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# Experimental Study of Accidental Industrial LPG Releases: Rain out investigation

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#### Abstract

The objective of the atmospheric dispersion research projects of INERIS are to develop models of flashing releases as encountered in realistic industrial environments. Equivalent source term models exist for flashing release in current long range dispersion models. Several factors can, however, invalidate simplified equivalent source models, especially in the very near field where obstacles can be found.

To perform these objectives, INERIS took part in an European project called FLIE [1] (Flashing Liquids in Industrial Environment). In this project, INERIS carried out large-scale experiments with propane and butane releases. The French ministry in charge of Environment also supported INERIS participation which dealt with the modelling of two-phase jets. This paper aims at presenting the large-scale experiments and the main results relative to the rain out observations.

The experimental set-up is located in the INERIS test site. It allows performing propane and butane liquid releases at ambient temperature with a regulated pressure from the saturation pressure to 15 bar with an orifice (circular or rectangular shape) of an equivalent diameter from 10 mm to 25 mm. It is possible to realize free jets but also impinging jets by introducing obstacle at a maximum distance of 2 meters from the release point.

To develop models requires experimental validation data. Up to now, little of experiments were carried out on a great scale with such a variety of measurements. The results revealed some new points related in particular to the rain-out which are seldom discussed in the literature and which can be useful to develop models of flashing releases. Another important lesson learnt is that pools formed by LPG release jets do not contain liquid exclusively but also a mixture with ice. This has a great influence on the pool evaporation.

## 1. Introduction

#### 1.1 Context

Liquefied gases are in widespread use in industry due to their chemical or physical properties. In parallel, numerous industrial processes use fluids that are overheated compared with ambient temperature conditions. Following a loss of containment around an installation, the hazards faced can be instantaneous, as with the explosion of flammable products, or differed as is the case with releases of toxic products.

In order to calculate safety perimeters around industrial installations, one of the objectives of atmospheric dispersion research projects of INERIS is to develop models of flashing releases in realistic industrial environments. Now, equivalent source term models exist for flashing releases. However, several factors can invalidate these simplified equivalent source term models, especially in the context of realistic industrial environments. For instance, liquefied pressurized gases released are rarely pure products, the shape and size of the breach can be strongly different than which had been usually used to develop existing models, in the very near field of the release, obstacles can be found, etc...

To perform his objective, INERIS carried out "large-scale" trials of "industrial" (that is not pure) propane and butane releases in order to develop a tool able to evaluate the gas and the liquid fraction (size of droplets forming the aerosol and liquid pool) in the near field of the flashing release. This INERIS paper aims to present the large scale experimental set-up and the main current results.

#### 1.2 What happens in the event of an accidental release?

The consequences of an accidental release will depend on the one hand on the initial storage conditions, and also on the environment. This is because the presence of obstacles of sufficient size may affect the jet and change the characteristics of the hazardous cloud.

At the breach level, in the case of an accidental release of pressurized liquefied gas, the product released in the ambient air is suddenly placed under temperature and pressure conditions that are such that a part of the liquid vaporizes violently. This phenomenon is generally called a "flash" or a "thermal fragmentation". The sudden vaporizing of a part of the liquid leads to the fragmentation of the liquid jet into fine droplets whose diameter and speed will be especially dependent on the difference between the initial substance temperature and the substance boiling point. A mechanical fragmentation can also be observed when shear stress due to friction turbulences break the liquid phase despite the surface tension forces [2].

In the absence of any obstacle, after a length of a few times the diameter from the breach, a twophase jet develops. This jet is made up of droplets in suspension in a mixture of gas and ambient air that is drawn along. The atmospheric dispersion of this two-phase jet is notably dependent on the environment. In cases where no obstacles are present along the path of the jet, the different contributions to the formation of a flammable or toxic cloud are as follow [3]:

- the quantity of product likely to be quasi-instantaneously vaporized and the droplet size distribution due to the flash phenomena at the breach level ;
- the quantity of product likely to form a pool on the ground (commonly called rain-out) that then evaporates into the atmosphere due to mass and heat transfer phenomena; and lastly,
- the quantity of product likely to be drawn into the air in aerosol form.

