

CRITERION FOR THE FORMATION OF A 'CLOUD-LIKE' RELEASE UPON DEPRESSURISATION OF A GAS VESSEL

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The initial stage of a single-phase gas escape from a depressurised vessel and the subsequent mixing with the ambient air is considered. A simple criterion is formulated which enables the release to be classified into the instantaneous (leading to formation of a gas cloud) or continuous (giving rise to a jet) type. The method is based on comparison of the characteristic time of the single-phase release (determined from the solution of the "internal" problem) with the characteristic times of turbulent mixing in an instantaneously released cloud, impulsively started transient jet as well as in a steady-state jet. Quantitative relationships defining the critical breach size are obtained for the cases of the gas escape from a high-pressure storage and from a pressure-supported gasholder. The most probable range of the fuel mass that can be involved in the burning fireball upon ignition of the gas released is assessed. The results are compared with available experimental data.

Key Words: Gas Release, Jet/Cloud Formation, Fireball, Source Terms

INTRODUCTION

The escape of explosive gases from pressurised storage vessels, pipes and vents into the atmosphere is one of the major dangers in the modern chemical industry.

Two main cases of release are normally considered in source models being used in the chemical process quantitative risk analysis (CPQRA) (e.g. AIChE/CCPS [1], TNO [2]). If the breach in the containment is relatively small the escaping gas forms a jet. This type of release is referred to as "continuous" and ignition of the escaping gas gives rise in this case to a jet fire. On the other hand, catastrophic failure of the pressurised tank leads to rapid ("instantaneous") escape of the substance into the atmosphere and to formation of a fuel-rich cloud which upon immediate ignition burns as a fireball (Marshall [3], NFPA [4]). Though the two limit cases of escape have been studied extensively, the criterion for classifying the release into the instantaneous or continuous type still remains mostly qualitative.

Ignition of flammable gas can lead to generation of shock waves and emission of thermal radiation, which can cause injury to people or damage nearby constructions. Overpressure is likely to be a principal hazard for a confined/congested environment, while strong heat radiation is one of the major hazards of ignition of the escaping gas in open space. In the case of the jet fire the burning rate is determined by supply of the fuel, and combustion proceeds until all the gas is burned. For a fireball the main parameter determining the geometry of the burning cloud, its duration and heat radiation flux is the initial mass of fuel

involved (e.g. Prugh [5]). For rapid (instantaneous) releases the gas is barely mixed with the air straight after the discharge and hence the mass of fuel in such a fireball can be assumed to be equal to the total mass of the stored gas. In the case of finite-duration releases the escaped gas still can form a cloud but mixing with the air reduces the fraction of the gas in the fuel-rich zone decreasing the hazards associated with the fireball. From this point of view a model for assessing the most probable range of fuel masses that can burn in the fireball would be very useful.

The purpose of this work is to develop a quantitative criterion that could be used in risk assessment practice to classify the single-phase gas release type into instantaneous or continuous given the initial parameters of the gas storage vessel and sizes of the breach. The method for the assessment of the critical conditions is based on comparison of the characteristic time of the release (determined from the solution of the "internal" problem) with the characteristic time of turbulent mixing of the escaping gas with the ambient air. The relationships defining the critical breach size as a function of the vessel geometry, gas properties and storage conditions are obtained. The method is applied to the analysis of the gas escape from a pressure-supported telescopic gasholder and from a high-pressure storage vessel. The most probable range of the fuel mass that can be involved upon immediate ignition in the burning fireball is assessed using the formulas obtained.

### CLASSIFICATION OF FINITE-DURATION RELEASES

Consider a flammable gas discharging through a circular breach of diameter  $d_b$  into quiescent atmosphere, the discharge velocity  $U_{out}$  being constant over the period of the release  $t_{out}$ . If the discharge time exceeds substantially the characteristic time of the turbulent mixing between the escaping gas and the ambient air we can expect that a jet will be formed and maintained during the release. In presence of an ignition source a jet fire will occur for such an escape. On the other hand, the gas released tends to form a fuel cloud if the discharge time is less than the characteristic time over which the gas mixes with the ambient atmosphere. In this case we can expect that only a minor fraction of the gas will be diluted below the lean flammability limit and upon immediate ignition almost all the gas can burn as a fireball. The two above-mentioned regimes are the limit cases between which intermediate releases occur (see Fig. 1).

To establish a criterion dividing the "puff" and jet releases we need to compare the total outflow time  $t_{out}$  with the characteristic mixing time relevant to dispersion and dilution of the released gas in the atmosphere. We assume that momentum dominates the buoyancy in the released cloud at least before the gas is diluted to combustible concentrations. We also neglect the influence of wind and consider here only unconfined releases.

Three flows can occur for the different release conditions and hence are relevant to the problem in question:

- instantaneously released gas cloud,
- time-dependent impulsively starting jet,
- steady turbulent jet.

For each of these flows we define the characteristic dilution time as the period after the release over which the concentration corresponding to the upper flammability limit (UFL) is achieved. This gives the time scale necessary for the released gas to become combustible.

The momentum-driven flow and dilution of the gas in an instantaneously released cloud can be described using the model developed by Landis *et al* [6] (a similar approach was offered earlier by Hardee and Lee [7], Roberts [8], the details of this model can be found in Appendix A). Using the temporal dependence of the average concentration of the gas in the cloud we obtain that for an instantaneous release the UFL is achieved in the cloud after the time (see the notation below):

$$t_{UFL}^{(cloud)} = \frac{d_b}{2U_{out}} \left( \frac{\rho_b}{\rho_a} \right)^{1/3} \left( \frac{\mu_a}{\mu_g} \right)^{4/3} C_{UFL}^{-4/3} \quad (1)$$

The dilution of the gas in a transient jet can be described using the approach offered in [7], Phillips [9] (see Appendix B). The characteristic time of the dilution to the upper flammability limit in the impulsively started transient jet is

$$t_{UFL}^{(\text{trans jet})} \approx (2.2 - 3.4) \frac{d_b}{U_{out}} \left( \frac{\rho_b}{\rho_a} \right)^{1/2} \left( \frac{\mu_a}{\mu_g} \right)^2 C_{UFL}^{-2} \quad (2)$$

Finally, the characteristic mixing time relevant to steady axisymmetric jets can be shown to be (see Appendix C)

$$t_{UFL}^{(\text{steady jet})} \approx 1.4 \frac{d_b}{U_{out}} \left( \frac{\rho_b}{\rho_a} \right)^{1/2} \left( \frac{\mu_a}{\mu_b} \right)^2 C_{UFL}^{-2} \quad (3)$$

It can be seen from (2) and (3) that both types of jet outflow have the same functional dependence of the characteristic time on the parameters of the gas and on the geometry and the velocity of the discharge, the constants differ only by a factor of about 2. In what follows we use a characteristic mixing time defined as

$$t_{UFL}^{(\text{jet})} = \frac{2d_b}{U_{out}} \left( \frac{\rho_b}{\rho_a} \right)^{1/2} \left( \frac{\mu_a}{\mu_b} \right)^2 C_{UFL}^{-2} \quad (4)$$

which is relevant to both kinds of jet releases.

