European Train the Trainer Programme for Responders

# Lecture 7

# Unignited hydrogen releases outdoors and their mitigation

## LEVEL IV

## Specialist officer

The information contained in this lecture is targeted at the level of specialist officer and above.

This topic is also available at levels I-III.

This lecture is part of a training material package with materials at levels I – IV : Firefighter, crew commander, incident commander and specialist officer. Please see the lecture introduction regarding competence and learning expectations

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Clean Hydrogen Partnership

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## Summary

Unwanted hydrogen releases are followed by the mixing of escaped gas with air, thus establishing the initial conditions for fire and explosion hazards. The unignited releases involve the escape of compressed gaseous hydrogen stored at high pressures at FCH systems and infrastructure. Accidental releases of compressed gaseous hydrogen fall into two main categories: permeation leaks and high-pressure jets. One of the important questions for Responders is whether a release of gaseous hydrogen will lead to the formation of a flammable mixture with air. By using similarity law and under-expanded jet theory, Responders will be able to evaluate the size of the flammable hydrogen envelope and to calculate hydrogen concentration decay along the jet axis. Additionally, Responders will learn how to calculate the blow-down time and to assess the pressure dynamics in a storage tank of known capacity. Finally, this lecture introduces Responders to the means of reducing the hazard distance from the point of the release as well as to prevention/mitigation/detection measures of unignited hydrogen releases.

## Keywords

Unignited release, similarity law, under-expanded jet theory, hydrogen envelope, hydrogen concentration decay, blow-down, hazard distance, mitigation measures, detection

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# 1. Target audience

The information contained in this lecture is targeted at the level of specialist officer and above. This lecture is also available at levels I: Fire fighter.

The role description, competence level and learning expectations assumed at specialist officer level are described below.

## 1.1 Roll description: Specialist

Specialists assist Incident Commanders with advice and by directing and supervising technical operations that involve the use of knowledge, skills or equipment related to a specific risk or response activity. Typical incidents involve hazardous materials, transportation vehicles, climatic events, structural and other built environment failures and emergency response logistics.

## 1.2 Competence level: Specialist

A scientific, engineering and empirical foundation in the subject area with skills, extended by experience, to apply that information, knowledge in an interpreted and useful way to enable the Incident Commander and other first responders under the specialist's direction to respond effectively and safety to the emergency situation.

## 1.3 Prior learning: Specialist

EQF 5 Comprehensive, specialised, factual and theoretical knowledge within a field of work or study and an awareness of the boundaries of that knowledge. A comprehensive range of cognitive and practical skills required to develop creative solutions to abstract problems. Exercise management and supervision in contexts of work or study activities where there is unpredictable change; review and develop performance of self and others.

## 2. Introduction and objectives

An unwanted hydrogen release can be caused either by an equipment failure or by a loss of leak tightness of a FCH system. The consequences of this event can be different, depending on the characteristics of the release and the location of a source of the leak, i.e. outdoors or indoors. If initially the release is unignited, there is still a possibility that it will ignite, after a certain delay, when an ignition source is present in the path of the release. If hydrogen release occurs indoors the consequences will be more severe compared with the releases happening outdoors as an explosive atmosphere may build up in a confinement. All these consequences can affect people, structures, and environments. Minor incidents may escalate leading to catastrophic outcomes.

This lecture is mainly focused on unignited hydrogen releases in an open environment. The classification of compressed hydrogen leaks is presented in the current lecture as well as some useful terminology. Responders are introduced to the similarity law and under-expanded jet theory, which will allow them to evaluate hydrogen concentration decay and determine hazard distance from the point of release to the lower flammability limit or any other concentration of interest. The means of reducing the hazard distance from a source of hydrogen release are detailed in this lecture. It also discusses prevention and mitigation/detection measures for unignited hydrogen releases from compressed hydrogen storage vessels.

By the end of this lecture Responders will be able to:

- Classify unignited hydrogen releases (leaks/jets) and distinguish between permeation leaks, sub-sonic, sonic and super-sonic jets; expanded and under-expanded jets, momentum- and buoyancy-controlled jets;
- Evaluate hydrogen concentration decay in the momentum-controlled jets using corresponding nomogram;
- Predict the distance, at which hydrogen jet becomes buoyant;
- Calculate the size of hydrogen flammable envelope, i.e. to determine the furthest point from the source of leak, at which a jet can be ignited;
- Assess a safe size of a TPRD (thermal activated pressure relief device) with the view to avoid the formation of a flammable hydrogen layer under a ceiling in the enclosure;
- Calculate the blow-down time for compressed hydrogen tanks of different capacities;
- Recognise the means of reducing hazard distances from the point of the release;
- State the main prevention techniques for hydrogen jets;
- Recognise the mitigation measures of hydrogen leaks consequences;
- Point out the detection means of hydrogen releases.

## 3. Useful definitions

*Blow-down* is a process where the storage pressure decreases with time during a leak.

*Expanded jet* is a jet with a pressure at the nozzle exit equal to atmospheric pressure.

*Mach number* (M or Ma) is a dimensionless number equal to the ratio of a local flow velocity to the local speed of sound.

*Permeation* is the movement of atoms, molecules, or ions into or through a porous or permeable substance.

*Hazard distance* is a distance from the (source of) hazard to a determined (by physical or numerical modelling, or by a regulation) physical effect value (normally, thermal or pressure) that may lead to a harm condition (ranging from "no harm" to "max harm") to people, equipment or environment (as per draft ISO TC197 definition).

*Under-expanded jet* is a jet with a pressure at the nozzle exit above the atmospheric pressure.

## 4. Compressed hydrogen leaks

As it is already known from previous lectures FC vehicles are equipped with on-board hydrogen storage tanks pressurised up to 70 MPa, and a refuelling infrastructure operates at pressures up to 100 MPa [1]. Due to the small size of its molecule, hydrogen is prone to leakage. Predominantly hydrogen releases/leaks originate from valves and connections [2], which may occur both indoors and outdoors. The releases can be unignited (i.e. non-reacting) or ignited (i.e. reacting). Although a full-bore rupture of a pipe or vessel is a rare event, it should be considered as a credible worst-case scenario. Special efforts should be made to prevent the unwanted hydrogen releases. A release of hydrogen either through a PRD or from a pipe rupture will result in a high-pressure jet.

Table 1 summarises the types of leaks and equipment or components generating hydrogen leaks [3].