In this case, the estimation of the main amounts of vapour released in the air consists in studying the evaporation of the formed droplets and the formation, and then the evaporation, of a liquid pool at ground level. This estimation is important to define a realistic source term that will be used in an atmospheric dispersion model.

In the case of a release in the presence of obstacles, the interaction between the jet and the obstacle may notably change the calculation of the source term. In that case, it is important to determine what becomes of the drops after impact. To do this, it is necessary to know the characteristics of the jet and the droplets prior to impact as well as those of the obstacle.

## 2. The experimental set-up

The experimental set-up, developed in order to understand these phenomena, is located in the INERIS site. The figure 1 shows the release field and the control room in the background. The figure 2 is a diagram of the experimental set-up.



Figure 1 : Photograph of the test field

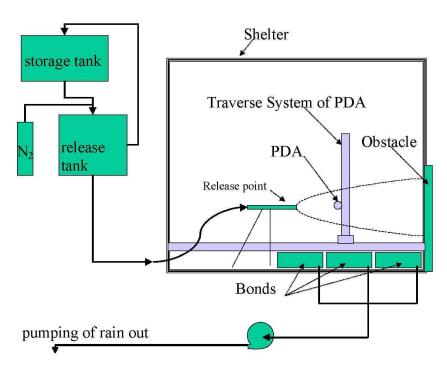


Figure 2 : Diagram of the experimental set-up

The experimental set-up allows us to perform propane and butane liquid releases (the release point is presented in figure 3) at ambient temperature with a regulated pressure from the saturation pressure to 15 bar with an orifice of an equivalent diameter from 10 mm to 25 mm. It is possible to carry out free jets but also impinging jet by introducing obstacle at a maximum distance of 2 meters from the release point.



Figure 3 : Photograph of the release point

The table 1 presents the values chosen for each of the main parameters that play a role on the jet and rain out characteristics. In this table, one will notice that the temperature of the fluid, even if it plays a fundamental role, was not a fixed parameter. Indeed, to heat a tank containing 1 ton of LPG in full safety is very constraining.

Parameter	Value	Commentary				
Product	Propane	We did not use "pure" gases, we preferred use typical gases usually				
	Butane	found in industrial sites.				
Storage pressure	Saturation Pressure + 0 bar Saturation Pressure + 1 bar Saturation Pressure + 3 bar Saturation Pressure + 6 bar	We decided to fix the gas pressure taking into account his saturation pressure (at ambient temperature) because we wanted to compare two different gases in their usual use in industry, that is ambient temperature and saturation pressure for storage use or ambient temperature and saturation pressure plus some bars due to filling operations or hydraulic pressure for example.				
Type of jet	Free jet Impinging jet	Free jet is an horizontal jet at 1.5 m from the ground For impinging jets, we used steel plates 3 mm thickness				
Distance of impact	0.8 1.6 2.6					
Exit orifice shape and size	Circular (diameter of 10, 15, 20 and 25 mm) Rectangular (9*9, 6*18 and 3.2*25 mm <sup>2</sup> )					

 Table 1. Values of parameters set for the experimental trials

During releases, several parameters are recorded:

- Ambient conditions: direction and speed of the wind, temperature, humidity and atmospheric pressure.
- Release tank: pressure, temperature at several heights in the tank and at the liquid/gas interface, weight of the tank (to calculate the mass flow rate).
- Release point: pressure and temperature just before the outside.
- Inside the jet: a Dual Phase Doppler Anemometer allows to measure speed and size of droplets at several locations in the jet.
- Rain out: Six bonds equipped with continuous weight measurements are used to study the phenomena of liquid pool formation.
- Obstacle: Six surface temperature thermocouples.

## 3. Rain out observations

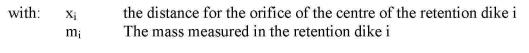
This section deals with main results obtained with rain out measurements for free and impinging jets of propane and butane.

#### 3.1 Free jets

Concerning free jets, only butane gave us measurable rain out. This can be explained by the much lower boiling point of propane (-42°C) than the boiling point of butane (-0.5°C). Moreover, the propane saturation pressure at ambient temperature (8.3 bar at 20°C) is higher than that of butane (2.1 bar with 20°C). Figure 4 and table 2 present the influence of the storage pressure (the storage temperatures are identical) on the distribution of the rain out on the ground. In the figure 4 and table 2, the % rain out is the ratio between the mass flow recorded in a retention dike and the total mass flow rate of the release.