Compare now the jet and cloud characteristic mixing times. Dividing Equation (1) by (4) we obtain that

$$\frac{t_{UFL}^{(\text{cloud})}}{t_{UFL}^{(\text{jet})}} = \frac{1}{4} \left( \frac{\rho_a}{\rho_b} \right)^{1/6} \left( \frac{\mu_g}{\mu_a} \right)^{2/3} C_{UFL}^{2/3} \ll 1$$

because the values of  $C_{UFL}$  are usually small (about 0.05 - 0.2 for typical hydrocarbons). This means that the gas dilution in the instantaneously released cloud proceeds much more intensively than in the jet and the time necessary for the cloud to be diluted to the combustibility limits is normally shorter than that in the case of the jet outflow. We offer the following classification scheme for the gas releases:

- $t_{out} \geq t_{UFL}^{(\text{jet})}$  : jet ("continuous") release;
- $t_{out} \leq t_{UFL}^{(\text{cloud})}$  : cloud ("instantaneous") release;
- $t_{UFL}^{(\text{cloud})} \leq t_{out} \leq t_{UFL}^{(\text{jet})}$  : intermediate ("cloud-like") release

The latter type of the release corresponds to the case where the gas escapes rapidly enough so that the steady jet can not form over the release time, but on the other hand the escape is slow enough so that some part of the gas mixes with the air reducing thus the mass in the fuel-rich zone.

### CRITICAL SIZE OF THE BREACH

The criterion (5) defining the type of the finite-duration release can be reformulated in terms of the conditions on the breach diameter  $d_b$ . Given the released mass  $M$ , the outflow conditions  $\rho_b$ ,  $U_{out}$  and the size of the breach  $d_b$ , the total discharge time can be expressed as  $t_{out} = 4M / C_d \pi d_b^2 \rho_b U_{out}$ . By equating this time to the characteristic mixing time for the cloud and jet releases (1), (4) and solving for the diameter  $d_b$  we find the following two critical breach diameters: the diameter  $d_c$  dividing the instantaneous and cloud-like releases

$$d_c^3 = \frac{8M}{C_d \pi \rho_b} \left( \frac{\rho_a}{\rho_b} \right)^{1/3} \left( \frac{\mu_g}{\mu_a} \right)^{4/3} C_{UFL}^{4/3} = \frac{8M}{C_d \pi \rho_{g,a}} \left( \frac{\rho_{g,a}}{\rho_b} \right)^{4/3} \left( \frac{\mu_g}{\mu_a} \right)^{4/3} C_{UFL}^{4/3} \quad (6)$$

and the diameter  $d_j$  dividing the cloud-like and jet releases

$$d_j^3 = \frac{2M}{C_d \pi \rho_b} \left( \frac{\rho_a}{\rho_b} \right)^{1/2} \left( \frac{\mu_g}{\mu_a} \right)^2 C_{UFL}^2 = \frac{2M}{C_d \pi \rho_{g,a}} \left( \frac{\rho_{g,a}}{\rho_b} \right)^{3/2} \left( \frac{\mu_g}{\mu_a} \right)^{3/2} C_{UFL}^2 \quad (7)$$

where the equality  $\rho_{g,a}/\rho_a = \mu_g/\mu_a$  was used. We also introduce the actual breach area  $S_b = \pi d_b^2/4$  and critical breach areas corresponding to the above critical diameters  $S_c = \pi d_c^2/4$  and  $S_j = \pi d_j^2/4$ . These areas can be used together with the diameters to characterise the breach. In terms of the breach sizes the criterion for different types of the release (5) is presented as

- $d_b \leq d_j$  (or  $S_b \leq S_j$ ) : jet (“continuous”) release;
- $d_b \geq d_c$  (or  $S_b \geq S_c$ ) : cloud (“instantaneous”) release;
- $d_j \leq d_b \leq d_c$  (or  $S_j \leq S_b \leq S_c$ ) : intermediate (“cloud-like”) release

This form of the criterion is more suitable for risk analysis practice than the relationship between the characteristic times (5) because it enables the release type to be determined from some well-defined values - geometry of the reservoir, properties of the gas and the storage conditions. The overall classification scheme is sketched in Fig. 2. We note that the criterion (8) can also be applied to non-circular breaches in which case the area  $S_b$  should be used as the primary characteristic of the breach while the effective breach diameter can be introduced as  $d_b = (4S_b/\pi)^{1/2}$ .

### MINIMUM MASS OF FUEL IN THE FIREBALL

When the gas is being released into the atmosphere through a breach, a distribution of concentration is being formed in which the fuel-rich zone (with the concentration of the fuel exceeding the UFL) is surrounded by a zone of combustible mixture in which the gas is diluted so that its concentration is between the flammability limits (see Fig. 1). If ignited immediately, the mixture between the lower and upper flammability limits is expected to burn quickly as a flash fire, while the fuel-rich core will burn more slowly (in the diffusion regime) as a fireball. So, the minimum mass of fuel in the fireball could be assessed to be equal to that in the fuel-rich zone surrounded by the surface on which the fuel concentration is  $C_{UFL}$ .

If the release is instantaneous ( $t_{out} \leq t_{UFL}^{(cloud)}$  or  $d_b \geq d_c$ ) the gas is barely mixed with air immediately after the depressurisation. Considering the worst-case scenario one can assume that the mass of fuel in a fireball after a catastrophic failure of the gas vessel is equal to the total mass of the gas stored.

To establish the lower boundary of probable fuel masses that can burn in a fireball in the case of the “cloud-like” releases ( $t_{UFL}^{(cloud)} \leq t_{out} \leq t_{UFL}^{(jet)}$  or  $d_j \leq d_b \leq d_c$ ) consider the critical case where the outflow time is exactly equal to the characteristic jet mixing time, i.e. the diameter of the breach is equal to  $d_j$  from (7).