Table 1. Leak sources and scenarios developed by EIGA (2007) [3]

| Equipment/component | Type of leak |
|---|---|
| Pipework | Pinholes, pipe split |
| Flange's | Gasket failure, thermal movement, material creep |
| Weld connection | Weld crack |
| Solder connection | Solder crack, solder melt |
| Union connection | Thermal movement, leak |
| Screw connection | Leak, sealant failure, creep, material split |
| Hose connection | Seal leak, material split, human error |
| Valves | Stem leak, seal leak, bonnet/housing split opened by impact |

| | |
|---|---|
| Hoses | Perforation split |
| Instruments | Element rupture |
| Regulators | Diaphragm rupture, seal leak, downstream rupture (overpressure) |
| Solenoid valves | Seal leak |
| Pumps | Perforation, seal leak |
| Cylinder | Perforation, rupture, permeation leak |

Typical flow rates of hydrogen releases vary significantly, from $10^{-5}$ g/s for permeation leaks to $10^{5}$ g/s for leaks on hydrogen pipelines. According to the European Regulation for type approval of hydrogen-powered vehicles (2010) the *safe permeation rate* for FC vehicles at 15 and 20 °C is limited to 6 and 8 mL/hr per L of a storage tank capacity [4]. It means for 170 L capacity of hydrogen storage permeation rate will be of $2.53\times10^{-5}$ g/s. A flow rate for a leak from a broken pipe (on a 150 kW FC) will be around 3 g/s. A release from an industrial pipeline (diameter 30 cm) at p = 2.5 MPa will have a flow rate of 100 kg/s. Currently, a release via a PRD (with a diameter of 5.08 mm) from a storage tank at 35 MPa will generate a flow rate of 390 g/s.

A permeation-induced release of hydrogen differs from plumes and jets: hydrogen releases slowly, in very small amounts, equally along the surface of a storage tank. The rate of hydrogen permeation through a particular material depends on several parameters, e. g. the temperature, internal pressure, and the membrane thickness. The higher the storage pressure the higher is the permeation rate (please refer to Lecture 3 'Hydrogen storage'). Three main phenomena drive the dispersion of permeated hydrogen: buoyancy, diffusion, and natural ventilation. Permeated hydrogen is distributed homogeneously in a garage-type enclosure [5]. Maximum allowable permeation rate at 20 °C for a passenger car is 8 mL/hr/L and for a FC-bus 5 mL/hr/L in a garage-type enclosure [6]. The simulations carried out at HySAFER of Ulster University demonstrated that with this level of permeation rate the hydrogen dispersion in a typical garage is not a problem [5].

An unscheduled release of hydrogen stored at high pressures can produce a highly *under-expanded* (pressure at the nozzle exit is above atmospheric pressure) turbulent jet that behaves differently compared to the *expanded* jets (pressure at the nozzle exit is equal to atmospheric pressure), which was extensively studied in the past. The schematic representation of a hydrogen release from high-pressurised storage vessel is shown in Figure 1. The majority of leaks from hydrogen storage and equipment will be in a form of an under-expanded jet, at least at the beginning [1].

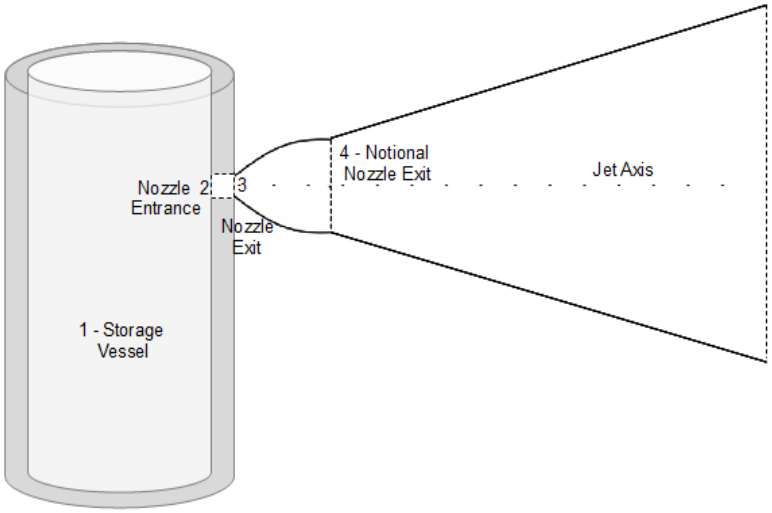



Figure 1. A scheme of a hydrogen release from high pressure hydrogen storage vessel.

## 4.1 Expanded and under-expanded jets

Hydrogen jets can be expanded or under-expanded. An *expanded jet* is a jet with a pressure at the nozzle exit equal to atmospheric pressure. In an *under-expanded jet*, the pressure at the nozzle exit has not fully dropped to the atmospheric one. At high pressures, a velocity at the exit from the nozzle remains locally sonic, but the exit pressure rises above ambient. As the result, the expansion down to ambient conditions takes place outside the nozzle [1]. The critical ratio between storage and ambient pressure for sonic (choked) flow of compressible gas to be established is about 1.9 at STP. If the pressure ratio is below 1.9 then the gas flow in an orifice is expanded (subsonic). Otherwise, for the ratio above 1.9, the releases are considered to be under-expanded (sonic or choked) flow.

For under-expanded jet, the flow expansion occurs near the nozzle exit and is characterised by the complex shock structure, which is well documented and published elsewhere [7]. The schematic for an under-expanded shock structure is presented in Figure 2a. In this diagram, one can see a distribution of the *Mach number* (M or Ma), which is a dimensionless number equal to the ratio of a local flow velocity to the local speed of sound. *Mach disk* is a strong shock, which is normal to the under-expanded jet flow direction. A numerically simulated initial stage of hydrogen under-expanded release is presented on Figure 2b, for pressure ratio in a storage tank and the atmosphere 160.

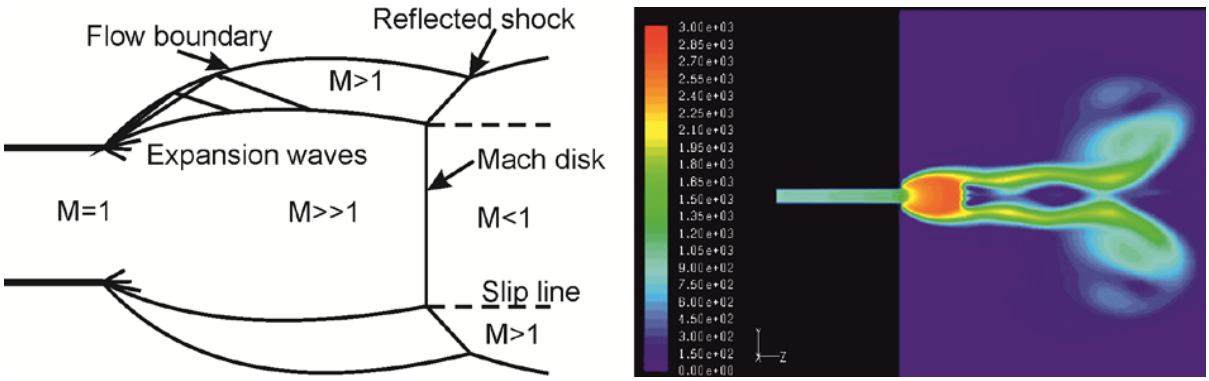



Figure 2. (a) The schematic of an under-expanded jet structure [8]; (b) an initial stage of under-expanded jet release at 16.1 MPa through a channel of 0.25 mm with a mass flow rate 0.46 g/s [1].

