

## Rainout Assessment after a Release of Hazardous Liquefied Gas: Effect of Physical and Storage Parameters

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In process industry it is common practice to store and handle dangerous substances, which are gaseous at atmospheric temperature, as pressurized or refrigerated liquids. In case of accidental release, a fluid in these conditions gives rise to a two-phase discharge. In such conditions, the liquid breaks up in an aerosol cloud, whose behavior in atmosphere affects dispersion distances. The physical phenomena occurring after the release to the atmosphere are mainly three: expansion to ambient pressure, liquid atomization (break-up) and rainout. In particular, the rainout fraction is a crucial parameter in quantitative risk analysis and therefore models capable of estimating whether it occurs or not are of paramount importance.

This study aims at emphasizing the influence of some physical parameters and storage related parameters on the rainout fraction, taking into account continuous and stationary releases of ammonia and LNG (Liquefied Natural Gas). These represent two different classes of substances: gas liquefied by pressure (superheated) and gas liquefied by refrigeration (subcooled), which undergo to different jet break-up mechanisms. The results obtained for these two compounds can then be extended to the whole range of pressurised liquids/liquefied gases.

A simple model has been developed to evaluate rainout occurrence, the simple approach here proposed (which has been validated comparing its results to those of other simulation models as well as to some available experimental data) allows to investigate in a simple and effective way the importance of the different physical and storage parameters involved during rainout.

### 1. Introduction

Dangerous materials that are gaseous at ambient temperature are commonly stored as pressurized or refrigerated liquids. In case of accidental release, these liquids give rise to a two-phase discharge. In such conditions, the liquid breaks up in an aerosol cloud, which behavior in atmosphere affects dispersion distances. The physical phenomena occurring after the release to the atmosphere are mainly three: expansion to ambient pressure, liquid atomization (break-up) and rainout.

In particular, the largest droplets can lead to the formation of an evaporating pool, while the smallest ones will stay into the gas cloud. Understanding whether a specific liquid release will lead to a pool formation or not is of paramount importance to define the inlet conditions for the cloud dispersion modeling leading to the safety distance estimation. This is quite cumbersome since the physical phenomena occurring after the release of a liquefied gas (namely, jet expansion up to the ambient pressure, mechanical and/or flashing liquid break-up, and rainout) are not easy to model in detail. The rainout fraction is a parameter of crucial importance in Quantitative Risk Analysis (QRA) and therefore lumped parameters mathematical models have been developed for mechanical and/or flashing liquid break-up, as well as for rainout (Cleary et al., 2007; Witlox et al. 2007). However, such models, while requiring dedicated computer codes (e.g., Witlox and Holt, 2000) still retain many approximations and their agreement with the available experimental data is questionable. It follows that for a first-attempt estimation of whether rainout will occur or not (which is the main information required for setting up the subsequent simulation of the atmospheric dispersion of the flammable cloud) simpler correlations would be desirable.

Among all the pressurized and refrigerated liquids in this paper the ammonia and the Liquefied Natural Gas (LNG) has been considered and the available experimental data of their rainout have been used to validate a short cut method for predicting rainout fraction.

## 2. Short-cut Rainout Correlation

A simple correlation to be used for predicting whether rainout is expected or not in a given situation should be able to account for the influence of the main physical and storage-related parameters influencing the liquid break up and the subsequent droplets evaporation (e.g., latent heat of vaporization, vapor pressure, liquid density and surface tension, drop-size, wind condition, ambient temperature, air entrainment).

A simple correlation has been developed to evaluate rainout occurrence, starting from the observation that rainout occurs when the fluid released after a loss of containment breaks out into droplets which take more time to evaporate (i.e. the evaporation time  $t_{evap}$ ) than to fall on the ground (i.e. flight time,  $t_{fl}$ ). This consideration leads to the following criteria for rainout existence:

$$\left\{ \begin{array}{l} \frac{t_{evap}}{t_{fl}} > 1 \\ \frac{t_{evap}}{t_{fl}} < 1 \end{array} \right. \quad \begin{array}{l} \eta_{rainout} > 0 \\ \eta_{rainout} = 0 \end{array} \quad (1)$$

Both the characteristic evaporation time and the characteristic flight time depend on droplets diameter. In turn, this characteristic dimension is determined by the mechanism that determines the liquid break up: shear stress (mechanic or aerodynamic breakage) or

liquid flash (thermodynamic breakage). The dominating mechanism depends on the physical characteristics of the liquid as well as on the storage conditions (De Vaull and King, 1992; Bricard and Friedel, 1998; Witlox et al. 2007), which therefore determine the characteristic droplet dimension and in turn the two characteristic times.