For this critical breach size the formation of the jet finishes just at the moment of the termination of the discharge, and to get an estimate of the gas mass in the fuel-rich zone we may use the spatial concentration distributions relevant to the steady jets (the details are given in Appendix D). Upon substitution of the critical diameter  $d_j$  from (7) into Equation (24) we get the critical fuel volume

$$V_{UFL} = \frac{\alpha^2 B^3}{3C_d} \left( \frac{\rho_a}{\rho_b} \right)^{1/2} \left( \frac{\mu_g}{\mu_a} \right)^{1/2} \frac{M}{\rho_b} = \frac{\alpha^2 B^3}{3C_d} \left( \frac{\rho_{g,a}}{\rho_b} \right)^{3/2} \frac{M}{\rho_{g,a}} \approx 0.5 \left( \frac{\rho_{g,a}}{\rho_b} \right)^{3/2} \frac{M}{\rho_{g,a}} \quad (9)$$

where the experimental values of the jet angle  $\alpha \cong 0.126$  (Moodie and Ewan [10]) and the concentration decay factor  $B = 4.3$  (Abramovich *et al* [11], Dowling and Dimotakis [12]) were used for calculation of the constant, the discharge coefficient  $C_d = 0.85$  [1].

The mass of the fuel in the fuel-rich zone is  $M_{UFL} = \rho_{g,a} V_{UFL}$  and we finally assess the minimum mass of fuel in the fireball as

$$M_{FB} = M_{UFL} \approx 0.5 \left( \frac{\rho_{g,a}}{\rho_b} \right)^{3/2} M \quad (10)$$

### OUTFLOW FROM LOW- AND HIGH-PRESSURE VESSELS

In the above formulas for the critical breach sizes (6), (7) and for the volume and mass of the gas in the fuel-rich zone (9), (10) the exit density of the gas  $\rho_b$  depends on storage conditions and on the regime of outflow.

If the storage pressure is low, the exit pressure is equal to the atmospheric pressure and the exit temperature only slightly differs from ambient. In this case the density of the escaping gas  $\rho_b$  is approximately equal to the density of the gas at the ambient conditions  $\rho_{g,a}$ . We get then the following relationships for the critical parameters:

$$d_c^3 = \frac{8M}{C_d \pi \rho_{g,a}} \left( \frac{\mu_g}{\mu_a} \right) C_{UFL}^{4/3} \quad (11)$$

$$d_j^3 = \frac{2M}{C_d \pi \rho_{g,a}} \left( \frac{\mu_g}{\mu_a} \right)^{3/2} C_{UFL}^2 \quad (12)$$

$$M_{FB} \approx 0.5 M \quad (13)$$

If the storage pressure in a vessel exceeds critical (which is about twice the atmospheric pressure), the discharge proceeds at sonic velocity (so-called choked flow), in this case the exit pressure is above ambient (see Appendix E). Pressure equalisation occurs in the immediate vicinity to the orifice, after which the flow is actually isobaric. To apply the above critical relationships to the choked outflow we make use of the "equivalent diameter" concept widely accepted for the description of underexpanded jets (e.g. [2], [10], Birch *et al* [13], Ewan and Moodie [14]). The equivalent diameter  $d_{eq}$  and the equivalent density  $\rho_{eq}$  are defined so that the mass flux through the orifice of this diameter is equal to the actual mass flux:

$$\frac{\pi d_b^2}{4} \rho_b U_{out} = \frac{\pi d_{eq}^2}{4} \rho_{eq} U_{out}, \quad \rho_{eq} = \rho_b \frac{P_a}{P_b}$$

Thus, for choked outflow from a high-pressure vessel the density of the escaping gas  $\rho_b$  in the relationships (1) - (7) should be substituted by the equivalent density  $\rho_{eq}$  and the geometric diameter of the breach  $d_b$  be substituted by the equivalent diameter  $d_{eq} = d_b (\rho_b / \rho_{eq})^{1/2} = d_b (P_b / P_a)^{1/2}$

To allow for the transient discharge rate we use the average pressure  $\bar{P}$  and density  $\bar{\rho}$  from Appendix E, Equation (28), to calculate the exit pressure and density  $P_b$  and  $\rho_b$  (see (26)) and apply the approximation (27) for the average/initial pressure ratio  $\eta$ . As a result we obtain the critical breach diameter  $d_c$ .

$$d_c^3 = \frac{8M}{C_d \pi \rho_{g,a}} \left( \frac{\rho_{g,a}}{\rho_b} \right)^{4/3} \left( \frac{\mu_g}{\mu_a} \right) C_{UFL}^{4/3} \left( \frac{P_a}{P_b} \right)^{1/6} = \frac{8M}{C_d \pi \rho_{g,a}} \left( \frac{k+1}{2} \right)^{\frac{8+k}{6(k-1)}} \left( \frac{\mu_g}{\mu_a} \right) C_{UFL}^{4/3} \left( \frac{P_a}{\eta P_0} \right)^{3/2} \approx \quad (14)$$

$$\approx \frac{13.2M}{\rho_{g,a}} \left( \frac{\mu_g}{\mu_a} \right) C_{UFL}^{4/3} \left( \frac{P_a}{P_0} \right)^{5/4} = 13.2 \left( \frac{\mu_g}{\mu_a} \right) C_{UFL}^{4/3} \left( \frac{P_a}{P_0} \right)^{1/4} V_0$$

and the diameter  $d_j$  :

$$d_j^3 = \frac{2M}{C_d \pi \rho_{g,a}} \left( \frac{\rho_a}{\rho_b} \right)^{3/2} \left( \frac{\mu_g}{\mu_a} \right)^{3/2} C_{UFL}^2 = \frac{2M}{C_d \pi \rho_{g,a}} \left( \frac{k+1}{2} \right)^{\frac{3}{2(k-1)}} \left( \frac{\mu_g}{\mu_a} \right)^{3/2} C_{UFL}^2 \left( \frac{P_a}{\eta P_0} \right)^{3/2} \approx \quad (15)$$

$$\approx \frac{3.2M}{\rho_{g,a}} \left( \frac{\mu_g}{\mu_a} \right)^{3/2} C_{UFL}^2 \left( \frac{P_a}{P_0} \right)^{3/4} = 3.2 \left( \frac{\mu_g}{\mu_a} \right)^{3/2} C_{UFL}^2 \left( \frac{P_a}{P_0} \right)^{1/4} V_0$$

where  $V_0 = M/\rho_0$  is the volume of the vessel, the constants were calculated for  $k = 1.4$ ,  $C_d = 0.85$ .