As it is shown in Figure 2a the local sonic velocity is established at the nozzle exit with the Mach number $M$ = 1. Then, the outflowing gas undergoes a rapid expansion and quickly accelerates to high Mach numbers (>>1, up to $M$ = 8 for 70 MPa storage pressure) accompanied with the decrease in pressure and density. A series of expansion waves are formed at the nozzle exit edge. These expansion waves are reflected as compression waves from the free surface at the jet flow boundary, coalesce and form a barrel shock and a Mach disk. Repeating barrel shocks and Mach disks structures downstream of the first Mach disk appear intermittently at pressure ratios below 20. For typical hydrogen storage pressures a short structure with only one Mach disk can be expected at the beginning of the *blow-down* (i.e. a process where the storage pressure decreases with time during a leak). As the gas with very high Mach number crosses the Mach disk, it undergoes a sudden decrease in velocity to subsonic speeds and increases in pressure (to the atmospheric) and density. The resulting flow structure after the Mach disk comprises a subsonic core ($M$ < 1) surrounded by supersonic shell ($M$ > 1) with a shear layer called slip line dividing these two regions.

## 4.2 Sub-sonic, sonic and supersonic jets

The Mach number of the jet, which is equal to a jet velocity ($U$) divided by the speed of sound in the gas ($C$) exiting into the ambient air, is an important parameter in determining its behaviour. For jets with a *subsonic flow* $U < C$, $M < 1$; the pressure at the exit plane is just equal (or very close) to atmospheric pressure. As stated earlier, many leaks from high pressure storage form an *under-expanded* jet with the *sonic* (choked) flow at the jet exit (when $U = C$, and $M$ = 1). The *supersonic* jets are characterized by the velocity larger than the speed of sound ($U > C$).

At high pressures, the exit velocity remains locally sonic, but the exit pressure rises above ambient with the result that expansion down to ambient conditions takes place outside the nozzle. For hydrogen, the critical pressure ratio for sonic (choked) flow of compressible gas is

1.89 at STP (standard temperature and pressure) ($\gamma$ =1.405) and 1.89 at NTP ($\gamma$ =1.39) according to the theoretical equation for choked flow conditions:

$$\frac{P_R}{P_N} = \left(\frac{\gamma+1}{2}\right)^{\gamma/(\gamma-1)} \quad (1)$$

where $P_R$ and $P_N$ are pressure in the reservoir and the nozzle exit respectively, and $\gamma$ is the ratio of specific heats. The equation (1) can be applied to estimate the pressure at the leak (nozzle) exit $P_N$ if the storage pressure at the reservoir $P_R$ is known [1].

Ishii et al. [8] reported that jets of a diatomic gas ($\gamma$ = 7/5) exhausted from an open-end of the test tube tend to be: *subsonic matched* jets for the ratios of pressures in the high- and low-pressure chambers (the only parameter controlling the jet strength) between 1 and 4.1; *sonic under-expanded* jets for pressure ratios in the range from 4.1 to 41.2; and *supersonic under-expanded* jets for pressure ratios above 41.2. Pressure ratio of 4.1 is above the critical ratio 1.9 for sonic (choked) flow. However, there is no contradiction: each of two pressure ratios (high-pressure chamber to tube exit, and tube exit to low-pressure chamber) can be $P_R/P_N$ = 2.05, which is close to the theoretical critical pressure ratio of 1.9 for sonic flow. The difference could be attributed to losses between high- and low-pressure chambers.

## 4.3 Momentum- and buoyancy-controlled jets

All jets can be divided into three types depending on the effect of buoyancy. These jet types are shown schematically in Figure 3 for a horizontally oriented jet. Fully *momentum-controlled jets* are not affected by the buoyancy, whilst fully *buoyancy-controlled jets* are diverted fast from horizontal to vertical direction of flow. The third type is a *transitional jet*, which has a momentum-controlled part located close to the nozzle exit, and a buoyancy-controlled part of the flow further downstream, when the jet velocity decreases and the diameter of the jet increases. It is important to know the moment when this transition takes place because it will have an impact on the hazard distance in particular and on first response strategy and tactics in general [1].

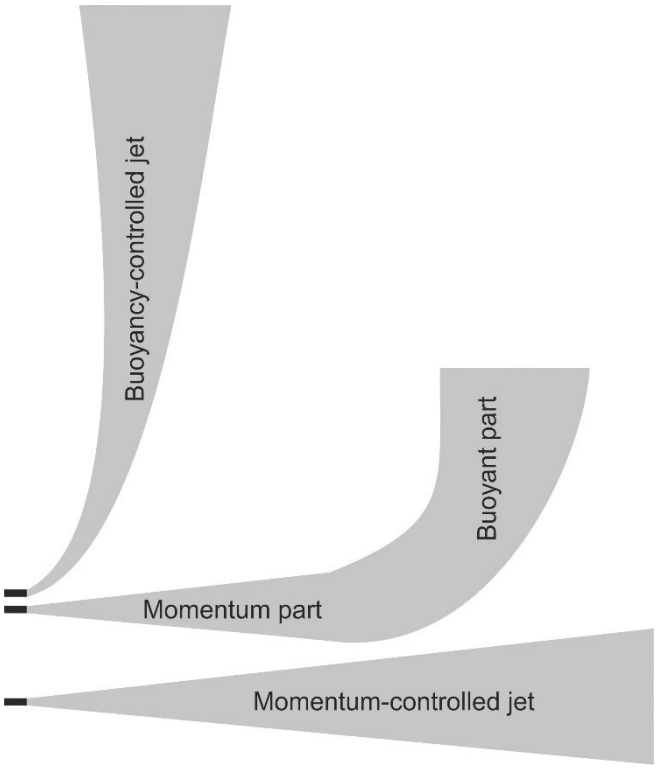



Figure 3. The schematic representation of momentum-controlled, transitional and buoyancy-controlled jets [1].

## 5. Hydrogen concentration decay in momentum-controlled jets

The concentration of hydrogen in a jet decays from 100% at the nozzle (i.e. at a point of leak) to the lower values along the jet axis and from axis to periphery. For some calculations it is more convenient to substitute an under-expanded jet from an actual nozzle with an expanded jet from so-called *effective* or *notional nozzle*. There are several notional nozzle theories. The most cited model is the one suggested by Birch et al. [9] in 1984. However, the former models are built on the ideal gas equation of state, and thus are not applicable to the storage pressures above 10 - 20 MPa, when the non-ideal behaviour of a gas must be accounted for. The first theory of *under-expanded jet* that accounts for a non-ideal behaviour of highly compressed hydrogen was published by Schefer et al. [10] in 2007. The notional nozzle diameter calculations by Schefer et al. [10] are similar to that of Birch et al. [9], yet the Abel-Noble equation is applied and the assumption by Birch et al. about the speed of sound at the notional nozzle is relaxed [9,10]. As a result, the theory of Schefer et al. [10] predicts uniform super-sonic velocities at the notional nozzle exit at high storage pressures.

Molkov et al. [11] developed an under-expanded theory, alternative to the one suggested by Schefer et al. [10]. This theory is based on the mass and energy conservation equations rather than on mass and momentum ones. Similar to Birch et al. [9], the model developed by Molkov

et al. [11] is based on the assumption of a uniform sonic flow through the notional nozzle. The calculation scheme used for the *under-expanded jet* theory is shown in Figure 4.

