

Lifting the Fog Off Hydrocarbon Mist Explosions

Stéphanie El-Zahlanieh, Shyarinya Sivabalan, Benoît Tribouilloy, Tribouilloy Benoit, David Brunello, Alexis Vignes, Olivier Dufaud

▶ To cite this version:

Stéphanie El-Zahlanieh, Shyarinya Sivabalan, Benoît Tribouilloy, Tribouilloy Benoit, David Brunello, et al.. Lifting the Fog Off Hydrocarbon Mist Explosions. AIChE Spring Meeting 2021 & 17. Global Congress on Process Safety (GCPS), Apr 2021, Online, United States. ineris-03270733

HAL Id: ineris-03270733 https://ineris.hal.science/ineris-03270733

Submitted on 25 Jun 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Lifting the Fog Off Hydrocarbon Mist Explosions

Stephanie El - Zahlanieh¹ LRGP, French National Centre for Scientific Research, University of Lorraine Nancy, France stephanie.el-zahlanieh@univ-lorraine.fr

Shyarinya Sivabalan¹, Tribouilloy Benoit², David Brunello¹, Alexis Vignes² and Olivier Dufaud¹ ¹ LRGP, French National Centre for Scientific Research, University of Lorraine Nancy, France ² INERIS, Accidental Risks Division, Parc Technologique ALATA Verneuil-en-Halatte, France

Keywords: Fuels, Hazard Identification and Risk Analysis, Process Safety, Explosion, Mist

Abstract

Explosion hazards do not only include gases, vapors and dusts, but also mists. Indeed, previous incident surveys identified dozens of accidents related to hydrocarbon mists generation and ignition. The ongoing occurrence of mist explosions demonstrates the need to assess the relevance of the current approaches for the evaluation of the mist ATEX risks and to determine reliable and standardized safety parameters for hydrocarbon mists.

For this study, different fluids with a high industrial interest were selected (e.g. ethanol, kerosene, and diesel) to be tested in a new apparatus based on the standardized 20L explosion sphere. A new fluid injection system was deployed using siphon/gravity-fed spray nozzles comprising a Venturi junction and proposing a wide variety of dispersion performances. This system was controlled using a specifically dedicated program, which ensures the versatility of the apparatus and its adaptability to the different liquids tested. It allows a fine control of the gas carrier flow, the liquid flow and both the injection and ignition times, which makes it possible to change the dilution rate for a desired droplet size distribution. The droplet size distribution (DSD) was determined for the various fluids dispersed into the 20L sphere. Several tests were performed to determine the minimum ignition energy (MIE), the lower explosivity limit (LEL), the maximum explosion pressure (P_{max}) and the maximum rate of pressure rise (dP/dt max) of hydrocarbon mists.

For instance, for a DSD centered at 55μ m, a maximum pressure of 9.2 bar and maximum rate of pressure rise up to 600 bar.s⁻¹ were found for ethanol. As for kerosene, for a DSD centered at 80 μ m, 8.8 bar and 570 bar.s⁻¹ were found respectively. Experimental results obtained between the LEL and the stoichiometric concentration were compared with numerical data obtained with Chemical Equilibrium with Applications and showed that they were coherent with the theoretical calculations and that tests were at least performed up to the stoichiometric concentration. A

sensitivity analysis was conducted to stress the influence of parameters such as the chemical nature of the fuel, the DSD, the ignition energy or the mist temperature (gas/mist ratio). For instance, tests performed on kerosene showed that as the DSD increases, the mist cloud becomes harder to ignite and hence the LEL increases. These first results already allow to propose a new protocol to determine safety parameters for hydrocarbon mists and provide tools to support the evaluation of mist ATEX risks.

1 Introduction

In the chemical and petrochemical industries, various processes are susceptible to lead to the formation of flammable hydrocarbon aerosols under certain conditions [1]. These unintended generations of hydrocarbon mists may ignite and give rise to explosions. Dozens of such explosions have been reported throughout the years despite the efforts taken to mitigate such incidents. In fact, in 1955, Eichhorn shed light on such matter in one of his publications in the Petroleum Refiner entitled "Careful! Mist can explode" [2]. The author has since introduced the possibility of mists igniting at temperatures well below their flash points. A few years later, in 1995, Eckhoff published a literature survey, in which the author reviewed studies concerning the generation, ignition, combustion, and explosion of flammable mists [3]. Interest in such a crucial subject has, since then, grown throughout the years. For instance, Santon [4] published an incident survey reporting 37 mist incidents, among which 9 explosions lead to 29 fatalities. Ten years later, Lees et al. [1] notably showed that 10% of reported releases on offshore oil and gas installations in the United Kingdom involved sprays or mists. More recently, a comprehensive review was performed by Yuan et al. [5] and led to the proposal of a systematic strategy to investigate aerosol explosion, which is in full accordance with the work presented here.

What defines mists? According to the Globally Harmonized System of classification and labelling of chemicals (GHS) [6], mists are defined as liquid droplets, generally of sizes ranging from 1 to about 100 μ m, of a certain substance or mixture suspended in a gas - usually air. The alternative term "smoke" is then used for aerosols with diameters lower than 1 μ m [7], while "sprays" consist of dispersed droplets of diameters generally greater than 50 μ m. Other definitions are also encountered. For instance, Ballal and Lefebvre [8] consider the term "mist" to be suitable up to 100 μ m [9]. As proposed by Eckhoff [10], the terms "spray" and "mist" are used arbitrarily in this paper as they are both relevant in this case.