In this figure, the vertical dotted lines present the position of the gravity centre estimated of the measured rain out for pressures (storage and release pressure are very near) of 2.6 bar, 3.8 bar and 5.6 bar. The gravity centre position, noted  $x_g$ , is calculated starting from the relation:

$$x_G = \frac{\sum_{i=1}^{i=6} x_i \times m_i}{\sum_{i=1}^{i=6} m_i}$$



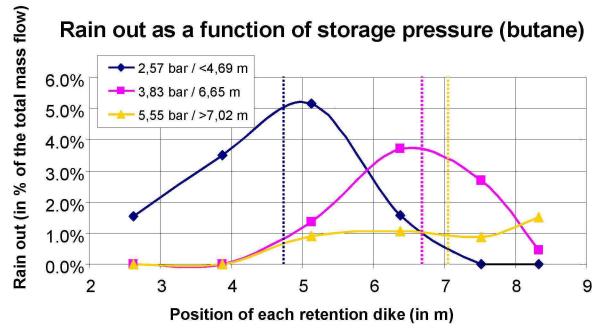


Figure 4 : Rain out as a function of storage pressure (butane)

	Positi						
Storage pressure (bar) / Position of the rain out gravity centre (m)	2.6	3.9	5.1	6.4	7.5	8.3	Total
of the fam out gravity centre (m)		Rain Out					
2.6 bar / <4.7 m	1.6%	3.5%	5.2%	1.6%	0.0%	0.0%	11.8%
3.8 bar / 6.7 m	0.0%	0.0%	1.4%	3.7%	2.7%	0.5%	8.2%
5.6 bar / >7.0 m	0.0%	0.0%	0.9%	1.1%	0.9%	1.5%	4.3%

 Table 2. Rain out distribution as a function of pressure storage

The rain out distribution for these three pressures shows that the mechanical fragmentation is dominating on the thermal fragmentation. Indeed, in his thesis [4], A. Touil showed that in the case of thermal fragmentation, the gravity centre of the rain out is often located close to the orifice. For these tests, we can explain the mechanical fragmentation by the fact that the superheat temperature (difference between the storage temperature and the boiling point of the fluid) of the butane is low ( $\leq 20^{\circ}$ C).

Concerning the influence of the storage pressure, the higher the pressure is, the more the gravity centre is far away from the orifice. That rises owing to the fact that the storage pressure influences directly the speed of the jet.

Concerning the total quantity of rain out, one notes obviously that the higher the pressure is, the less the rain out is significant. That is related to the fact that by increasing the pressure, the speed of the jet increases and thus mechanical fragmentation generate finer drops which can then evaporate more easily.

It is significant to note that the total rain out can exceed 12% of the initial released mass flow. Indeed, the liquid recovered in the six retention dikes corresponds inevitably to an underestimation: we can't capture all the liquid part touching down on the ground. In addition, it should not be forgotten that the quantity of rain out depends on the altitude of the breach compared to the ground. For these tests, this parameter was fixed at 1,5 m. This means that if tests had been carried out at lower altitudes, the measured quantities doubtless would have been larger.

#### 3.2 Impinging jets

For impinging jets, propane rain out can be observed. Total rain out is more significant for butane than for propane. As indicated in the case of the free jets, that is explained by the differences in boiling point and saturation pressure of the two gases.

Figure 5 represents, for butane and propane, the rain out part measured according to the distance between the obstacle and the outlet. The main tendency is that the longer the distance between the obstacle and the orifice is, more the rain out is small. That can be explained by the fact that the more the impact takes place late, the more the droplets forming the aerosol have time to evaporate before impact. It should be noted that it is possible to connect the measured rain out

with the liquid fraction theoretically calculated for the same free jet at a distance equal to that of the obstacle.