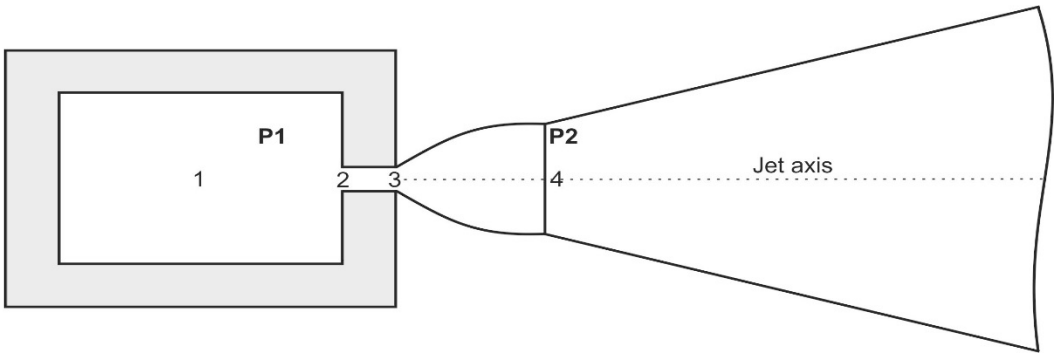



Figure 4. The scheme of an under-expanded jet: 1 - high pressure vessel; 2 - nozzle entrance; 3 - nozzle exit (= notional nozzle entrance); 4 - notional nozzle exit (3-4: no entrainment); P1 - storage pressure; P2 - atmospheric pressure (after the jet expansion) [1].

It is assumed that the flow velocity in the storage reservoir (1) is zero. The flow parameters at the entrance to the leak channel, nozzle entrance, are referred to as (2), and at the actual nozzle exit - as (3). For sonic and super-sonic flows, the parameters at the nozzle exit (3) are those for choked flow and therefore the nozzle exit velocity is equal to the local speed of sound (Mach number $M = 1$). The notional nozzle is between the actual nozzle exit (3) and the notional nozzle exit (4). At the notional nozzle exit (4) jet parameters correspond to a fully expanded jet with the pressure equal to the ambient one and uniform flow velocity equal to a local speed of sound. In some cases, the essential minor and friction losses, in the flow pathway (2) - (3), cannot be neglected, for example in the case of a very narrow crack [1].

As discussed previously combustion is generally of a primary concern when considering harm criteria. An unscheduled hydrogen release from high pressure equipment and infrastructure will form a highly *under-expanded jet*. This could lead to a formation of a large *flammable hydrogen-air envelope*. The size of the flammable envelope is the deterministic hazard distance from the source of the release. In the event of its subsequent ignition people, structures, equipment, and environment may be involved in a fire. The presence of an ignition source within the envelope could initiate severe *jet fires*, *deflagration*, and, potentially, *deflagration-to-detonation transition*. It must be noted that thermal effects of jet fires and pressure effects of deflagration or detonation could override the hazard distance determined by the size of flammable envelope [1]. Thus, the knowledge of the laws related to hydrogen dispersion and a flammable cloud formation, including *axial concentration decay* for arbitrary jets with various parameters is essential for decision making for Responders.

If the flammable envelope reaches a location of an air intake in high-rise buildings, the consequences for occupants and building structure may be catastrophic. Thus, it is important to determine the furthest point from the leak at which a jet can be ignited. This is generally defined by hydrogen concentration of 4 vol. %, i.e. by the LFL (lower flammability limit). Although this choice of hydrogen concentration is subject to debate, direct flame contact as a

result of a flash fire will also occur if a person is within the 4 vol. % hydrogen envelope when hydrogen ignition occurs. The flammable envelope size, i.e. distance to LFL of 4 vol. %, increases proportionally to the diameter [1].

The *similarity law*, discussed below, is used for the prediction of the *axial concentration decay* of a leaking gas for sub-sonic, sonic, and super-sonic jets. The similarity law is shown to be valid in a wide range of conditions from expanded to highly *under-expanded jets*. It can be applied for calculation of *hazard distances* informed by the size of the flammable envelope. The non-ideal behaviour of hydrogen at high pressures and the under-expansion of the flow in a nozzle exit are taken into account. In earlier publications by Molkov and his colleagues [11, 14] these distances were referred to as separation or hazard distances the definitions of which varied from one document to another. Some examples of the definitions are given below:

*"a minimum distance, which separates specific targets (e.g. people, structures or equipment) from the consequences of potential accidents related to the operation a hydrogen facility"* [12].

*"a minimum separation between a hazard source and an object (human, equipment or environment) which will mitigate the effect of a likely foreseeable incident and prevent a minor incident escalating into a larger incident"* [3].

The following factors affect the hazard distances:

- the nature of the hazard,
- the operating conditions and the design of the analysed equipment/facility,
- the type of target/object (people, structures, equipment),
- the environment between the latter and the source of hazard. In this way, the harm potential for people or structures can be evaluated and compared with the harm criteria.

In order to define where the flammable hydrogen-air mixture is formed, it is important to know how the concentration decays, from 100 vol. % at the nozzle to the LFL, i.e. to 4 vol. %. The original form of the similarity law (equation 2) for the axial concentration decay in the expanded round jets was suggested by Chen and Rodi [13] in 1980:

$$C_{ax}^{m} = 5.4\sqrt{\frac{\rho_N}{\rho_S}}\frac{D}{x} \qquad (2)$$

where $C_{ax}^{m}$ is the axial mass fraction of hydrogen in the jet at the distance $x$ from the nozzle, $\rho_S$ is the density of the surrounding gas, i.e. air (1.205 kg/m$^3$ at NTP), $D$ is the real nozzle diameter exit, $\rho_N$ is hydrogen density in the nozzle exit (the only unknown parameter).

The "unknown" density $\rho_N$ can be calculated with the use of the *under-expanded jet theory* developed and validated at the HySAFER center, Ulster University. The distance to the LFL is proportional to the leak diameter. Thus, the design of FCH systems has to satisfy technological requirements to mass flow rate and at the same time to take into account the requirement to

minimise the internal diameter of piping to decrease potential hazard distance as it follows from Equation (2). One should keep in mind that $\rho_N$ also affects the flammable envelope size of 4 vol. % (i.e. mass fraction $C_m$ of 0.00288) – the larger operating pressure of a FCH application, the larger notional nozzle density $\rho_N$, and, consequently, the longer hazard distance.