The volume of the gas in the fuel-rich zone and the minimum mass of fuel in the fireball are obtained for the high-pressure releases from (9), (10) as

$$V_{UFL} \approx 0.5 \left( \frac{\rho_{g,a}}{\rho_b} \frac{P_b}{P_a} \right)^{3/2} \frac{M}{\rho_{g,a}} = 0.5 \left( \frac{2}{k+1} \right)^{3/2} \frac{M}{\rho_{g,a}} \approx 0.38 \frac{M}{\rho_{g,a}}, \quad M_{FB} = \rho_{g,a} V_{UFL} \approx 0.38M \quad (16)$$

### EXAMPLE CALCULATIONS

We give here two examples of the application of the relationships obtained. First we assess the critical breach size and find the possible fuel mass in the fireball that can occur if a pressure-supported gasholder with the natural gas (NG) is damaged. Then the escape of NG from a high-pressure storage vessel is considered.

#### Low Pressure Gasholder

Pressure-supported gasholders are widely used for the storage of the natural gas. The gasholder consists of several telescopically moving sections so that the height of the gasholder varies with actual quantity of the gas within it. A relatively low overpressure (about 20 mbar) exists permanently within the gasholder supporting its sections above the ground. If the roof of the gasholder is damaged, the holder will deflate as its sections will move down telescopically until all the gas stored escapes into the atmosphere. The pressure difference (defined by the weight of the roof) can be assumed to be constant during the deflation which means that the outflow rate is also maintained approximately constant.

Since the storage pressure is only slightly higher than the ambient one and the temperature within the gasholder is equal to the atmospheric temperature, the density of the escaping gas is approximately equal to its density at the ambient conditions, i.e.  $\rho_b \cong \rho_{g,a}$ . The breach diameter  $d_c$  for which the total outflow time  $t_{out}$  is equal to the characteristic time of the dilution in the instantaneously released cloud  $t_{UFL}^{(cloud)}$  is (see Equation (11)):

$$d_c^3 = \frac{8M}{C_d \pi \rho_{g,a}} \left( \frac{\mu_g}{\mu_a} \right) C_{UFL}^{4/3} \approx 0.14V_0$$

where  $M$  is the total mass of the gas escaped (which for the deflating gasholder coincides with the total mass stored),  $V_0$  is the volume of the gasholder before the depressurisation. Upon the substitution of the parameters of the natural gas ( $\mu_0 = 17 \text{ kg/kmole}$ ,  $\rho_0 = 0.715 \text{ kg/m}^3$  at  $T_a = 293 \text{ K}$ ,  $C_{UFL} = 15\% \text{ vol.}$ ) and of the air ( $\mu_a = 29 \text{ kg/kmole}$ ) we get finally the diameter  $d_c$  and the area  $S_c$  of the breach:

$$d_c = 0.52V_0^{1/3}, \quad S_c = \frac{\pi d_c^2}{4} = 0.21V_0^{2/3}$$

A typical gasholder filled to its full capacity has the height  $H_0$  approximately equal to  $2/3$  of its diameter  $D_0$ , so that  $V_0 = \frac{1}{4}\pi D_0^2 H_0 \approx \frac{1}{6}\pi D_0^3$ . The ratio of the damaged area corresponding to the above diameter and the total roof area  $S_0 = \pi D_0^2 / 4$  is

$$S_c/S_0 = (d_c/D_0)^2 \approx 0.14$$

Thus, the release can be considered as instantaneous if the damaged area exceeds 14% of the roof area.

The critical breach diameter  $d_j$  and corresponding damaged area  $S_j$  dividing the jet and “cloud-like” releases can be found from (12)

$$d_j^3 = \frac{2M}{C_d \pi \rho_{g,a}} \left( \frac{\mu_g}{\mu_a} \right)^{3/2} C_{UFL}^2 \approx 7.6 \cdot 10^{-3} V_0 \quad \text{or} \quad d_j \approx 0.20V_0^{1/3},$$

$$S_j = \frac{1}{4}\pi d_j^2 = 0.03V_0^{2/3}, \quad S_j/S_0 = (d_j/D_0)^2 \approx 0.025$$

and the minimum mass of fuel in the fireball for this case is assessed from (13) as  $M_{FB} \approx 0.5M$

To be more specific we calculate the above parameters for the gasholder with the diameter  $D_0 = 30 \text{ m}$ , height  $H_0 = 20 \text{ m}$  and the volume  $V_0 = 1.4 \cdot 10^4 \text{ m}^3$ , the mass of the natural gas stored is  $M = 10^4 \text{ kg}$ . We may see that if the diameter of the breach is less than  $d_j = 0.2 \cdot (1.4 \cdot 10^4)^{1/3} = 4.8 \text{ m}$  (the damaged area  $S_j = 18.2 \text{ m}^2$ ), the escaping gas will form a turbulent jet and upon ignition a jet fire will occur. If the diameter of the breach is more than  $d_c = 0.52 \cdot (1.4 \cdot 10^4)^{1/3} = 12.5 \text{ m}$  (the damaged area  $S_c = 123 \text{ m}^2$ ) a cloud of the escaping gas will be formed and upon ignition it can give rise to the fireball with fuel mass of up to  $10^4 \text{ kg}$ . In the intermediate cases, when the diameter of the opening is between 4.8 and 12.5 m, a “cloud-like” release is expected, a minimal mass of fuel in the fireball is  $M_{FB} = 0.5 \cdot 10^4 = 5000 \text{ kg}$ . The range  $(5-10) \cdot 10^3 \text{ kg}$  of the fuel mass can be used in the risk assessment as an input data for the effect models [1 - 4]. We note finally that the above calculations are valid for telescopic gasholders only for which the escape of the gas is followed by decrease in the volume of the holder maintaining the positive overpressure inside the vessel. If the vessel is not telescopic or fails to telescope down because of some mechanical damage, the pressure-driven release will proceed until pressure equalisation, after which diffusive counterflow will govern the release. The above model does not apply to this case.

### High-Pressure Gas Vessel

Consider a storage vessel filled with the natural gas (NG) at high initial pressure  $P_0 \geq 10 \text{ bar}$ . Substitution of the properties of the natural gas (see above) into (14), (15) gives the following critical sizes of the breach:

$$d_c^3 = 13.2 \left( \frac{\mu_g}{\mu_a} \right) C_{UFL}^{3/2} \left( \frac{P_a}{P_0} \right)^{1/4} V_0 \approx 0.62 \left( \frac{P_a}{P_0} \right)^{1/4} V_0$$

$$d_j^3 = 3.2 \left( \frac{\mu_g}{\mu_a} \right)^{3/2} C_{UFL}^2 \left( \frac{P_a}{P_0} \right)^{1/4} V_0 \approx 0.032 \left( \frac{P_a}{P_0} \right)^{1/4} V_0$$

The minimum mass of fuel that can burn in the fireball (see (16)) is  $M_{FB} \approx 0.38M$ .