The ongoing occurrence of mist explosions demonstrates the need to assess the relevance of the current approaches for the evaluation of the mist ATEX risks. Indeed, the importance of hazardous area classification (HAC), as well as the lack of tools which correlate the dispersion of mists with their flammability and explosion severity, is abundantly expressed in literature [5,9]. For instance, European ATEX regulations require assessing the risk of formation and ignition of explosive atmosphere (ATEX) associated with the production of a flammable mists. Nevertheless, due to the lack of tools, risk analysis and area classification prove difficult. This lack of knowledge is clearly highlighted in some guides such as the EI15 [11] in which it is stated that "there is little knowledge on the formation of flammable mists and the appropriate extents of associated hazardous areas".

In order to mitigate the occurrence of such explosions and to assess the flammability and explosion severity of hydrocarbon mists, the factors and criteria of liquid handling, as well as the fluids' safety parameters should be identified. If such definitions are achieved, the classification of hazardous areas (HAC) would be a step closer for mists, and the improvement of current ATEX standards and regulatory provisions concerning mists would then be achievable. In other words, "lifting the fog off hydrocarbon mist explosions" may finally be possible.

In this study, different fluids with a high industrial interest were selected to be tested in a new apparatus based on the standardized 20 L explosion sphere. The mist generated was characterized, notably by its droplet size distribution, and explosion tests were performed to determine the minimum ignition energy (MIE), the lower explosivity limit (LEL), the maximum explosion pressure (P_{max}) and the maximum rate of pressure rise (dP/dt_{max}) of hydrocarbon mists. The main goals of this article are i) to demonstrate through some examples that it is possible to characterize the flammability and explosion severity of mists using modified standard set-ups such as the 20 L sphere, ii) to show the influence of some operating conditions on the safety parameters of hydrocarbon mists.

2 Materials and Methods

2.1 Fuel Selection

A large variety of fuels has been involved in mist explosion incidents, ranging from vegetable oils to hydraulic oils and crude oils [4]. During an oil mists Joint Industry Project (JIP) led by the Health and Safety Executive (HSE) [12], a list of fluids was proposed comprising fuels of industrial interest and specific physicochemical properties. In addition, based on the properties that were identified to be the most affecting on oil mist hazards, a fluid classification system was developed by the Health and Safety Executive. This system divided the chosen fluids into four release classes based on their flashpoint and their ease of atomization, a parameter represented by the Ohnesorge number. For this paper, three fuels were chosen to be studied under certain conditions: ethanol, Jet A1 kerosene and diesel fuel. The choice of each fluid can be justified by both industrial and scientific considerations.

Ethanol

The industrial value of ethanol is continuously growing throughout the years with its emergent use as an engine fuel or fuel additive for automobiles. Such demand for ethanol or ethanol-fuel blends is and will keep leading to an increase in its production and transport, hence increasing the requirements to manage fire or explosion risks. Moreover, flammability and explosion severity studies performed on ethanol vapors as well as mists, such as Mitu and Brandes [13], Thimothée [14], and Pinheiro et al. [15], can be considered abundantly sufficient to help calibrate the testing methods chosen for this paper.

Jet A1 kerosene

In 2009, Santon showed that 7 incidents, over 29 detailed in his review, were related to kerosene mists. They were mostly related to transportation activities, from cargo accident to aviation kerosene explosion [4]. One incident that highlights the dangers of flammable aviation kerosene

and air mixtures is the Trans World Airlines (TWA) flight 800. In fact, this tragic incident led to the loss of 230 lives in an airplane explosion resulting from the ignition of the flammable fuel and air mixture in the center wing fuel tank [16]. Bowen and Shirvill [17] also pointed out the risks behind the potential atomization of kerosene.

Diesel fuel

In their incident review, Lees et al. [1] mentioned that data from the UK Hydrocarbon Release Database (HCRD) showed that 20% of mist reported incidents took place from diesel sources. In fact, the authors stated that out of 48 mist/spray release flash fires reported between 2000 and 2005, 11 involved diesel releases.

Table 1 shows the main physicochemical properties of the selected fuels. It should be highlighted that Ohnesorge number (Oh), used for the HSE classification, represents the ratio between the viscous forces to surface tension and inertial forces, i.e. at a given Reynolds number, increasing Oh will improve liquid atomization.