It can be concluded from the similarity law (2) that for hydrogen jets released to a stagnant air from a given storage pressure (i.e. fixed value of $\rho_N$) the ratio of a distance to a fixed concentration, $x = L$, expressed as a percentage by mass, to the nozzle diameter, $D$, is a constant, i.e. $L/D$ = const. This means that the hazard distance is proportional to the leak diameter. Please note that the mass fraction ($C_m$) can be calculated from Equation (3) based on the volumetric (mole) fraction ($C_V$):

$$1/C_m = 1 + (1/C_V - 1)M_S/M_N \qquad (3)$$

where $M_S$ and $M_N$ are the molecular masses of a surrounding gas (28.84 g/mol is accepted for air) and nozzle gas (hydrogen), respectively. For example, the mass fraction of 0.0282 corresponds to the volumetric fraction of 0.295 (29.5 vol. % of hydrogen in air, i.e. a stoichiometric mixture); the mass fraction of 0.044365 corresponds to the volumetric fraction of 0.401 (40.1 vol. %, i.e. to a mixture with a maximum burning velocity), 0.013037 – 0.16 (16 vol. %), 0.008498 – 0.11 (11 vol. %), 0.005994 – 0.08 (8 vol. %), 0.00288 – 0.04 (4 vol. % - LFL), 0.00141 – 0.02 (2 vol. %), 0.0007 – 0.01 (1 vol. %).

In the case of fully expanded, round jets the similarity law (2) gives the following ratios of a distance to particular axial hydrogen concentration (in vol. %), $L$, to the characteristic nozzle size, $D$ (for atmospheric nozzle pressure and expanded subsonic jets, with density ratio $\rho_S/\rho_N$ = 14.38: $(L/D)_{30\%}$ = 49.1; $(L/D)_{4\%}$ = 491; $(L/D)_{2\%}$ = 1003; $(L/D)_{1\%}$ = 2019. For example, the distance to the LFL of hydrogen in air (4 vol. %) in the momentum-controlled expanded jets is equal to 491 nozzle diameters. Please note that the correlation (2) by Chen and Rodi [13] was validated by concentration measurements in vertical jets up to the ratio of $L/D$ = 50 only. Its applicability above this range should be confirmed [1].

If the similarity law by Chen and Rodi [13] is applied in its original form (2) to the expanded jets, then the calculation of hazard distances for unignited hydrogen round jets would be straight forward: by using Equation (2) with hydrogen density at the nozzle exit $\rho_N$ = 0.0838 kg/m$^3$ (NTP). However, for the *under-expanded jets* there is one unknown parameter, the density of the gas in the actual nozzle exit, $\rho_N$. This density can be calculated by the under-expanded theories, either without losses [11] or with losses [14].

The similarity law for hydrogen concentration decay along the axis of the momentum-controlled jets, as well as the experimental data for hydrogen under-expanded releases, are shown in Figure 5.

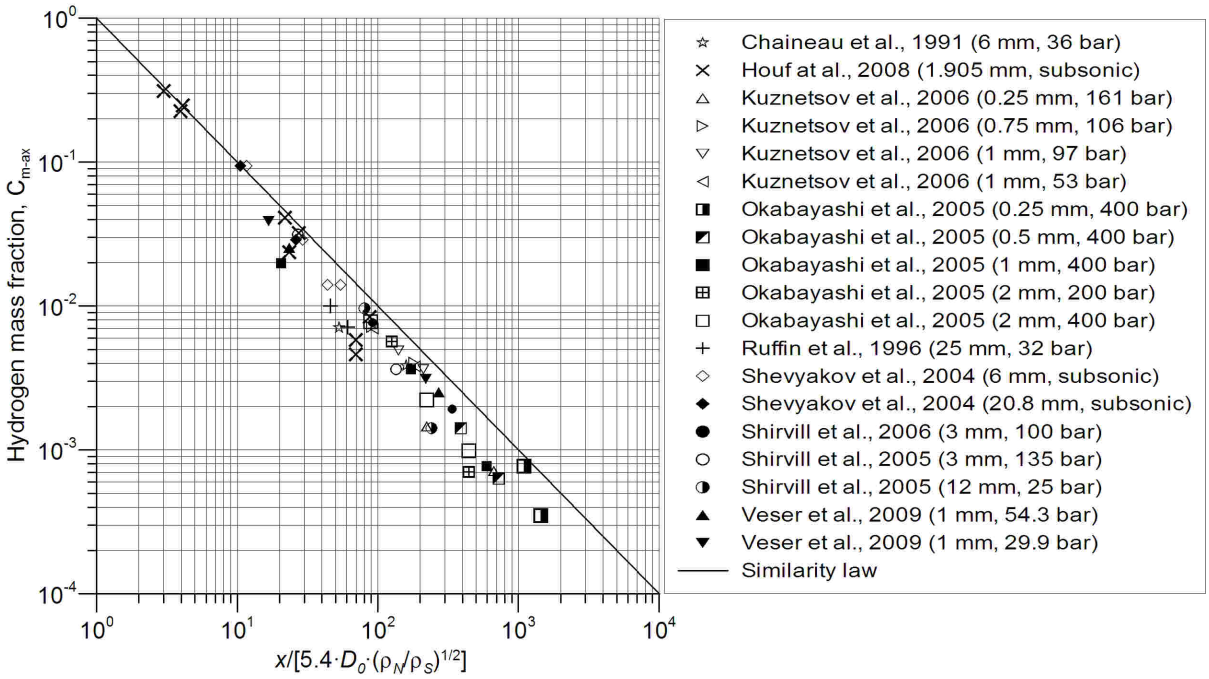



Figure 5. The similarity law (straight, solid line) and the experimental data (symbols) on the axial concentration decay in the momentum-controlled expanded and under-expanded hydrogen jets [1].

The experimental data on the axial concentration decay of pure hydrogen in momentum-controlled sub-sonic, sonic and super-sonic jets, released from the vessels of different volume pressurized up to 40 MPa, and through nozzles with diameters ranging from 0.25 to 100 mm are reported in [2]. The concentration of hydrogen in the air was in the interval between 1 and 86.6 vol. %. The similarity law was validated to the ratios *x/D* in the range from 4 to 28,580. Laminar and turbulent, expanded and under-expanded momentum-controlled jets were used to validate the universal character of the similarity law [2]. Please note that the similarity law is conservative in relation to the experimental data points, which is due to the effect of friction and minor losses in the experiments.

As indicated earlier, the mass fraction of hydrogen corresponding to the LFL (4 vol. %) is 0.00288. If we use this value along with the air density of 1.205 kg/m$^3$ (NTP), Equation (2) will be simplified to the following formula:

$$x_{4\%} = 1708 \cdot \sqrt{\rho_N} \cdot D \tag{4}$$

which can be used to evaluate the size of flammable envelope to 4 vol. % of hydrogen (hazard distance for unignited hydrogen release).

The extent of the unignited hydrogen release can be calculated using two methods:

1. The nomogram for hydrogen concentration decay.
2. e-Laboratory (https://hyresponder.eu/e-platform/e-laboratory/) .

The *nomogram* for the graphical evaluation of hydrogen concentration decay with the use of the similarity law and the under-expanded jet theory without losses is shown in Figure 6. The nomogram consists of four main graphs entitled: "volumetric to mass fraction", "the similarity law", "choose leak diameter", and "choose density in the nozzle exit", and one additional graph "calculate density in the nozzle exit by storage tank pressure and temperature" (based on calculations by the under-expanded jet theory without losses).