We calculate now the critical breach sizes and the minimum mass of fuel in a fireball for a vessel of the volume  $V_0 = 100 \text{ m}^3$  at two storage pressures  $P_0 = 20 \text{ bar}$  (the total mass of the NG is  $M = 1.4 \cdot 10^3 \text{ kg}$ ) and  $100 \text{ bar}$  ( $M = 7 \cdot 10^3 \text{ kg}$ ).

In the first case we obtain that the jet outflow can occur if the breach diameter is less than  $d_j = 1.1 \text{ m}$  (the damaged area  $S_j = 1.0 \text{ m}^2$ ) and the cloud will escape if the breach diameter exceeds  $d_c = 3.1 \text{ m}$  (the damaged area  $S_c = 7.5 \text{ m}^2$ ). The lower boundary for the fuel mass that can be involved in the fireball is assessed as  $M_{FB} \approx 0.38 \cdot (1.4 \cdot 10^3) = 530 \text{ kg}$ . In the second case we obtain  $d_c = 2.7 \text{ m}$  (the damaged area  $S_c = 5.7 \text{ m}^2$ ),  $d_j = 1.0 \text{ m}$  (the damaged area  $S_j = 0.8 \text{ m}^2$ ) and  $M_{FB} \approx 0.38 \cdot (7 \cdot 10^3) = 2700 \text{ kg}$ . Thus, in the first case as much as  $(0.53-1.4) \cdot 10^3$  and in the second case  $(2.7-7.0) \cdot 10^3 \text{ kg}$  of the NG can be involved in the fireball on instantaneous ignition. We see that the critical breach sizes depend on the storage conditions quite weakly, so that their typical values can be calculated given the typical storage conditions and the geometry of the reservoir.

## DISCUSSION AND CONCLUSIONS

In the current paper a quantitative criterion for the classification of the release type into instantaneous, continuous or intermediate is offered. The criterion is derived from the analysis of the mixing of the escaping gas with the ambient air in typical situations (in an instantaneously released cloud, in an impulsively started transient jet and in a steady jet) and comparison of the characteristic mixing times with the outflow time.

In each of these cases experimentally verified models of gas dilution were used so that the data on the characteristic times can be considered as quite reliable. The criterion as a whole still needs the experimental verification for which a comprehensive set of data on outflow from a finite-volume vessel and on behaviour and parameters of the resulting cloud would be necessary.

The best-studied types of the outflow are the continuous jets issuing from a relatively small orifice and, on the other hand, the instantaneous releases (including the discharge of the liquefied gases and BLEVE) upon the total loss of containment. Only limited attempts have been made to investigate the intermediate releases, nevertheless the published data can be used to check at least the consistency of the criterion.

Chaineaux *et al* [15, 16] studied the outflow of the methane from a  $120 \text{ dm}^3$  vessel. The orifice diameters were 6, 12 and 24 mm, the initial pressure of the gas was 10 MPa. Substitution of the volume, the pressure and the UFL concentration into Equation (15) gives the critical diameter  $d_j = 10 \text{ cm}$ , which is well above the orifice sizes used in the experiments. Hence, for all the orifices our criterion predicts the jet outflow. This is quite consistent with the observations reported: in all the tests a jet developed over the first few seconds after which the jet persisted for several dozens of seconds changing with time to follow the continuously decreasing pressure in the vessel.

Another type of the discharge was reported in [6] where rapid releases were modelled. The plumes were generated using a "cannon" consisting of a 30.5 cm long, 10.2 cm diameter pipe with a hemispherical cap on one end and a rupture disk holder at the other, the volume of the vessel was  $2750 \text{ cm}^3$ . The burst pressures of the rupture disks were 3.22, 7.17, 20.52 and 70.69 bar. In the experiments the cannon was filled with mixture of the nitrogen and a tracer enabling video recording of the generated plume. All experiments reported showed the formation of an elongated cloud so that the authors referred to the release type as instantaneous. Since the test gas was non-flammable it is impossible to use our criterion directly, but for the sake of checking the consistency of the criterion we calculated the critical diameters  $d_c$  assuming the UFL concentration to be that of methane (15% vol.), this value is also typical for other hydrocarbons.



The diameters obtained from Equation (14) for the above pressures were 10.4, 9.8, 8.9, 7.9 cm correspondingly, i.e. the orifice diameter used in the experiments was above (except for the lowest pressure) the critical diameter dividing the cloud and “cloud-like” releases.

Thus, though being very limited, this comparison of the predicted values with the experimental data shows that the criterion does not contradict the available observations. Besides, the values of the critical breach sizes obtained for both the low-pressure gasholder and the high-pressure vessel in the examples above appear to be quite reasonable and consistent with the intuitively clear notions of the “small hole” and “catastrophic damage”.

Further experimental study of releases from storage and process vessels would be desirable to establish the accuracy and applicability limits of this criterion. The experiments could comprise releases of different gases from several vessels of different volumes and at diverse exit hole diameters. The representative parameters that can be used to characterise the outflow conditions and the properties of the gas are:

$$\Psi = \frac{d}{V_0^{1/3}} \left( \frac{P_0}{P_a} \right)^{1/2}, \quad \xi = \left( \frac{\mu_g}{\mu_a} \right)^{1/3} C_{UFL}^{4/9}$$

where for the low-pressure releases the outflow parameter  $\Psi$  reduces simply to  $d/V_0^{1/3}$ . In these coordinates the critical condition dividing the cloud and cloud-like outflow is  $\Psi = 1.44 \xi$  for low-pressure and  $\Psi = 2.36 \xi$  for high-pressure releases. The critical condition dividing the cloud-like and jet outflows is  $\Psi = 0.91 \xi^{3/2}$  for low-pressure and  $\Psi = 1.47 \xi^{3/2}$  for high-pressure releases (see Equations (11) - (16)). In Fig. 3 the critical conditions are presented in coordinates  $(\xi, \Psi)$  for both low- and high-pressure releases. The upper (straight) line divides the cloud and cloud-like outflows, the lower line corresponds to the boundary between the cloud-like and jet releases. The left vertical axis relates to low-pressure outflows while the right one corresponds to high-pressure releases.