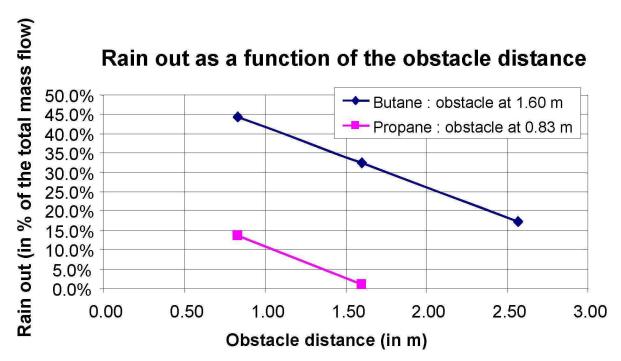


Figure 5: Rain out as a function of the obstacle distance

Qualitatively, it is interesting to note that after releases, the liquid present in the retention dikes is sufficiently cold (below the boiling point) so that the mass transfer seems to govern the pool evaporation [5]. The observation of rain out records as a function of time during release tests shows that less than one minute is required to observe liquid accumulation in retention dikes. This might correspond to the time required to cool down the obstacle and retention dike enough to be able to "capture" a fraction of the liquid part of the jet.

This duration of one minute is of course a function of many parameters (nature of the gas, release and ambient conditions, distance from the obstacle, nature of obstacle and retention dike, etc...) but for a first approach, it is an order of magnitude that we observed in many cases in these tests carried out with LPG and also in the case of Ammonia [6] tests carried out 6 years before.

Thus, a simple method to characterize the source term resulting from an impinging two-phase jet, can be to considerer that, during the first minute of release, all the droplets are vaporized in contact with the obstacles and that, beyond the first minute, part of the droplets will be captured by the obstacles becoming enough cold to contribute to the pool formation of which it will be necessary to evaluate the evaporation rate.

The figure 6 shows an example of a rain out record as a function of time that comes from an impacting jet of propane. The experimental conditions are :

- storage pressure : 9.6 bars,
- orifice diameter : 10 mm,

- distance of the obstacle distance : 0.86 m,
- ambient conditions : temperature : 22.5 °C, relative humidity : 65%, temperature humidity dew point : 16.2°C.

Water of the atmosphere makes part of the rainout due to the phenomenon of jet-entrainment that mixes the released propane with moist air. The measured mass in the retention dike results from an equilibrium between rain out of this mixture and his evaporation. When all the phenomena are in a steady-state in terms of temperature, a slope can be observed on the curve. The component evaporation is certainly very weak: during the jet, the "air" above the pool could be saturated with LPG vapours. In a first approximation the evaporation component can be neglected so the slope corresponds to the accumulation on the ground of a part of droplets and a part of condensed water coming from ambient humidity.

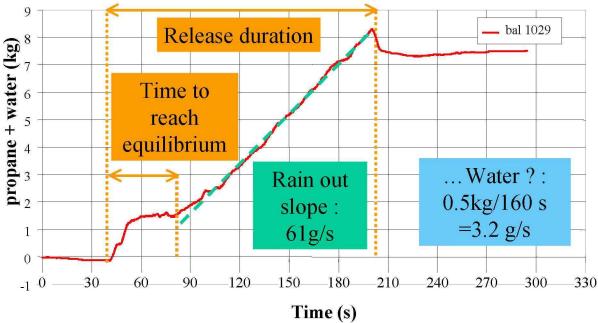


Figure 6 : Example of a rain out record as a function of time

Moreover, after releases we observed the almost systematic formation of "ice" in the retention dikes like on the obstacle. The nature of this "ice" was not analyzed but it is very probable that it is not entirely made up of water. Indeed, the observation of this solid reveals that while it's melting, cavities appears on the solid surface as if a gas (is it LPG or a LPG hydrate?) was desorbing after being captured by initial solidification.

Thus, it appears that the determination of the pool evaporation rate will have to take into account the possible presence of a solid part whose nature and behaviour is not well defined.

#### 4. Conclusion

In order to develop models of flashing releases as encountered in realistic industrial environments, INERIS carried out large-scale experiments with propane and butane. In the first part, this article presented the large scale experiments. The second part was dedicated to the main results concerning the rain out formation.

We presented the influence of type of product, storage pressure, type of jet and distance of impact over the rain out formation.

Moreover, we highlighted difficulties to interpret rain out measurements and to evaluate pool evaporation rate due to the presence of the water in the atmosphere.

Following these tests, it seems significant to continue the investigations to understand the influence of the air moisture on the formation of the rain out. Moreover, pool evaporation model taking into account a solid part in the pool will have to be developed.

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