt{\rho_l D \sigma}} \quad (2)$$

where μ_l and ρ_l are the dynamic viscosity and density of the droplet, respectively, U_l is the liquid velocity, D is the orifice diameter and σ is the surface tension.

The transition to atomization breakup was determined by Ohnesorge (1936) to take place at Ohnesorge numbers given by:

$$Oh = 745 Re_l^{-1.22} \quad (3)$$

and, subsequently by Miesse (1955):

$$Oh = 100 Re_l^{-0.92} \quad (4)$$

Using either the Ohnesorge (1936) and Miesse (1955) correlations, and Bernoulli's equation for the exit velocity:

$$\Delta P = \frac{1}{2} \rho U_l^2 \quad (5)$$

the pressure differential (ΔP) needed to produce atomization breakup can be determined as a function of hole size for a range of substances. Care should be taken in extrapolating the data beyond the range of conditions used to derive the correlations, i.e. diameters smaller than 0.5 mm or orifices much larger than 4 mm. It should also be noted that these correlations only quantify the onset atomization of the liquid jet at the orifice. Mists may still be generated at lower pressures due to secondary aerodynamic breakup or impingement of the jet on nearby surfaces. In fact, in some cases impingement may provide the dominant mechanism for spray breakup. An alternative method for characterising the onset of atomization, based on the Weber number, We , was presented in the review by Birouk and Lekic (2009).

For high velocity jets, primary breakup is controlled principally by the action of the surrounding air on the liquid stream. A secondary factor is the presence of turbulence in the liquid stream issuing from the orifice. The presence of turbulence disturbs the surface of the

² Also referred to as the Laplace number or 'Z' number.

liquid jet, making it more susceptible to aerodynamic breakup. Primary breakup is therefore enhanced by either increasing the liquid velocity or decreasing the liquid viscosity. The atomization process is always accelerated by an increase in air resistance, hence an increase in air density enhances the breakup process. These factors are examined in more detail in the reviews of Lefebvre (1989) and Birouk and Lekic (2009).

Following the primary atomization process, large liquid droplets or filaments may fragment into smaller droplets due to secondary aerodynamically-driven breakup. Pilch and Erdman (1987) identified six different mechanisms involved in secondary breakup of spray droplets, each of which was characteristic of a particular Weber number range. Below a critical Weber number, We_c , the droplet size remains stable and further breakup does not take place. An empirical correlation describing how We_c varies as a function of Oh was developed by Brodkey (1969). However, this requires knowledge of the slip velocity between the droplet and the surrounding air.

The resulting average droplet sizes from pressurized spray releases tend to be larger than those produced by condensation aerosols, and the sprays are usually polydisperse (i.e. featuring a range of droplet sizes). Smaller droplets are produced by increasing the liquid density, reducing the viscosity or reducing the surface tension. Increasing the reservoir pressure also usually produces smaller droplets.

The size of droplets and the way in which the spray spreads is strongly affected by the nozzle shape. There have been many measurements of the droplet size spectrums produced by different sprays, see for example Pimentel (2006). However, empirical correlations for the size spectrum are mostly only valid for a particular nozzle and set of operating conditions.

To predict the volume of flammable mist produced by a pressurized liquid release there are currently two types of approach available. Firstly, Computational Fluid Dynamics (CFD) models that compute the flow in both the air and the liquid phases and can predict or prescribe primary and secondary breakup (e.g. Trinh *et al.*, 2007). These tend to be complex and costly, and therefore are not used routinely for industrial risk analysis. Secondly, there are engineering models that are based more heavily on empiricism or on simplifications such as the assumption of Locally Homogenous Flow (LHF). In principle, it may be possible to use these latter models for the purposes of HAC. They include methods used to quantify droplet sizes, spray cone angles, jet penetration and impingement (Simmons, 1977; Lefebvre, 1989; Ruff *et al.*, 1990; Pimentel, 2006; Mather and Lines, 1999; Abramovich, 1963; Ghosh *et al.*, 1991). A review of these and other models is provided in the report by Gant (2011).

3.2 Condensation Aerosols

Condensation aerosols are produced when the concentration of vapour rises above the saturation vapour pressure. This can occur when vapour at high temperature is mixed with cold air, for example, in the plume of the visible “steam” produced from a boiling kettle. It is not necessary to raise the temperature of liquid above its boiling point in order to produce this effect.

In the context of industrial hazards, condensation aerosols are known to be partly responsible for production of flammable mists in crankcases of marine diesel engines (Freeston *et al.*, 1956). Here, an overheated surface of a malfunctioning bearing, for example, vaporizes lubrication oil that subsequently condenses as it passes into cooler regions of the crankcase. The source of ignition is often the same heated surface that produced the vapour. In principle, similar behaviour could be produced if a combustible liquid was spilled onto a hot surface, such as an engine exhaust manifold.