Measurement of the parameters of escaping clouds could give the data necessary to classify the release as continuous, puff or cloud-like, the boundaries between these regimes could be compared with the current predictions plotted in Fig. 3. The values of the parameter  $\xi$  characterising the properties of the gas (its molecular mass and UFL concentration) were calculated for some widely used gases - hydrogen, volatile hydrocarbons and ammonia. These values are presented in Fig. 3 by vertical lines drawn for corresponding abscissa values. It can be seen that for the above substances the values of  $\xi$  fall between 0.3 and 0.7, this range seems to be the most important and has to be studied in the experiments.

We finally note that the criterion  $d/V_0^{1/3}$  should be important for the statistical analysis of accidents on pipelines and storage vessels. Also, the approach offered in this study for single-phase releases can be extended to include the two-phase releases of liquefied gases, which would be essential for quantitative risk analysis practice.

#### ACKNOWLEDGEMENT

The authors would like to thank the EPSRC (grant GR/K 13486) and the Royal Society for support of this study.

NOTATION

<i>a</i>	-	shape factor in the cloud model, [-],
<i>A</i>	-	constant in the axial velocity decay law, [-],
<i>B</i>	-	constant in the axial mass concentration decay law, [-],
<i>C</i>	-	volume (molar) concentration, [-],
<i>d, D</i>	-	diameter, [m],
<i>G</i>	-	outflow mass rate, [kg/s],
<i>h</i>	-	height of the cone in the integral model of jet, [m],
<i>k</i>	-	ratio of the specific heats, [-],
<i>M</i>	-	mass of the gas, [kg],
<i>P</i>	-	pressure, [Pa],
<i>r</i>	-	radius, [m],
<i>R = 8.31</i>	-	universal gas constant, [J/mole·K],
<i>S</i>	-	breach area, [m <sup>2</sup> ],
<i>T</i>	-	temperature, [K],
<i>V</i>	-	volume, [m <sup>3</sup> ],
<i>Y</i>	-	mass concentration, [-],

Greek

<i>α</i>	-	jet cone angle in the Gaussian model, [-],
<i>β</i>	-	cone angle for the instantaneously started jet, [-],
<i>μ</i>	-	molecular weight, [kg/kmole],
<i>η</i>	-	ratio of the average and initial discharge rates, [-],
<i>ρ</i>	-	density, [kg/m <sup>3</sup> ],

Subscripts

<i>0</i>	-	initial conditions,
<i>a</i>	-	ambient conditions,
<i>ax</i>	-	axial value,
<i>b</i>	-	breach,
<i>C</i>	-	value relevant to cloud formation,
<i>eq</i>	-	equivalent diameter and velocity,
<i>FB</i>	-	fireball
<i>g</i>	-	gas,
<i>g,a</i>	-	density of the gas at ambient conditions,
<i>J</i>	-	value relevant to jet formation,
<i>out</i>	-	outflow,
<i>UFL</i>	-	upper flammability limit,

Superscripts

-	-	average value.
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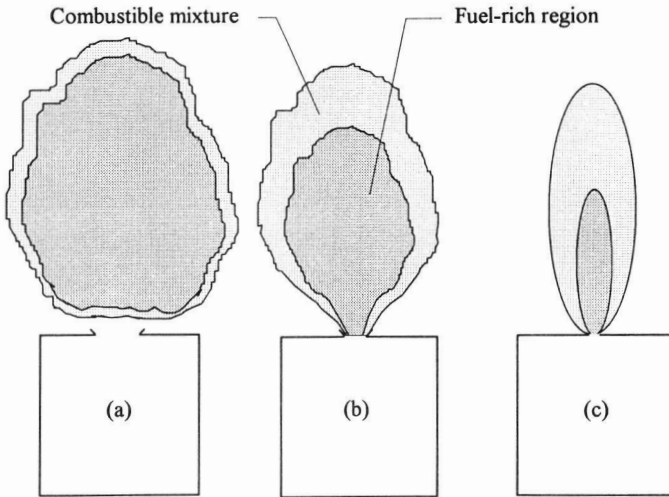


Figure 1. Possible types of finite-duration releases : a - cloud, b - "cloud-like", c - jet

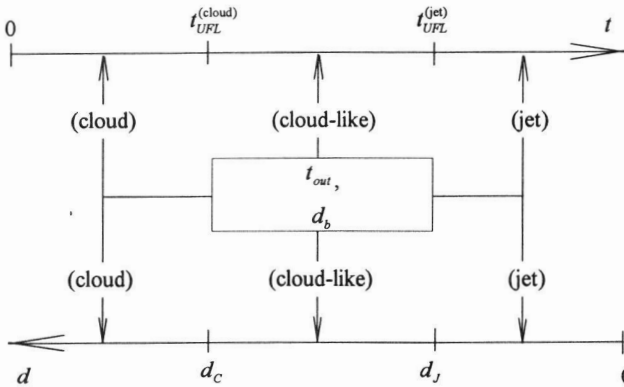


Figure 2. Criterion for realisation of different types of outflow as a relationship between the outflow time  $t_{out}$  and the characteristic times  $t_{UFL}^{(cloud)}$  and  $t_{UFL}^{(jet)}$  or, equivalently, as a relationship between the breach diameter  $d_b$  and the critical diameters  $d_c$  and  $d_j$

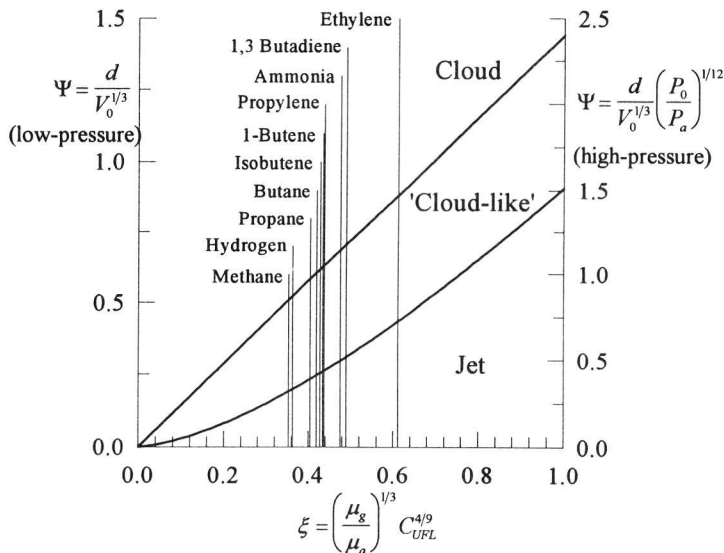


Figure 3. Boundaries between the jet, cloud-like and cloud releases. The abscissa  $\xi$  defines the properties of the gas, the ordinate  $\Psi$  describes the outflow conditions. The left ordinate axis corresponds to low-pressure releases, the right one - to high-pressure outflow. The vertical lines present the values of  $\xi$  for different gases.