	Ethanol 96%*	Aviation fuel Jet A1**	Diesel***
Flashpoint (°C)	13-17	≥ 38	> 52
Density (kg.m ⁻³)	790	750 - 840	750 - 850
Viscosity (cSt)	1.2	8	1 – 2.5
Surface Tension (kg.s ⁻²)	0.022	0.026	0.027
HSE Release Class	'Unclassified'	Class I (Oh ratio ≥ 2 , Flashpoint < 125°C)	Class I (Oh ratio ≥ 2 , Flashpoint < 125°C)
Flammability limits (%)	3.3 – 19	0.6 - 6	0.5 - 8
Auto-ignition temperature (°C)	363	210	232 - 260

Table 1. Physicochemical properties of the three selected fuels

* MSDS from ThermoFisher Scientific ** MSDS from Neste *** MSDS from HOLLYFRONTIER

2.2 Mist Generation System

The accidental formation of an explosive flammable mist cloud can be caused by several phenomena. For instance, it can be due to the evaporation of fuels in heated areas, their subsequent circulation and condensation in colder areas [3]. In addition, leaks or ruptures due the damages or corrosion in vessels and pipelines can lead to high pressure releases and hydrocarbon mist

generation. To better represent a spray or mist caused by a leak or rupture, various generation methodologies could be adopted. The wide use of sprays in industrial applications lays out several options to choose from. Nevertheless, the experimental procedure proposed to characterise the ignition sensitivity and explosion severity of mists aims to be standardized, so that the results can be compared and that generic safety measures can be proposed. Therefore, the selected generation system should be easy to find and implement but should also be able to mimic "idealized" conditions of a mist release.

In this work, a siphon/gravity-fed set-up comprising a Venturi junction with two inlets (an air inlet and a liquid inlet) was used to generate mist. This generation system's main component is a spray nozzle set composed of a fluid and an air cap (Spraying Systems) through which the liquid/air jet passes and fragments. Mist spray generation was investigated as a function of three relevant parameters stated by Kooij et al. [18]: nozzle type, spraying pressure and fluid properties. Three nozzle sets, presented in Figure 1, of different orifice diameters were chosen to represent three ranges of droplet size distribution. In addition to the nozzle set, the air injection pressure, which shows important influence on the droplet size, was varied at least between 3 and 5 bar. These pressures were chosen according the maximum pressure tolerance of the three nozzle sets.



Figure 1. The three spray nozzle sets: (a) N1 (b) N' (c) N2

2.3 Mist Characterization

Gant et al. [9] pointed out that characterizing a mist properly before its ignition is of great importance since the safety parameters of mists are highly affected by their droplet size distribution, concentration and turbulence. These three parameters are studied by using an in-situ diffraction laser sensor as well as by implementing Particle Image Velocimetry; in order not to overload this manuscript, PIV experiments will not be developed here.

Droplet Size Distribution (DSD)

The time evolution of droplet size distributions was determined by in-situ laser diffraction (Helos/KR-Vario by Sympatec GmbH) and will be later supplemented by APS spectroscopy (aerodynamic diameter measurement - TSI) to characterize the submicron droplets. The Helos laser sensor used for such measurement is designed to analyse the droplet size of using 3 high-resolution measuring ranges (R1, R3 and R5) from 0.5 μ m to 875 μ m. The apparatus measures the droplet size distribution (DSD) directly through the transparent windows (borosilicate glass) of a modified 20L open sphere similar to the standard explosion vessel [19]. R3 lens was mainly used during this study as it covers a range of droplet diameters from 0.5/0.9 μ m to 175 μ m. The acquisition frequency was set at 2 distributions per millisecond. The measurements given by the sensor are notably the volume diameter d₁₀, d₅₀, d₉₀ and the D_{3,2} (Sauter Mean Diameter, SMD). In order to have an approximation of the DSD near the kernel spark produced by an ignition source described in more details in Section 2.4, the height of the sensor was adjusted to a height corresponding to the location of the ignition source.

2.4 Ignition Sources

Potential ignition sources in accidental fires or explosions can vary from electrical discharges or sparks to hot surfaces or to malfunctioning electrical circuits. In previous studies on explosion limits and deflagrations pressures, ignition sources such as exploding wire ignitors, spark ignition, pyrotechnical ignitors, and hot spot ignitors have been used [8, 20, 21].

For this study, wide ranges of energies, starting from 1 mJ, were required. Therefore, both spark ignition as well as chemical pyrotechnical ignitors were chosen. In tests performed with spark ignition, two stainless steel electrodes were placed in the center of the 20 L explosion sphere with a separating distance of 6 ± 0.1 mm. The electrodes were insulated from the sphere's wall using Teflon plugs. Both electrodes were connected to a KSEP 320 high voltage unit, which was also bypassed by a custom control system. The maximum power generated by such system is 225 W, i.e. 225 J.s⁻¹. Hence, to deliver an energy of 100 J, a permanent spark would be generated during 444.44 milliseconds.

To avoid generating a spark for a long duration and leading to the sedimentation of droplets, permanent spark ignition was used up to 100 J. For higher energies, chemical pyrotechnical ignitors (Sobbe GmbH) of energies ranging from 100 J to 5 kJ were used. These ignitors were actuated electrically by a low-voltage electrical signal sent by the KSEP 310 unit. Both ignition sources were compared at 100 J to ensure that they both deliver approximate results.

2.5 Mist Flammability and Explosion Severity

The Modified 20 L Ignition and explosion Severity Test device (MIST)

The standard 20 L explosion sphere used for dust explosion tests has been modified with the installation of the mist generation system at the bottom of the sphere, the removal of the dust container, as shown in the figure below, and the installation of two electronic valves to control the inlet flowrates as well as the liquid/air ratio. Before injecting the fuel/air mixture, the sphere was partially vacuumed to a calculated pressure, so that, when the mist was fully injected, an atmospheric pressure would be attained. The two ignition sources mentioned in Section 2.4 were

used for this test. The control of the system as well as the acquisition of the data were performed using a new control system and a LabView program prepared by the LRGP. This system controls the KSEP 310 and 332 units (Cesana AG), the inlet electronic valves (gas and liquid) and allows a safe operation of the test equipment and an optimum evaluation of the explosion results with an acquisition frequency of 5000 measurements per second.