The use of the nomogram for the calculation of the distance from the nozzle (for example 1 mm in diameter) to the 4 vol. % of hydrogen in air (blue-coloured dash line) along the axis of the release from a storage tank at a pressure of 70 MPa and temperature 300 K is demonstrated below.

1. Draw the vertical line downward from the point on the horizontal axis "hydrogen volumetric fraction", corresponding to the concentration of interest (4 vol. % or 0.04), until the intersection with the line of the graph "volumetric to mass fraction" (left-hand, top corner in Figure 6).
2. Draw the horizontal line from this intersection point to the intersection with the similarity law line in the right-hand top corner graph "the similarity law" (Figure 6).
3. Draw the vertical line downward from the intersection point obtained on "the similarity law" graph until the intersection with line corresponding to 1 mm diameter on the graph "choose leak diameter" (Figure 6). Please note that there are eight lines on the "choose leak diameter" graph, which correspond to the following leak diameters (from top to bottom): 15 mm, 10 mm, 5 mm, 3 mm, 2 mm, 1 mm, 0.5 mm, 0.1 mm). These figures are shown at the right side of the graph.
4. Calculate the density using the additional graph "calculate density in the nozzle exit by storage tank pressure and temperature" located at the bottom of the nomogram using given pressure (in our example - 70 MPa) on the ordinate axis and a line corresponding to the chosen temperature (300 K). This is shown by two thick grey arrows on the "calculate density in the nozzle exit by storage tank pressure and temperature" graph. The density calculated graphically at the nozzle exit for 70 MPa and 300 K is about 23 kg/m$^3$.
5. Coming back to the "choose leak diameter" graph, draw the horizontal line from the intersection point on the line "1 mm" until the intersection with an imaginary line corresponding to 23 kg/m$^3$ (located between two lines, 20 kg/m$^3$ and 50 kg/m$^3$, shown in the graph). Please note that there are five lines at the graph corresponding to the densities of 1 kg/m$^3$, 3 kg/m$^3$, 10 kg/m$^3$, 20 kg/m$^3$, and 50 kg/m$^3$ from top to bottom, respectively. These values are shown at the left side of the graph.
6. Draw the vertical line downward from the intersection point with the imaginary line corresponding to 23 kg/m$^3$ to the intersection with the abscissa axis 'distance to concentration of interest' on the "choose density in the nozzle exit" graph. Thus, the

calculated graphically distance from the nozzle exit to hydrogen concentration of 4% by volume is about 7.7 m.

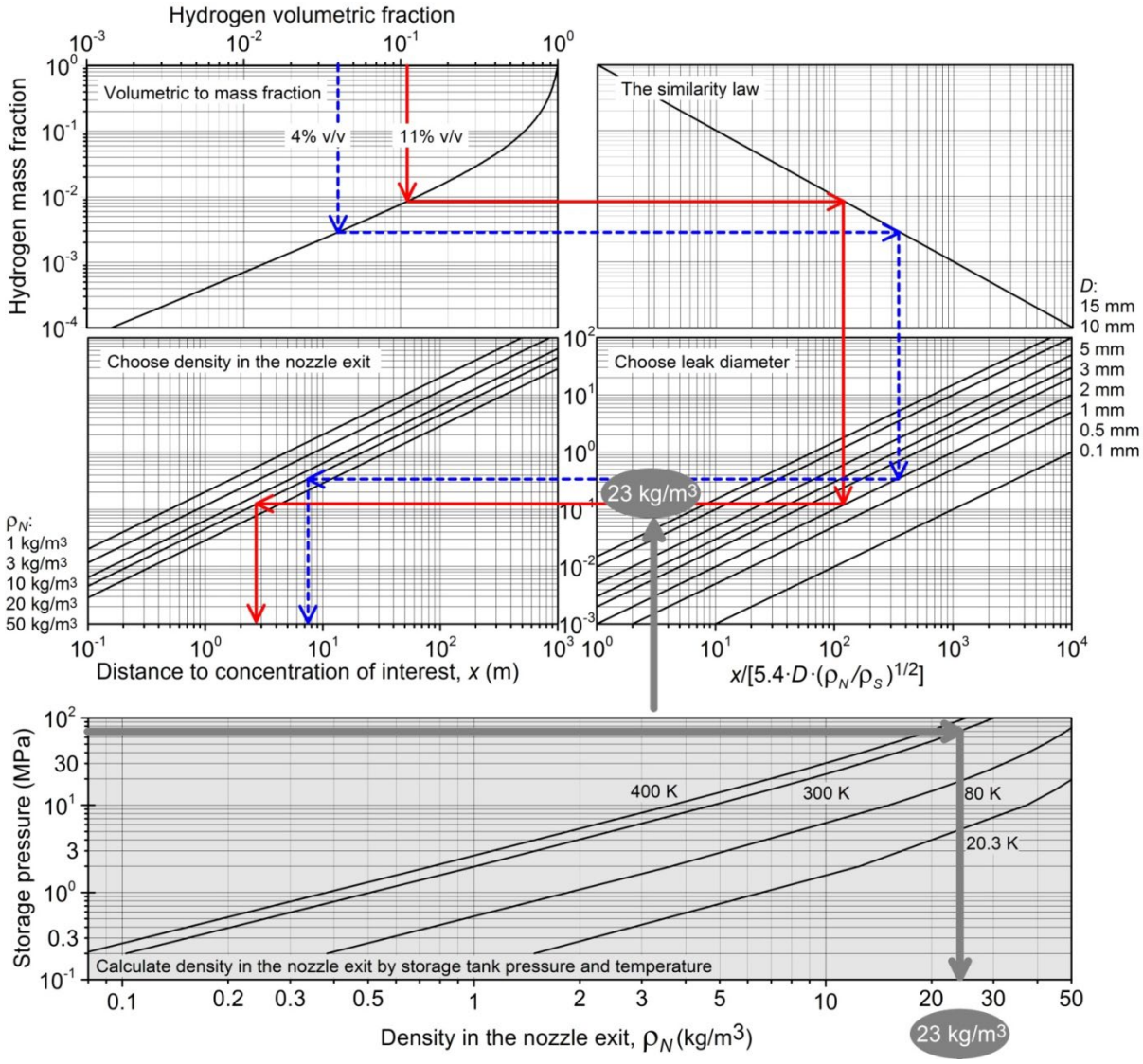



Figure 6. The nomogram for concentration decay calculation in unignited jets [1].

The use of Equation (2) for the similarity law with more accurate value of hydrogen density at the nozzle calculated by the *under-expanded jet theory* (23.95 kg/m$^3$) and air density of 1.205 kg/m$^3$ (NTP) gives a distance of 8.36 m for 4 vol. % of hydrogen in air. The error of graphical calculations is at the acceptable level and below 10%.

Task for trainees: using the nomogram in Figure 6 evaluate the distance to 11 vol. % of hydrogen concentration in air assuming the same storage pressure 70 MPa, storage temperature 300 K and physical nozzle diameter 1 mm (corresponds to an average flame tip location).

e-Laboratory tool 'Jet parameters model' provides quick and simple way to evaluate the density of the gas in the nozzle by entering the following data: pressure in the reservoir, temperature in the tank, orifice diameter and ambient pressure.