APPENDICES

A. Dilution in an instantaneously released gas cloud

The dynamics of the gas and air mixing in a cloud formed after a very short duration release was studied in [6] experimentally, a model for dilution of the instantaneous release was also offered and compared with the measured data. This model is based on the assumption that the total momentum of the gas cloud remains constant as the cloud mixes with the ambient air due to frontal capture [7]. A very similar approach was also used in [8] to model the instantaneous release of liquefied gas giving rise to a hemispherical cloud.

The following relationship for the increase in the volume of the released cloud was obtained in [6]:

$$\frac{V}{V_b} = \left( \frac{4}{3a} \frac{\rho_b}{\rho_a} \frac{U_{out} t}{r_b} \right)^{3/4} \tag{17}$$

where  $a$  is the shape factor relating the volume of the cloud  $V$  to its equivalent radius  $r$  so that  $V = a\pi r^3$ . The experiments showed that a good correlation of the model prediction with the measured data is achieved if the value of  $a = 4/3$  is used, so that the term  $4/3a$  becomes 1 in (17).

We use this model to assess the characteristic time of the dilution in the released cloud which we define as a period over which the given molar (volume) concentration  $C$  of the gas is achieved within the cloud. The temporal dependence of the volume concentration  $C$  in the cloud is

$$C = \frac{\rho_g \mu_a}{\rho_a \mu_g} = \frac{\rho_b V_b \mu_a}{\rho_a V \mu_g} = \left( \frac{\rho_b}{\rho_a} \right)^{1/4} \left( \frac{\mu_a}{\mu_g} \right) \left( \frac{U_{out} t}{r_b} \right)^{-3/4}$$

where  $\rho_g$  is the partial density of the gas in the cloud. We find then that the time necessary for the cloud to be diluted to the volume concentration  $C$  is

$$t_C^{(cloud)} = \frac{d_b}{2U_{out}} \left( \frac{\rho_b}{\rho_a} \right)^{1/3} \left( \frac{\mu_a}{\mu_g} \right)^{4/3} C^{-4/3} \quad (18)$$

### B. Dilution of the gas in a transient jet (integral model)

An impulsively started jet can be described on the basis of the approach developed in [7] for liquefied gases, the only difference with the current model is that in [7] a wedge-shaped cloud was considered while we analyse here a cone-shaped jet.

We represent the jet by an expanding cone of radius  $r$  and height  $h$ , so that  $r = \beta h$  and the volume of the jet is  $V = \pi r^2 h / 3 = \pi r^3 / 3\beta$ . By equating the momentum of this cone  $3/2 \rho U V$  to the total momentum  $\pi d_b^2 / 4 \rho_b U_{out}^2 t$  released by the time  $t$  and expressing the velocity of the jet as  $U = dh/dt = dr/\beta dt$  we obtain the following equation:

$$\frac{3}{2} \left( \frac{1}{3\beta} \pi \rho_a r^3 \right) \frac{1}{\beta} \frac{dr}{dt} = \left( \frac{\pi d_b^2}{4} \rho_b U_{out}^2 \right) t$$

We find then that the length of the transient jet increases with time according to the square-root law

$$\frac{h}{d_b} = \beta^{-1/2} \left( \frac{\rho_b}{\rho_a} \right)^{1/4} \left( \frac{U_{out} t}{d_b} \right)^{1/2}$$

This dependence correlates well with the experimental data on transient impulsively started jets obtained by the image processing method in Shirakashi *et al* [17]. The volume of the starting jet depends on time as

$$V(t) = \frac{\pi}{3} \left( \frac{\rho_b}{\rho_a} \right)^{3/4} \beta^{1/2} (U_{out} d_b t)^{3/2}$$

The mass of fuel released by the time  $t$  is  $M(t) = \frac{\pi d_b^2}{4} \rho_b U_{out} t$  and the average volume concentration of the gas in the cloud is

$$C = \frac{M(t)}{\rho_a V(t)} \left( \frac{\mu_a}{\mu_g} \right) = \frac{3}{4\beta^{1/2}} \left( \frac{\rho_b}{\rho_a} \right)^{1/4} \left( \frac{\mu_a}{\mu_g} \right) \left( \frac{d_b}{U_{out} t} \right)^{1/2}$$

The time necessary to dilute the gas to the concentration  $C$  is then

$$t_C^{(trans. jet)} = \frac{9}{16\beta} \frac{d_b}{U_{out}} \left( \frac{\rho_b}{\rho_a} \right)^{1/2} \left( \frac{\mu_a}{\mu_g} \right)^2 C^{-2} \approx (2.2 - 3.4) \frac{d_b}{U_{out}} \left( \frac{\rho_b}{\rho_a} \right)^{1/2} \left( \frac{\mu_a}{\mu_g} \right)^2 C^{-2} \quad (19)$$

where an experimental range of values for  $\beta = (0.166 - 0.25)$  [6, 9] is used to calculate the constant.

### C. Dilution of the gas in a steady turbulent jet

The axial distributions of velocity  $U_{ax}$  and mass concentration  $Y_{ax}$  of the gas in a free axisymmetric jet issuing from an orifice of the diameter  $d_b$  are described by the hyperbolic law

$$\frac{U_{ax}}{U_{out}} = A \left( \frac{\rho_b}{\rho_a} \right)^{1/2} \frac{d_b}{x} \quad Y_{ax} = B \left( \frac{\rho_b}{\rho_a} \right)^{1/2} \frac{d_b}{x} \quad (20)$$

where  $x$  is the axial distance reckoned from the virtual source,  $U_{out}$  is the discharge velocity,  $\rho_b$  and  $\rho_a$  are the densities of the issuing gas and of the ambient air respectively. The empirical constants  $A$  and  $B$  have been measured by a number of authors (e.g. [11], [12], Sforza [18], Birch *et al* [19]), their average values are  $A = 6.5$ ,  $B = 4.3$ .