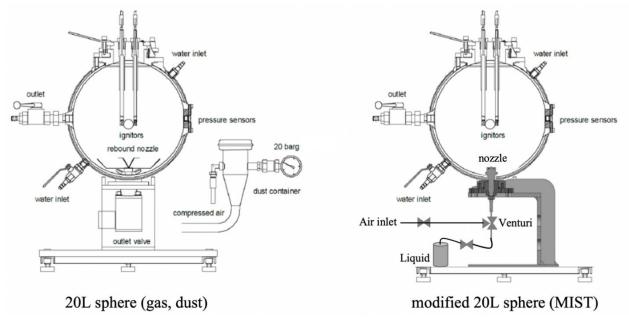


Figure 2. Modification of the standardized 20L explosion sphere Left: standard 20 L sphere – Right: modified 20L sphere

3 Results and Discussion

3.1 Droplet Size Distribution (DSD)

Ethanol mists were generated in the open 20L sphere using the three nozzle sets presented in Section 2.2. DSD measurements were performed during a 4 second generation period and up to 1 second after the valves' closure, with intervals of 50ms (2 DSD per ms and an average value each 50 ms). As it is shown in Figure 3, three ranges of droplet diameters can be obtained by changing the nozzle set. Nevertheless, it should be noted that with nozzle sets N' and N2, a peak corresponding to smaller diameters (between 6 and 18μ m) was always observed, demonstrating the persistence of 'primary droplets' for nozzle N' and of a fragmentation phenomenon for N2. It should be noticed that, in the case of mist generation through the N' nozzle set, hybrid mixtures (vapor – liquid phase) can be created during the ignition step due to the presence of fine droplets being easily vaporized along with larger droplets.

Since fluids' properties are one of the many relevant parameters affecting the droplet size distribution of a mist [18], a sensitivity study has been undertaken to study the influence of preheating or pressurizing the liquid on the DSD. A metallic reservoir that holds up to 10 bar of internal pressure and 100 °C was designed to store the liquid hydrocarbons. Granulometry tests

were also performed on kerosene JetA1 and diesel fuel mists and demonstrated similar DSDs as these shown in Figure 3.

Being able to independently control the DSD and the flow (i.e. the fuel equivalence ratio) is an essential point of any study on the risks associated with mists. This will allow to control and study the effect of the fuel equivalent ratio, the droplet size distribution and the chemical nature of the fuel, and to determine their impact on the ignition sensitivity and the explosion severity of hydrocarbon mists.

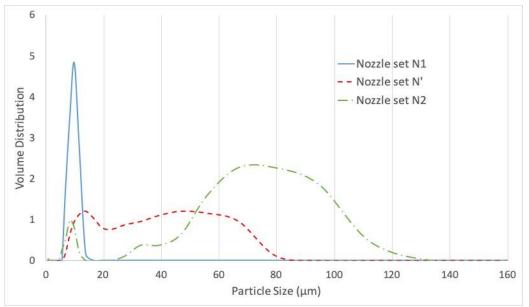


Figure 3. Droplet size volume/mass distribution at t = 2 s of ethanol mist generated at P=3bar using nozzle sets N1, N' and N2

3.2 Minimum Ignition Energy

The minimum ignition energy is the smallest amount of spark energy needed to ignite the most easily ignitable mixture of a flammable substance in air. Such parameter is one of the most important parameters for the assessment of explosion risks and hazardous areas. Krishna [22] stated that a mist becomes more easily ignitable as its mean droplet diameter decreases. This statement can be justified by the fact that the liquid must be partially vaporized before it ignites, and with small diameter droplets, the high surface-area-to-volume ratio leads to a faster evaporation process [9]. Therefore, the nozzle set N1, which generates the smallest mist droplets (Figure 3), was chosen for this test.

For instance, Jet A1 mists were dispersed into the modified 20 L explosion sphere during a fixed injection time, corresponding to the stoichiometric concentration. As the MIE was lower than 100 J, the ignition source used for this part was the spark ignitors. Results showed that the MIE of JetA1 mists of a DSD around $8 - 10 \mu m$ lies between 1 and 3 J. On the contrary, previous studies, such as Shepherd et al. [23], showed that energies between 5 mJ and 1 J are sufficient to ignite a mixture of Jet A vapor fuel in air. This large dissimilarity in values shows how mists can behave differently from vapors and how the presence of droplets can affect the ignition sensitivity. It

should be noted that the electrode shape, spark gap, circuit capacitance and the spark duration are parameters that influence the values of the minimum ignition energy [9, 24].