Droplets produced from condensation aerosols are usually small in size ($< 50 \mu\text{m}$) and the mist is relatively monodisperse. For this reason, this method of mist formation is frequently used in fundamental studies of mist flammability, e.g. Burgoyne and Cohen (1954), Hayashi *et al.* (1974; 1976), Cameron and Bowen (2001).

Models do not appear to have been developed to predict the production rate of condensation aerosols, from a given liquid vaporization rate and ventilation rate, although in principle it should be reasonably straightforward to do so for a single-component substance, such as decane. For multi-component liquids (e.g. diesel), a model would be more complex to develop, although useful guidance may be found in previous work on multi-component spray modelling, for example, that of Muralidhar *et al.* (1995a; 1995b).

4. Future Area Classification Guidance

4.1 Selection of Equipment

To select the appropriate equipment for use in zoned areas, consideration needs to be given to the MIE, MESG, MIC, MHSIT of the mist. In general, measurements reported in the literature indicate that mists are more difficult to ignite than their equivalent vapours. The MIE, MESG and MHSIT all appear to be of similar magnitude or higher for a mist as compared to the vapour. Equipment for use in a mist atmosphere could therefore be selected based on its rating for the equivalent vapour atmosphere, in terms of gas group and temperature class. Further analysis and measurements are required to confirm that this is a safe approach, as the data is relatively limited. For multi-component substances (e.g. diesel), the matter is complex since lighter fractions evaporate preferentially in a spray, producing airborne droplets with a different composition to that of the liquid in the pressurized reservoir. A conservative approach may be to base the selection of equipment on the most reactive component present in a significant proportion in the liquid (i.e. the component with the lowest MIE, MIC and MESG). Consideration also needs to be given to environments where there is the potential for prolonged contact between mists and hot surfaces, which could result in cracking of liquid hydrocarbons and the formation of more reactive vapours.

4.2 Definition of the Flammable Cloud Extent

The value of the mist LEL is important since the area classification would use this to determine the extent of the flammable cloud. In nearly-quiescent mists, the LEL decreases as the droplet size is increased, due to the sedimentation effect. A plausible “worst-case” scenario would involve a spray directed onto the ceiling of an enclosure, producing a continuous widespread mist of falling droplets in the size range of $100 \mu\text{m}$ to $150 \mu\text{m}$. The LEL in this case has been found experimentally to be around 10% of the equivalent vapour LEL. Taking half-LEL to define the extent of the hazardous area would then be equivalent to 5% of the vapour LEL.

Such a low value of the mist LEL may be considered to be overly conservative in some cases. For example, if the source of ignition was located on the ceiling of an enclosure, the effect of droplet sedimentation may be limited. The mist droplets would not be falling downwards from any height in these circumstances, and it might be considered that the mist LEL should therefore be considerably higher than 5% of the vapour LEL.

It is also uncertain whether or not the sole reason for the low values of the LEL measured in flame tube experiments is the speed at which the droplets are falling relative to the air. If this were the case, then a spray could potentially also drive droplets into a flame at a similar rate to the droplets falling in a flame tube, and lead to a similar reduction in the LEL. Similar

considerations may also apply if the ignition source was situated in a location where the ventilation flow projected a high mass flux of droplets against the ignition source, such as in the ventilation outlet of a gas turbine enclosure. One of the arguments against this hypothesis is that the measured LEL in high-momentum sprays has been found to be higher than the vapour LEL. However, these findings are based mainly on data from one study by Anson (1953) and are complicated by polydisperse nature of the spray in that case.

Another consideration in the flame tube experiments is that the buoyancy-driven flame travels vertically up the tube in these tests, which increases the relative speed between the droplets and the flame. It is unclear whether this motion of the flame is necessary to enhance the fuel concentrations at the flame front and lower the LEL. This subject merits further investigation, in order to identify the dominant physical mechanisms responsible for the reduction in the LEL.

Mists produced by condensation aerosols tend to have droplet sizes smaller than 50 μm . If the only means of generating a mist is from a condensation aerosol, rather than a spray, it may be appropriate to use a higher level to define the flammable cloud extent than 5% of the equivalent vapour LEL.

To ignite mists composed of large droplets requires an energetic ignition source, such as a naked flame. In environments where only relatively low-energy ignition sources are present (i.e. those produced by small electrical sparks), it may therefore be appropriate to use a higher LEL for the cloud extent. Further work is necessary to determine an appropriate safe level. In environments where flames are present (such as rooms containing furnaces or boilers) or where hot surfaces could produce a strong ignition source, it might be appropriate to assume 5% of the equivalent vapour LEL for the HAC methodology, but this needs to be confirmed by further analysis.