By comparing these two characteristic times (that is, when  $\frac{t_{evap}}{t_{fl}} = 1$ ) it is possible to

identify a threshold droplet diameter,  $D_{lim}$ , for rainout: droplets larger than this threshold will rainout, and the rainout fraction,  $\eta_{rainout}$ , will be roughly proportional to the difference between the droplet diameter and the threshold diameter, as follow:

$$\eta_{rainout} = \frac{(D_{drop} - D_{lim})}{D_{drop}} \quad (2)$$

The evaporation time for the droplet can be easily evaluated integrating the material balance and the energy balance for the evaporating droplet:

$$t_{evap} = \frac{1}{4} D_{drop}^2 \left( \frac{\Delta H_{evap} \rho_{liq}}{Sh \wp_{AB} (\rho_{\infty} - \rho_{sup}) h^v + Nu \ k (T_{drop} - T_{\infty})} \right) \quad (3)$$

where  $D_{drop}$  is Sauter mean droplet diameter,  $\Delta H_{evap}$  released liquid evaporation enthalpy,  $\rho_{liq}$  released liquid density,  $Sh$  Sherwood number,  $\wp_{AB}$  material diffusivity,  $\rho_{\infty}$  air density,  $\rho_{sup} = \frac{P_v(T)MW}{RT}$  is the droplet superficial density (MW molecular weight and R perfect gas constant),  $h^v$  vapor enthalpy,  $Nu$  Nusselt number,  $k$  thermal exchange coefficient,  $T_{drop}$  droplet temperature, and  $T_{\infty}$  air temperature.

Equation 3 shows that the evaporation time depends from the droplet diameter both directly and indirectly, for the presence of  $Sh$  and  $Nu$ .

The flight time of the droplet can be obtained by the integration of the momentum equation for the droplet along horizontal and vertical directions, applying the following assumptions:

- laminar conditions:  $C_D = \frac{24}{Re}$ ,
- constant vertical velocity:  $\frac{du_{drop,z}}{dt} = 0$ ,

The flight time for the droplet is instead evaluated as follow:

$$t_{fl} = \frac{z}{\frac{g(\rho_{liq} - \rho_{\infty})D_{drop}^2}{18\mu} + u_{\infty,z}} \quad (4)$$

Where  $z$  is the release height,  $g$  gravity acceleration,  $\mu$  the dynamic viscosity, and  $u_{\infty,z}$  vertical component of wind velocity  $u_{\infty}$ .

After having defined the evaporation time and the flight time for the droplet it is possible to evaluate their ratio, assuming that:

- the droplet velocity ( $u_{drop}$ ) is constant,
- the wind direction is horizontal ( $u_{\infty,z} = 0$ ),
- the droplet temperature ( $T_{drop}$ ) is constant,
- the release direction is horizontal.

Solving the simple relation  $t_{evap} - t_{fl} = 0$ , the limit droplet diameter is determined and then, knowing the initial droplet diameter, also the rainout fraction can be estimated.

The initial droplet diameter is evaluated choosing the smallest one between the two calculated considering both mechanical and aerodynamic fluid break-out.

The two correlations used to evaluate the diameter of droplets originated by mechanical ( $D_{mec}$ ) and aerodynamic ( $D_{aer}$ ) break-out are the following:

$$D_{mec} = \frac{\sigma We_{crit}}{u_f^2 \rho_{\infty}} \quad (5)$$

where  $\sigma$  is the surface tension, the critical Weber number  $We_{crit}=12.5$ , and  $u_f$  droplet velocity after jet expansion.

$$D_{aer} = 0.833 \cdot 10^{-3} - 0.0734 \cdot 10^{-3} \ln E_p \quad (6)$$

where

$$E_p = -\Delta H_{evap} - [P_v(T_{st}) - P_{\infty}]v_{st} + [P_{st} - P_v(T_{st})]v_{st} \quad (7)$$

is the partial expansion energy,  $P_v$  is the vapor pressure,  $T_{st}$  the storage temperature of the released fluid,  $P_\infty$  the pressure at the end of the expansion,  $P_{st}$  is the storage pressure, and  $v_{st}$  the specific volume of the released fluid at storage conditions.