3.3 Lower Explosive Limit

The lowest concentration of a mixture at which a propagation of a flame is supported away from the ignition source is defined as the Lower Explosive Limit (LEL). For d_{50} ranging from 8 to 10 µm, the LEL of the three selected fluids was determined as shown in Table 2. Burgoyne [25] stated that the lower explosive limit of hydrocarbon mists, specifically tetralin, is around 50 g.m⁻³. The difference between the results obtained and those found by Burgoyne [25] can be explained by the different sources used to ignite the mists (a pilot flame in the case of Burgoyne). Moreover, the experimental apparatus used to measure the LEL might influence the values [9]. It should also be noted that a single average diameter is often given in order to describe the whole DSD, whereas it can greatly vary as a function of the turbulence and time due to coalescence phenomenon. In 1995, Eckhoff concluded that the LEL of a spray will range between 100 g.m⁻³ and 500 g.m⁻³ regardless of the fuel volatility [3]. Dufaud et al. [26] also found a LEL in the order of 250 g.m⁻³ for lube oil mists. Table 2 shows that lower values of LEL can be obtained.

	Ethanol 96%	Aviation fuel Jet A1	Diesel
LEL (g.m ⁻³)	77 ± 5	94 ± 5	73 ± 5

As stated by Gant [9], the LEL of a mist is linked to the size of the droplets. In fact, Burgoyne [27] studied the effect of the droplet size on the lower limit of flammability and showed that in upward flame propagation tests, the LEL values tend to decrease with increasing drop diameter; however, with a downward flame propagation such indications become harder to pinpoint with increasing drop diameters due to the presence of droplets falling vertically downwards. Tests on kerosene JetA1 mists generated with the three nozzle sets and ignited with 100 J chemical ignitors showed that as the DSD increases, it becomes harder to ignite the mist cloud and hence the LEL increases (Table 3), a result well coherent with that found by Zabetakis [28].

Table 3. Effe	ct of the DSD on	the LEL of keros	ene JetA1 mists
---------------	------------------	------------------	-----------------

Nozzle set	Mist DSD range (µm)	D10 (µm)	D50 (µm)	SMD (µm)	LEL (g.m ⁻³)
N1	8-10	7	9.2	9	94 ± 5
N'	40-60	10	15	14.5	127 ± 5
N2	80-100	8	63	17	166 ± 5

3.4 Explosion Severity

3.4.1 Validation Tests

As presented in Section 2.5, the 20 L sphere was modified to allow the generation of mists and chemical ignitors of 100 J were used as ignition source. In accordance with the European standard PR EN15967:2020 "Determination of the maximum explosion pressure and maximum rate of pressure rise of gases and vapors", a MATLAB program was prepared to determine P_{max} as well as the maximum rate of pressure rise dP/dt_{max} using both two-point and five-point derivatives.

Preliminary tests were performed on ethanol to calibrate the test method used. They show that the maximum explosion pressure reaches 7.5 barg, whereas the maximum rate of pressure rise reaches 567 bar.s⁻¹ (Figure 4) for an ethanol mist concentration of 128 g.m⁻³ ($6.3\%_{v/v}$) of a DSD of about 8 - 10 µm generated by the nozzle set N1 with an injection pressure of 3 bar and at ambient temperature. These values are in good agreement with Mitu and Brandes [13] who found an absolute maximum explosion pressure of about 8 bar for an explosion of vapor ethanol/air mixture with a fuel/air ratio $\varphi = 1.15$.

Chemical Equilibrium with Applications (CEA) was also used as a comparison tool between experimental results and theoretical calculations of liquid or gas phase combustion. Numerical data on the combustion of ethanol at different fuel/air ratios were obtained using CEA (Figure 5). Such results are consistent as the explosions performed in the MIST sphere are not under adiabatic conditions; therefore, it is normal to obtain an experimental P_{max} well below the theoretical adiabatic overpressure. Figure 5 also shows that tests were at least performed up to the stoichiometric concentration. The use of the CEA program permits to obtain theoretical equilibrium compositions of combustion products which can help in modeling mist explosions more finely.

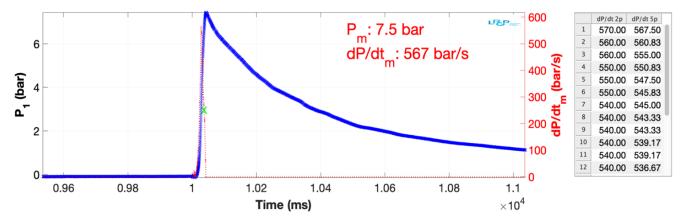


Figure 4. Time evolution of the explosion pressure of ethanol mist in the 20L explosion sphere

(generation at ambient temperature using nozzle set N1)

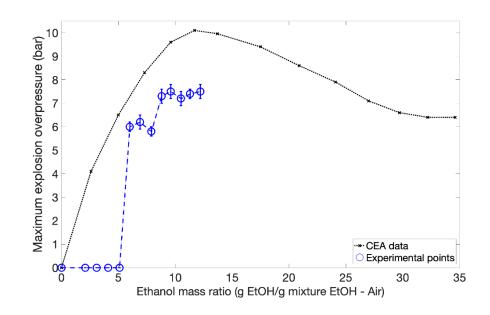


Figure 5. Comparison between theoretical CEA overpressure values and experimental results