4.3 Unconfined Spray Releases

Where there is the possibility to generate a mist from a pressurized spray release, an initial calculation could be performed to assess whether the operating pressure is sufficient to cause liquid atomization. At pressures below a certain level, for a given hole size, it may be appropriate to assume that no significant mists will be produced. This is conditional upon the liquid being released into an unconfined space where it is unlikely to impinge on nearby surfaces and break up.

For pressures above this threshold, the maximum extent of the hazardous zone could potentially be obtained from a simple ballistic trajectory model. This model neglects aerodynamic effects, droplet breakup and droplet evaporation, and is therefore likely to be highly conservative but it may nevertheless be useful for an initial scoping study. The accuracy of this approach could be assessed by comparing predictions to experimental data, such as the recent measurements of Witlox *et al.* (2011) and Bettis and Jagger (2011). If the distances predicted by these simple equations are not supported by the experimental studies, it may be necessary to use a more sophisticated model. Ballistic models do not account for mist formation when spray droplets impinge on nearby solid surfaces, and the subsequent dispersion of the mist as a more passive cloud. Theoretical studies of the sedimentation speed and wind advection speeds, based on a post-impingement droplet size spectrum and initial cloud height, could be used to investigate this further. The results could also be confirmed with some experiments.

The flammable cloud volume for unimpeded spray releases in the open air could be determined using the LEL, as defined above, by making various assumptions about the orifice characteristics, breakup behaviour and spray entrainment. This would be particularly useful in

order to determine a limiting release size, below which the flammable cloud would be so small that its ignition would not pose a significant hazard. For secondary releases (i.e. those not anticipated to occur during the course of normal plant operation), it could result in an area being classified as non-hazardous. This approach is consistent with the concept of “negligible extent”, described in existing area classification standards for flammable gas releases (BSI, 2009). To develop this model, rather than simply assume a particular type of spray breakup behaviour based on a single set of experiments, a sensitivity study could be undertaken that considered a range of different empirical correlations previously developed for different types of nozzles, to account for the possible range in release conditions. This could include, for instance, correlations for both circular orifices and flat-fan types of nozzle.

4.4 Confined Spray Releases in Ventilated Enclosures

For spray releases in ventilated enclosures, the same approach described above could be used to determine the rate at which a mist is produced. The flammable cloud volume could then be determined using a similar method to that recommended by Webber *et al.* (2011) for flammable gas leaks in enclosures, in which it is related to the ventilation rate and size of the enclosure. As noted by Webber *et al.* (2011), it may also be necessary to develop rules of thumb to cope with confined spaces around potential leak sources, where sprays impinging on nearby surfaces combined with poor local-ventilation could result in the build up of significant mist volumes.

4.5 Joint Industry Project (JIP) on Mist Explosion Hazards

On 5 December 2011 a start-up meeting was held for the Mist Explosion Hazards Joint Industry Project (JIP), which is being coordinated by HSL. The project is sponsored by 15 companies and organisations, including HSE, with a total budget of £450k. The objective is to develop practical criteria that define the likelihood of flammable mist formation, which can be used as part of an area classification exercise. The scope of work therefore includes studying the formation of flammable mists, area classification zone and extent, mitigation measures, protected equipment concepts and equipment selection. In the present early stages of the project, work is being undertaken to classify the range of substances into categories according to both their propensity to form mists and their flammability. Further information on the scope of work and details of how to join the JIP are available on request.

5. Conclusions

This literature review has provided a summary of information needed for development of HAC guidance for mist explosion hazards. It has focused on two key aspects: mist flammability and the generation of mists. For the first issue of flammability, the properties of mists has been discussed with reference to five quantities: the LEL, MIE, MESG, MIC, and MHSIT. The second main issue of mist generation has been examined for mists produced by pressurised sprays and condensation aerosols. Tentative proposals have been suggested for developing HAC guidance based on the prediction of the flammable cloud size in unconfined releases and those in ventilated enclosures.

The principal aim of the present work has been to review the existing literature. Although tentative proposals for the development of an area classification methodology for mists have been put forward, these have not yet been scrutinised rigorously. On closer examination, some of the concepts may be found to be impractical or provide results that are unsustainably under- or over-conservative. These matters will be explored in the ongoing JIP on mist explosion hazards.