The similarity law for *under-expanded jets* and the nomogram are performance-based tools for calculation of hazard distances for unignited releases. They are the alternative to the prescriptive codes. For instance, the International Fire Code, (2006) contains a table of prescribed distances between a hydrogen system and various potential targets, including an air intake to a building [15]. However, this code does not provide or require any information either on the hydrogen storage parameters (pressure and temperature) and a leak size from the system. This code is an example of an archaic prescriptive approach that contradicts the modern requirements to codes and standards to be performance-based [1].

The standard NFPA 55 [16] in 2010 moved one step further, compared to the prescriptive International Fire Code [15] in 2010, by presenting a set of 4 equations to calculate distances to the axial concentration of 4 vol. % of hydrogen in air. Molkov [1] indicated two weak points in this standard. To "simplify" the use of the methodology by practitioners each of 4 equations is assigned to a quite wide range of pressures. This results in a "programmed" over-prediction and under-prediction of the distance depending on the storage pressure. However, the most serious disadvantage in the calculation of the distance by NFPA 55 [16] is the use of a leak size of only 3% of the flow area of the pipe (this choice is based on leak frequencies of other gases available to gas industry) [1]. Indeed, the similarity law states that for the same density of hydrogen in the nozzle exit $\rho_N$, i.e. for the same storage pressure, the hazard distance is directly proportional to the leak diameter. Thus, the questionable choice of a leak size as 3% of cross-section area of the pipe in NFPA 55 [16] results in the "prescribed" decrease of the distance by reciprocal to square root from 0.03, i.e. by 5.77 times. It is clear that in the case of full-bore rupture of a hydrogen pipe the consequences could be catastrophic if this "3%" approach is widely applied and enforced by the regulators. The risk-informed approaches cannot compromise the science-informed engineering design. The alternatives to this "forced" decrease of the hazard distance is safety engineering design of hydrogen systems, e.g. reduction of pressure in the pipe when possible and the decrease of the pipe diameter to the minimum size required by the technological reasons and/or substitution of one large pipe by a number of smaller pipes, use of PRD with enhanced safety characteristics, etc. [1].

The conclusion can be drawn that the similarity law may be used to calculate the hazard distance more accurately compared to currently available standards and codes. This methodology is scientifically underpinned and can account for friction and minor losses in a leak pathway when/if necessary. Last but not the least, the hydrogen safety engineering guarantees the freedom to consider worst-credible scenario of full bore rupture rather than the questionable 3% of cross-section area of pipe approach as suggested by some codes and standards [1].

## 6. Reduction of a hazard distance from a hydrogen release

Equation (4) demonstrates that the hazard distance for unignited jets, $x$, is proportional to the diameter of the release, $D$, and grows as a square root from the storage pressure (term density

$\rho_N$). With this in mind, the key safety requirement for unignited hydrogen release can be formulated as: *reduce the diameter of piping (which is a leak diameter for a credible worst-case scenario) to the technologically affordable* minimum, align with the ALARP ('as low as reasonably practicable') or ALARA ('as low as reasonably achevable') principle, which is often used in the regulation and management of safety-critical and safety-involved system. The same can be applied to the diameter of the Pressure Relief Devices (PRDs) or Temperature Activated Pressure Release Devices (TPRDs) [2].

## 6.1 When a jet becomes buoyant?

Depending on the buoyancy effect the jets can be momentum-controlled, transitional or buoyancy controlled as demonstrated in Figure 3. The hazard distance for unignited hydrogen release is higher for a horizontally oriented leak as opposed to the diagonal or vertical. In many cases vertical, unignited jet, which happens outdoors will not pose any hazard to people or cause damage to buildings and structures. For Responders, however, it is important to know whether a leak is originally momentum- or buoyancy-controlled, or at what hydrogen concentration the flow regime changes from momentum to buoyant for the same jet (for example, in the case of vertical downward directed leaks).

The technique presented below, allowing distinguishing between momentum- and buoyancy-controlled flow in expanded and under-expanded jets, is based on the work of Shevyakov et al. [17, 18] that was carried out with the expanded jets only. Figure 7 shows, in logarithmic coordinates, the dependence of the ratio of distance to nozzle diameter $x/D$ (ordinate) for a particular volumetric concentration of hydrogen in air on the *Froude number Fr* (abscissa) in its classical form:

$$Fr = U^2/gD \tag{5}$$

where $U$ is the velocity at the nozzle exit (notional nozzle exit for under-expanded jets), m/s, $g$ is the gravitational acceleration (standard acceleration of gravity on Earth is 9.80665 m/s$^2$), $D$ is the nozzle diameter (notional nozzle exit diameter for under-expanded jet), m.

For under-expanded jets in Figure 7 parameters to be used in (5) - the notional nozzle exit diameter $D$ and the velocity at the notional nozzle exit $U$ - were calculated by the *under-expanded jet theory* [11]. Both expanded and under-expanded jets obey the same functional dependence with accuracy of 20% acceptable for engineering applications.

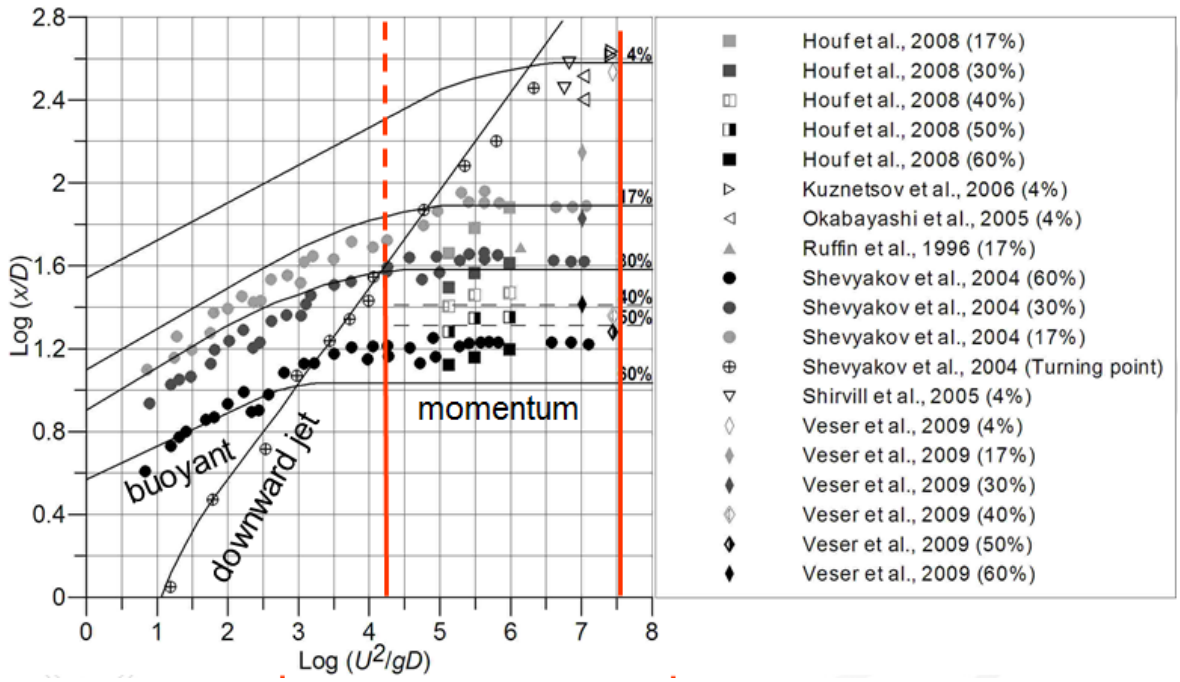



Figure 7. The dependence of the distance to nozzle diameter ratio for particular concentration of hydrogen in air on the Froude number [17, 18].