<u>3.4.2</u> Effect of the Chemical Nature of the Fluid

The three fluids presented in Section 2.1 were tested under the same injection conditions (Nozzle set N1, ambient temperature, $P_{inj} = 3$ bar) and using the same 100 J chemical ignitors. Figures 6 and 7 present the variation of both the maximum explosion overpressure and the maximum rate of pressure rise for diesel fuel, kerosene Jet A1, and ethanol. It should be noted that the lines are only arbitrary point-to-point connections and that a fitting will be adopted later on. As it can be seen, in the figures below, the three mists behave very differently once ignited. For ethanol, a maximum value of P_{max} of 7.5 barg and a maximum rate of pressure rise of 567 bar.s⁻¹ were found at a mist concentration of 128 g.m⁻³. However, in the case of kerosene JetA1, 6.4 barg for P_{max} and 356 bar.s⁻¹ for dP/dt_{max} were found at a mist concentration of 159 g.m⁻³, which is consistent with tests performed by Zheng et al. [20] on high flash point jet fuels (RP-5 and RP-3) resulting in values of about 6.5 bar as Pmax. As for diesel fuel mists, even though its values of rate of pressure rise are not much elevated, it can be seen that diesel can indeed ignite and produce an explosion at a temperature well below its flashpoint.

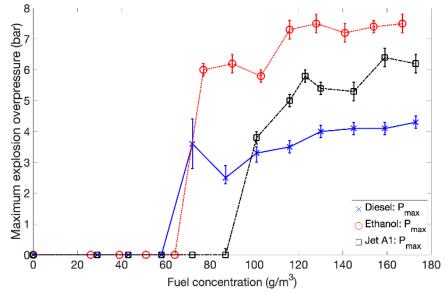


Figure 6. Variation of the maximum overpressure as a function of fuel concentrations in the 20L sphere

(generation at ambient temperature using nozzle set N1)

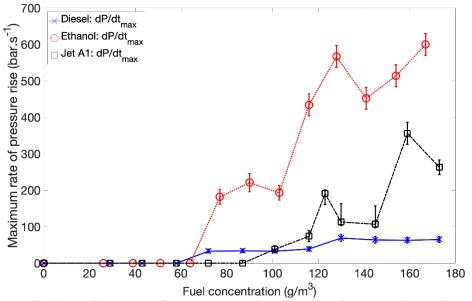


Figure 7. Variation of the maximum rate of pressure rise as a function of fuel concentrations in the 20L sphere

(generation at ambient temperature using nozzle set N1)

<u>3.4.3</u> Effect of the Ambient Temperature

The MIST sphere is equipped with a water jacket to control its temperature. To study the influence of the ambient temperature on the explosion severity of mists, the temperature of the sphere was notably increased to 60°C for kerosene JetA1, which corresponds to a temperature greater than its flashpoint. Tests were performed at various concentrations with spark ignition source at 100 J. Results showed how ambient conditions can affect the rate of explosion pressure rise from values of 168 bar.s⁻¹ at $T = 20^{\circ}C$ to about 540 bar.s⁻¹ at $T = 60^{\circ}C$ at a mist concentration of 87 g.m⁻³. This change can be explained by an increase of the vapor concentration around the droplet before ignition, reaching the lower explosive limit. The ignition step is then governed by a gas combustion regime and not limited by the droplet evaporation as it is the case at 20° C. It can be seen in Figures 8 and 9 that this effect is more pronounced on the maximum rate of pressure rise than on the maximum explosion pressure, highlighting the impact of the temperature on the mist combustion kinetics. It should also be underlined that the effect of the temperature is less perceptible at high mist concentrations. In fact, the vapor pressure of kerosene increases from 4 to 18 mbar when the temperature rises from 20 to 60°C [29]. This implies that, even at 60°C, kerosene cannot be fully vaporized for concentrations greater than 125 g.m⁻³. Moreover, apart from the very first moments of ignition, the flame temperature will rapidly rise over the auto-ignition temperature, the impact of the initial temperature being then negligible. As a consequence, for mist concentrations greater than 125 g.m⁻³, it seems consistent that the difference between vapor and mist explosive behavior becomes smaller.

In addition to the augmentation of the sphere's temperature, pyrotechnical ignitors of high energies (1, 2, 5 and 10 kJ), which permit to increase the temperature locally, were tested. The results were consistent with that obtained by increasing the sphere's temperature.

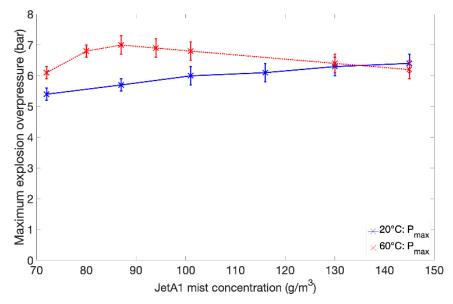


Figure 8. Variation of the maximum overpressure as a function of Jet A1 mist concentrations in the 20L sphere at two different temperatures (generation using nozzle set N1)

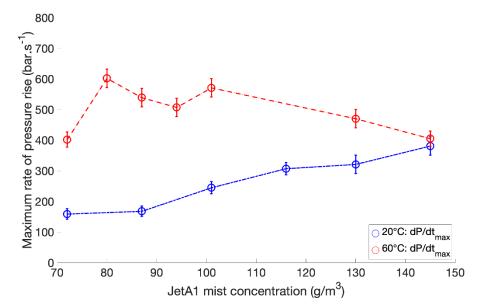


Figure 9. Variation of the rate of pressure rise as a function of Jet A1 mist concentrations in the 20L sphere at two different temperatures (generation using nozzle set N1)