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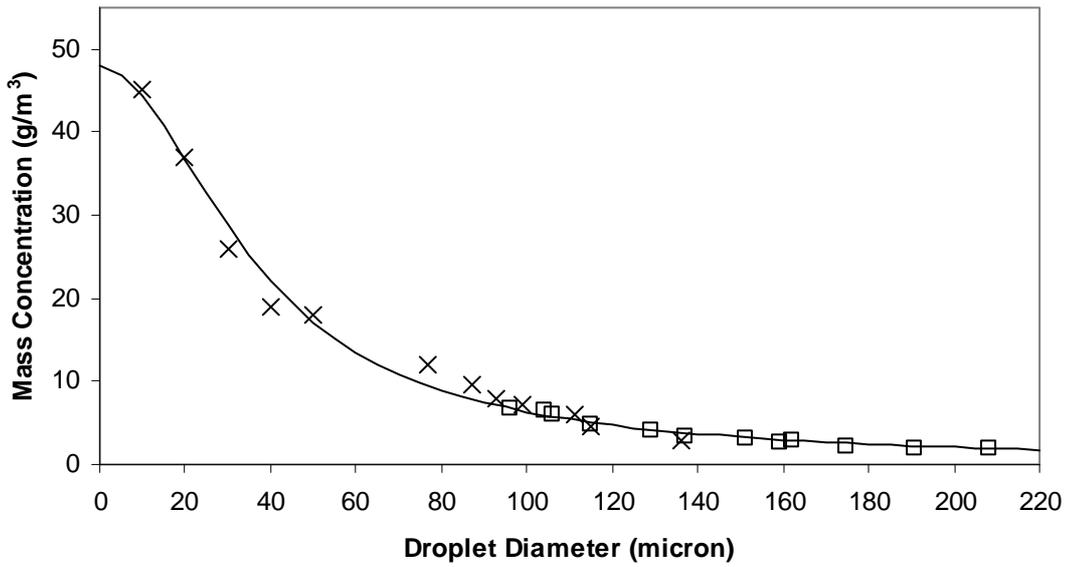


Figure 1 LEL for monodisperse mists for tetralin from Burgoyne (1963): × measured, □ calculated, — empirical model result.

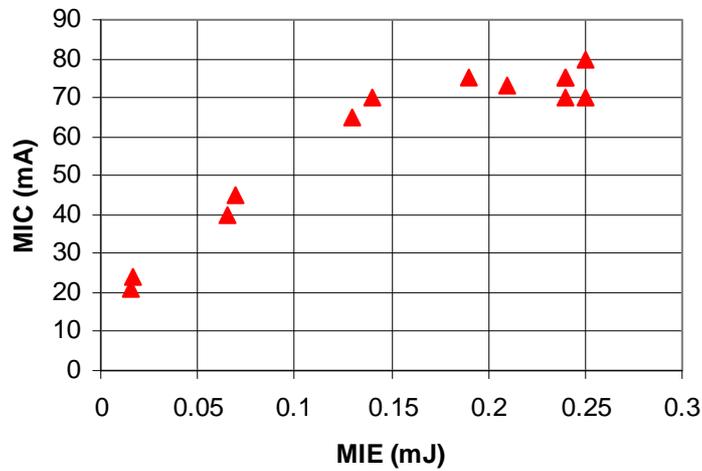


Figure 2 MIE versus MIC for various flammable vapours

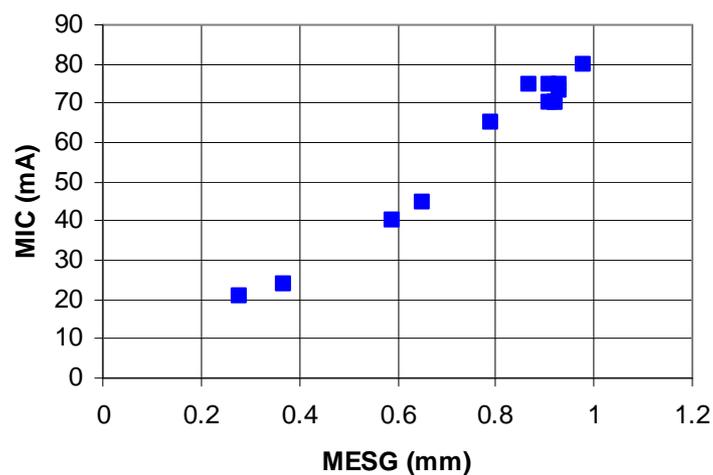


Figure 3 MESG versus MIC for various flammable vapours

Table 1 Hot manifold ignition test results, temperatures for runaway oxidative self-heating and auto-ignition temperatures, from Jagger *et al.* (2003)

<i>Fluid</i>	<i>Hot manifold ignition temperature³ (°C)</i>	<i>Runaway temperature (°C)</i>	<i>AIT (°C)</i>
Water glycol mixture	$> 704 \pm 7$	132.5 ± 7.5	425
Mineral oil	386 ± 5	200 ± 10	341
Oil water emulsion	$> 704 \pm 7$	180 ± 10	385
Rapeseed derivative	466 ± 5	180 ± 10	390
Polyol ester	436 ± 5	155 ± 15	400
Poly glycol ether	396 ± 5	155 ± 15	380
Phosphate ester	$> 700 \pm 5$	> 210	425

³ In some cases, hot surface ignition was not obtained. This was the case, for example, for water glycol at surface temperatures up to 704 °C.