### 3. Results

This simple approach has been validated comparing its results to those of other simulation models as well as to some available experimental data; in particular varying the height of the release, the storage temperature, and the storage pressure. Moreover, it was applied considering only a mean diameter, but it would define a limit diameter also in the case of a distribution of droplet diameters (i.e., droplets larger than the  $D_{lim}$  rainout); of course in this case, the formula should be weighted on the distribution function. For the sake of example, Figure 1 and Figure 2 compare the typical results obtained using this simple correlation for LNG and ammonia respectively, with the results of one of the more detailed models available (Witlox and Holt, 2000). We can see that the simple correlation is able to represent quite correctly the rainout fraction, and in particular it is able to foresee the presence or absence of rainout when some physical or operating parameter (the storage pressure in this case) changes.

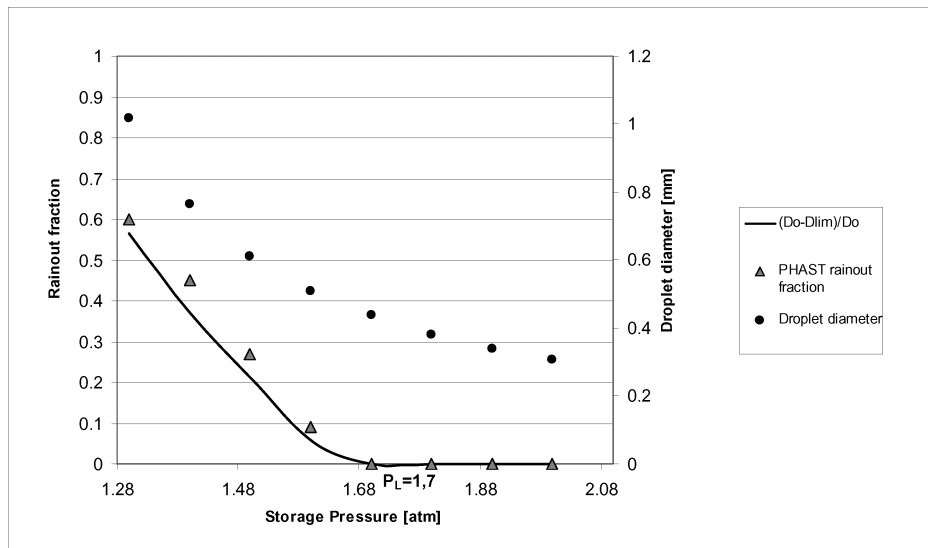


Figure 1 Rainout fraction and characteristic droplets diameter for a 1 m height release from a tank at constant temperature as a function of the storage pressure - LNG

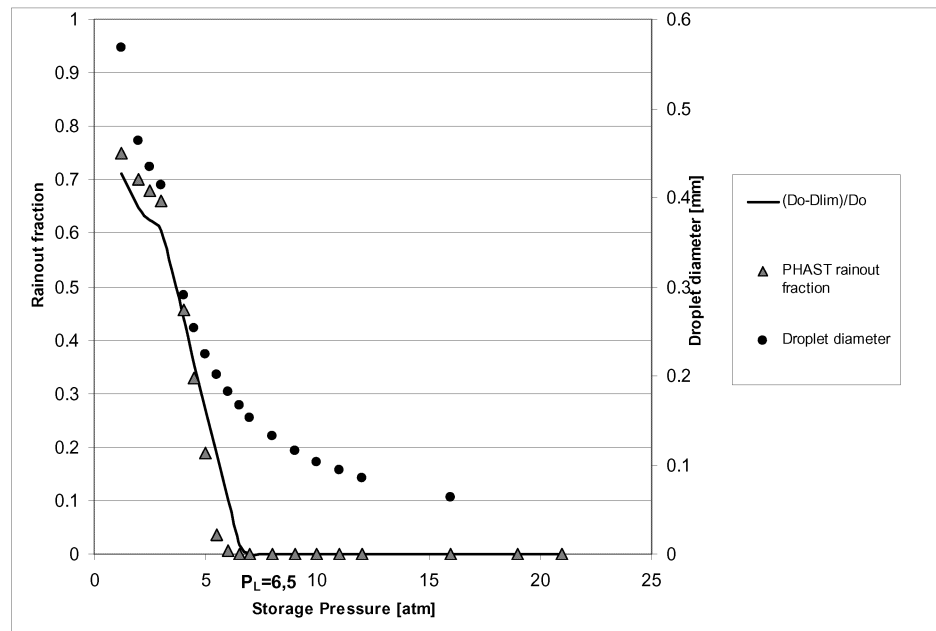


Figure 2 Rainout fraction and characteristic droplets diameter for a 1 m height release from a tank at constant temperature as a function of the storage pressure - ammonia

## References

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