3.4.4 Effect of the Droplet Size Distribution

The droplet size distribution is one important parameter to be taken into account while studying the flammability and the explosion severity of a mist. In fact, in low-momentum sprays or quiescent mists, large droplets are affected by gravity and hence fall faster than smaller droplets, which tend to decrease the average mist mass concentration [9]. Such effect can be seen in Table 4 where both the P_{max} and dP/dt_{max} decreased with increasing droplet size distributions at a mist concentration of about 165 g.m⁻³. Obviously, this is not the only phenomenon to consider and the droplet surface can also play a significant role in the evaporation phase (d² combustion law) as well as in the heat transfer, especially for radiative transfers.

Table 4. Explosion severity of JetA1 using the three nozzle sets for mist generation
--

	Nozzle set N1	Nozzle set N'	Nozzle set N2
P _{max} (barg)	6.2	3.5	0.8
dP/dt _{max} (bar.s ⁻¹)	263	46	28

4 Conclusion

This study aims to demonstrate that it is possible to characterize the ignition sensitivity and explosion severity of hydrocarbon mists with a single set-up. These tests should be performed on simple or/and standard equipment which can be found in industries or laboratories, in order to be able to compare the results and propose adequate solutions for explosion risk management. The Modified 20 L Ignition and explosion Severity Test device, developed in this study, can be a suitable apparatus to assess mist explosivity.

Experiments carried out on ethanol, kerosene JetA1 and diesel have allowed to validate the procedures and setups and show the influence of some operating conditions, such as the ambient temperature, the fuel equivalence ratio and the droplet size distribution, on the safety parameters of hydrocarbon mists. For instance, MIE values for JetA1 mists of 8 - 10 μ m were found to lie between 1 and 3 J, a value much greater than that corresponding to JetA1 vapors (50mJ). The LEL of the three hydrocarbon mists showed to be 77 g.m⁻³ (3.8%_{V/V}), 94 g.m⁻³ (1.2%_{V/V}) and 73 g.m⁻³ (0.86%_{V/V}) for ethanol, JetA1 and diesel mists respectively. Moreover, it was notably demonstrated that the LEL of JetA1 mist increases with an increasing DSD. The rate of pressure rise is also greatly influenced by the combustion kinetics and its rate-limiting step (evaporation, fuel oxidation...). The effect of the sphere temperature was also studied, showing an increase in both P_{max} and dP/dt_{max} of JetA1 mist explosions when the temperature of the sphere was increased from 20°C to 60°C and thus highlighting the influence of the fuel phase (gas, gas-aerosol, liquid aerosol) before ignition. It was also shown that, over the tested diameter range, increasing DSD leads to a diminution of the explosion severity.

Additional tests are currently performed at various initial turbulence levels and on a larger range of DSD with several conventional fuels or hydrocarbons (e.g. iso-octane, biodiesel, light fuel oil, lube oil...). This experimental approach will be coupled with both CFD simulation, to better understand the hydrodynamics within the explosion vessel, and combustion modeling, to highlight the influence of the rate-limiting step of mist explosion on the safety parameters.

Acknowledgements

This work was supported financially by the French Ministry for the Ecological Transition and the French Ministry of Higher Education, Scientific Research and Innovation. The authors would also like to thank the Health and Safety Executive (UK) for the fruitful exchanges on the topic. The contents of the work, including any opinions and/or conclusions expressed, are those of the authors alone and do not necessarily reflect French Ministries policies.