To obtain the characteristic time over which the gas is diluted to the volume concentration  $C$  by turbulent mixing with the ambient air in the jet we find out first the coordinate of the axial point corresponding to this concentration, or to the mass fraction equal to

$$Y_{ax} \approx C \left( \mu_g / \mu_a \right) \quad (21)$$

where  $\mu_g$  and  $\mu_a$  are the molecular weights of the gas and of the ambient air respectively. Upon substitution of (21) into the concentration decay law (20) we obtain the coordinate  $x_c$  of this point:

$$x_c = B \left( \frac{\rho_b}{\rho_a} \right)^{1/2} \left( \frac{\mu_a}{\mu_g} \right) \frac{d_b}{C}$$

The mixing time, i.e. the time necessary for the released gas to reach this axial location can be calculated from the velocity distribution (20) as

$$t_c^{(\text{steady jet})} = \int_{x_b}^{x_c} \frac{dx}{U_{ax}(x)} = \frac{(x_c^2 - x_b^2)}{2AU_{out}d_b} \left( \frac{\rho_a}{\rho_b} \right)^{1/2}$$

where  $x_b$  is the coordinate of the orifice with respect to the virtual source. From (20) and the condition  $U_{ax}(x_b) = U_{out}$  we conclude that  $x_b = Ad_b \left( \rho_b / \rho_a \right)^{1/2}$  and  $x_b/x_c = A/B \left( \mu_a / \mu_g \right) C \ll 1$  for small enough  $C$ . Thus, the time  $t_c^{(\text{st. jet})}$  corresponding to dilution of the gas in the axisymmetric jet is

$$t_c^{(\text{steady jet})} = \frac{B^2}{2A} \frac{d_b}{U_{out}} \left( \frac{\rho_b}{\rho_a} \right)^{1/2} \left( \frac{\mu_a}{\mu_b} \right)^2 C^{-2} \approx 1.4 \frac{d_b}{U_{out}} \left( \frac{\rho_b}{\rho_a} \right)^{1/2} \left( \frac{\mu_a}{\mu_g} \right)^2 C^{-2} \quad (22)$$

### D. Volume of the gas in a fuel-rich zone

We consider the Gaussian model of the turbulent jet which is a good approximation to the experimentally observed concentration fields (see [2, 11]). Introduce the cylindrical coordinate system with  $x$  axis aligned with jet axis and  $r$  to be the radial coordinate. The volume concentration at some point  $(x, r)$  is  $C(x, r) = C_{ax}(x) \exp\left(-r/\alpha x\right)^2$  where the axial volume concentration  $C_{ax}(x)$  can be found from (20), (21),  $\alpha$  is the tangent of the jet cone angle defined so that on the line  $r = \alpha x$  the concentration is  $e^{-1}$  of its axial value  $C_{ax}(x)$ .

The volume occupied by the fuel gas in the fuel-rich zone is

$$V_{UFL} = 2\pi \int_0^{x_{UFL}} \int_0^{r_{UFL}} C(x,r) r dr dx \quad (23)$$

where  $x_{UFL}$  is the axial length of the fuel-rich zone,  $r_{UFL}(x)$  is the radius of this zone in the  $x$  cross-section, so that  $C(x, r_{UFL}) = C_{UFL}$ . The inner integral in (23) is calculated as

$$\int_0^{r_{UFL}} C_{ax}(x) \exp\left(-\left(\frac{r}{\alpha x}\right)^2\right) r dr = C_{ax}(x) \frac{(\alpha x)^2}{2} \left(1 - \frac{C_{UFL}}{C_m(x)}\right)$$

After substitution of  $C_{ax}(x)$  from (20)-(21) and the second integration we get

$$V_{UFL} = \frac{\pi \alpha^2 (Bd_b)^3}{6 C_{UFL}^2} \left(\frac{\mu_a}{\mu_g}\right)^{3/2} \quad (24)$$

### E. Initial and average rates of gas discharge from high-pressure vessels

The relationships defining the mass flux of the gas escaping through an orifice in a depressurised vessel are derived elsewhere (e.g. Perry and Green [20]). For high-pressure vessels the flow from the orifice is sonic (choked) and the discharge rate is

$$G = C_d \frac{\pi d_b^2}{4} \left(\frac{2}{k+1}\right)^{\frac{k+1}{2(k-1)}} (kP\rho)^{1/2} = C_d \frac{\pi d_b^2}{4} \left(\frac{2}{k+1}\right)^{\frac{k+1}{2(k-1)}} \left(\frac{k\mu}{RT}\right)^{1/2} P \quad (25)$$

where  $C_d$  is the discharge coefficient,  $P$ ,  $T$  and  $\rho$  are the parameters of the gas in the reservoir.

The pressures  $P$  at which the flow is choked as well as the gas exit pressure  $P_b$  and density  $\rho_b$  are

$$P > P_a \left(\frac{k+1}{2}\right)^{\frac{k}{k-1}}, \quad P_b = P \left(\frac{2}{k+1}\right)^{\frac{k}{k-1}}, \quad \rho_b = \rho \left(\frac{2}{k+1}\right)^{\frac{1}{k-1}} \quad (26)$$

If a finite-volume vessel is depressurised the adiabatic choked discharge rate decreases with time as

$$G(t) = G_0 \left(1 + \frac{G_0(k-1)}{2M} t\right)^{-\frac{k+1}{k-1}}$$

where  $G_0$  is the initial discharge rate corresponding to the storage conditions  $P_0$ ,  $\rho_0$  and  $M$  is the total mass of the gas stored (e.g., [2], Woodward and Mudan [21]). The average discharge rate over the period of the sonic flow as determined in [21] is  $\bar{G} = \eta \cdot G_0$  where  $\eta$  decreases monotonously with increase in the initial/ambient pressure ratio, for nearly critical conditions  $\eta$  is equal to 1, while at high initial pressures ( $P_0/P_a \approx 100$ ) the value of  $\eta$  reaches about 1/4. The formula obtained for  $\eta$  in [21] is too complicated to be used in the current analysis, that is why we searched for a simple approximating formula that could be used instead of it. It was found that at high pressures ( $P_0/P_a > 10$ ) the dependence of  $\eta$  on the pressure ratio can be approximated with accuracy sufficient for the estimation purposes as

$$\eta \approx 0.6 \left(P_a/P_0\right)^{1/6} \quad (27)$$

Since the discharge rate is proportional to the pressure (or the density) of the gas (see (25)), the average pressure and density of the gas during the release are

$$\bar{P} = \eta \cdot P_0, \quad \bar{\rho} = \eta \cdot \rho_0 \quad (28)$$

Thus, the transient discharge can be approximated by a constant-rate outflow proceeding at the average pressure and density in the vessel.