5 References

- [1] P. Lees, S. Gant, R. Bettis, A. Vignes, J.-M. Lacome and O. Dufaud, Review of recent incidents involving flammable mists, (2019) 23.
- [2] J. Eichhorn, Careful! Mist can explode, Petroleum Refiner. 34(11) (1955) 194–196.
- [3] R.K. Eckhoff, Generation, ignition, combustion and explosion of sprays and mists of flammable liquids in air: a literature survey, 1995.
- [4] R.C. Santon, Mist fires and explosions an incident survey, (2009).
- [5] S. Yuan, C. Ji, H. Han, Y. Sun and C.V. Mashuga, A review of aerosol flammability and explosion related incidents, standards, studies, and risk analysis, Process Safety and Environmental Protection. 146 (2021) 499–514. https://doi.org/10.1016/j.psep.2020.11.032.
- [6] ST/SG/AC.10/30/Rev.8, Globally harmonized system of classification and labelling of chemicals (GHS)., 8th revised version, UNITED NATIONS, New York and Geneva, 2019.
- [7] R.P. Fraser and P. Eisenklam, Liquid Atomization and the Frop Size of Sprays, Institution of Chemical Engineers, 1956.
- [8] D.R. Ballal and A.H. Lefebvre, A general model of spark ignition for gaseous and liquid fuelair mixtures, Symposium (International) on Combustion. 18 (1981) 1737–1746. https://doi.org/10.1016/S0082-0784(81)80178-6.
- [9] S. Gant, Generation of flammable mists from high flashpoint fluids: literature review, Health and Safety Executive, Research Report RR980. (2013). https://www.hse.gov.uk/research/rrpdf/rr980.pdf.
- [10] R.K. Eckhoff, Chapter Five Explosions of Clouds of Combustible Liquid Droplets in Air, in: R.K. Eckhoff (Ed.), Explosion Hazards in the Process Industries (Second Edition), Gulf Professional Publishing, 2016: pp. 209–232. https://doi.org/10.1016/B978-0-12-803273-2.00005-0.
- [11] Energy Institute, Model code of safe practice: Area classification code for installations handling flammabe fluids, (2015). https://publishing.energyinst.org/topics/asset-integrity/eimodel-code-of-safe-practice-part-15-area-classification-for-installations-handlingflammable-fluids.
- [12] G. Burrell and S. Gant, Liquid classification for flammable mists, Health and Safety Executive, Research Report RR1108 (2017).
- [13] M. Mitu and E. Brandes, Influence of pressure, temperature and vessel volume on explosion characteristics of ethanol/air mixtures in closed spherical vessels, Fuel. 203 (2017) 460–468. https://doi.org/10.1016/j.fuel.2017.04.124.
- [14] R. Thimothée, Experimental characterization of a laminar flame propagation in a two-phase medium (aerosol) in microgravity conditions, Theses, Université d'Orléans, 2017. https://hal.archives-ouvertes.fr/tel-01994264 (accessed February 28, 2020).
- [15] A.P. Pinheiro, J.M. Vedovoto, A. da Silveira Neto and B. G.M. van Wachem, On ethanol droplet evaporation in the presence of background fuel vapor, (2019) 8.
- [16] T. Al Mughanam, Operational Flammability Limits of Commercial Fuels A Review, MATEC Web Conf. 249 (2018) 03013. https://doi.org/10.1051/matecconf/201824903013.
- [17] P.J. Bowen and L.C. Shirvill, Combustion hazards posed by the pressurized atomization of high-flashpoint liquids, Journal of Loss Prevention in the Process Industries. 7 (1994) 233– 241. https://doi.org/10.1016/0950-4230(94)80071-5.

- [18] S. Kooij, R. Sijs, M.M. Denn, E. Villermaux and D. Bonn, What Determines the Drop Size in Sprays?, Phys. Rev. X. 8 (2018) 031019. https://doi.org/10.1103/PhysRevX.8.031019.
- [19] A. Santandrea, M. Gavard, S. Pacault, A. Vignes, L. Perrin and O. Dufaud, 'Knock on nanocellulose': Approaching the laminar burning velocity of powder-air flames, Process Safety and Environmental Protection. 134 (2020) 247–259. https://doi.org/10.1016/j.psep.2019.12.018.
- [20] L. Zheng, L. Changbo, A. Gaojun, X. Chunhua, Z. Youjie, R. Lianling, W. Xudong, X. Lifeng, W. Gang, W. Feng and S. Limin, Comparative Study on Combustion and Explosion Characteristics of High Flash Point Jet Fuel, Procedia Engineering. 84 (2014) 377–383. https://doi.org/10.1016/j.proeng.2014.10.447.
- [21] S. Yuan, C. Ji, A. Monhollen, J.S.-I. Kwon and C. Mashuga, Experimental and thermodynamic study of aerosol explosions in a 36 L apparatus, Fuel. 245 (2019) 467–477. https://doi.org/10.1016/j.fuel.2019.02.078.
- [22] K. Krishna, Measurement and Prediction of Aerosol Formation for the Safe Utilization of Industrial Fluids, 2003. http://psc.tamu.edu/files/library/center-publications/theses-anddissertations/kk_phd_chen.pdf (accessed September 24, 2020).
- [23] J.E. Shepherd, J.C. Krok and J.J. Lee, Jet A Explosion Experiments: Laboratory Testing, (1997) 74.
- [24] C.J.R. Coronado, J.A. Carvalho, J.C. Andrade, E.V. Cortez, F.S. Carvalho, J.C. Santos and A.Z. Mendiburu, Flammability limits: A review with emphasis on ethanol for aeronautical applications and description of the experimental procedure, Journal of Hazardous Materials. 241–242 (2012) 32–54. https://doi.org/10.1016/j.jhazmat.2012.09.035.
- [25] J.H. Burgoyne, Mist and spray explosions, Chemical Engineering Progress. 53 (1957) 121– 124.
- [26] O. Dufaud, A. Charvet, G. Mougel, M. Molière, S. Luthun, D. Brunello, L. Perrin, S. Delimoges and M. Couchot, Generation, Characterization and Ignition of Lube Oil Mists, in: ASME Turbo Expo 2015, ASME, Montreal, Canada, 2015: p. Paper No. GT2015-43524, p. V04BT04A039; 8 pages. https://doi.org/10.1115/GT2015-43524.
- [27] J.H. Burgoyne, The flammability of mists and sprays, Proc. 2nd Symp. on Chemical Process Hazards. (1963) 1–5.
- [28] M.G. Zabetakis, Flammability characteristics of combustible gases and vapors, Bureau of Mines, Pittsburgh, PA (United States), 1964. https://doi.org/10.2172/7328370.
- [29] J.E. Shepherd, C.D. Nuyt and J.J. Lee, Flash Point and Chemical Composition of Aviation Kerosene (Jet A), (1999) 38.