Figure 7 shows five theoretical curves (solid lines) and the experimental data by Shevyakov and co-authors for expanded jets [17, 18], and the data by other researchers for under-expanded jets. Practically all under-expanded jets in hydrogen-related incidents/accidents will be in the momentum-controlled regime as follows from the available tests applied to validate the correlation in Figure 7. Four of five theoretical curves in the graph are related to hydrogen concentrations of 4 vol. %, 17 vol. %, 30 vol. %, and 60 vol. % respectively. Each of these four curves has an ascending buoyant part and a momentum "plateau" part [1].

It is important to stress that the Froude number (*Fr*) at the transition from the buoyant part of the curve to the momentum one depends on the concentration of hydrogen under consideration. For example, at the axial concentration of hydrogen equal to 60 vol. % the jet stays in momentum-controlled regime until the *Log* (*Fr*) > 3.5, while for the jet to be in the momentum-controlled regime at the axial location where hydrogen concentration drops to 4 vol. % (LFL) the nozzle exit Froude number has to be much higher, i.e. *Log* (*Fr*) > 6.5 (i.e. three orders of magnitude higher) [1].

The fifth curve on Figure 7 labelled as the "downward jets" is of a special interest for those who will make decisions at the scene of an accident. It gives for a jet directed vertically downward a dimensionless distance from the nozzle to the turning point, where the jet changes its direction of flow from downward to upward. To calculate the distance to the turning point only knowledge of the Froude number at the nozzle (notional nozzle) is required. The fifth curve intersects each of the four other curves in Figure 7 in the region of *transition from momentum-dominated to buoyancy-controlled flow* as expected [1]. In order to use the correlation in Figure 7 the following steps should be applied:

1. Calculate the nozzle exit Froude number, *Fr*, and its logarithm. Apply the under-expanded theory to calculate the notional nozzle exit diameter and the velocity in the notional nozzle exit for under-expanded jets.
2. Draw a vertical line upward from the point on the abscissa (horizontal) axis equal to the calculated Froude number logarithm. The intersection of this vertical line with the line marked as "downward jets" on the graph indicates the concentration, above which the jet is qualified as momentum-dominated (solid red line), and below which the jet is qualified as buoyancy-controlled (dash red line).

For example, if a jet exit Froude number is *Log* (*Fr*) = 4.25, the intersection of the vertical line with the line "downward jets" is at the location of the theoretical curve corresponding to 30 vol. %. Thus, the jet is in the momentum-dominated regime when the concentration in the jet is above 30 vol. % and it becomes buoyant when the concentration on the jet axis is below 30 vol. % (further downstream of the axial concentration of 30% by volume).

This technique is quite simple to apply and at the same time can be very useful for Responders. For instance, the hazard distance for a horizontal jet release can be essentially reduced as only a length of the momentum-dominated part of the jet can be taken as an indication of the hazard distance rather than aggregated (both momentum- and buoyancy-controlled parts of the jet) distance to 4% by volume (LFL) [1].

## 6.2 What is a safe PRD diameter?

The similarity law (2) is a simple and a thoroughly validated tool for hydrogen safety engineering for both expanded and under-expanded round jets. Let us consider the following scenario: a FC forklift is parked in a warehouse. The forklift is equipped with a hydrogen storage tank fitted with a pressure relief device (PRD). Let us calculate what diameter the PRD should have to provide a safe upward release of hydrogen from the on-board storage at 35 MPa. In this case we would like to exclude the formation of a flammable layer under a warehouse ceiling, which is 10 m above the PRD. To realize this safety strategy the concentration on the jet axis at a distance of 10 m should be equal to or less than 4 vol. % (corresponding mass fraction of hydrogen is 0.00288). The density of hydrogen in the nozzle exit, calculated by the under-expanded jet theory for the storage pressure of 35 MPa, is $\rho_N$ = 14.4 kg/m$^3$. Thus, the diameter of the PRD can be calculated from the similarity law (2) as equal or less than 1.5 mm:

$$D = \frac{C_{ax}^m}{5.4}\sqrt{\frac{\rho_S}{\rho_N}}x = \frac{0.00288}{5.4}\sqrt{\frac{1.205}{14.4}}10 = 0.0015m\,, \qquad (6)$$

To finalize this safety strategy for the use of hydrogen-powered forklifts in the warehouse a requirement to the fire-resistance rating of the on-board storage tank must be formulated and testing carried out. Indeed, the fire-resistance rating should be greater than the blow-down time (i.e. the time needed to empty the storage vessel) of the storage tank to exclude its catastrophic failure in the case of external fire. It is clear that the use of a PRD with a larger diameter would

create either a flammable cloud or a jet flame with higher hazards, e.g. dangerous overpressure during "delayed ignition" or deflagration of initial cloud, and associated risks [1]. The ignited hydrogen release (or jet fires) will be discussed in a subsequent lecture.

## 7. Blow-down of a compressed hydrogen storage tank

A blow-down is a process associated with the emptying of storage tank content through an orifice of a TPRD/vent/leak. The blow-down time will depend on the mass of hydrogen stored, volume of the tank, the pressure, and the orifice diameter.

For example, Tables 2 and 3 indicate volumes, pressures, weights and blow-down times to atmospheric pressure (in s) of hydrogen for on-board storage of selected demonstration or prototype FC vehicles. For a vehicle with the highest volume of storage tank (vehicle 3, 156 L, 70 MPa) the blow-down time is the highest. Although for typical orifice diameters of leaks, between 4 and 6 mm, the blow-down time ranges from 124 s (just over 2 minutes) to 54 s (slightly less than 1 minute), implying that this is a very rapid process.

Table 2. The examples of the on-board hydrogen storage parameters for different FC vehicles

| Vehicles | Manufacturer | Model | Pressure (MPa) | Volume (L) | Weight (kg) | Status |
|---|---|---|---|---|---|---|
| 1 | Hyundai | Tucson Hybrid FCEV | 35 | 152 | 3.6 | Demonstration |
| 2 | VW | Touran HyMotion | 35 | 81 | 1.9 | Prototype |
| 3 | Toyota | FCHV-adv (2008) | 70 | 156 | 6.2 | Prototype |
| 4 | VW | Tiguan HyMotion | 70 | 81 | 3.2 | Vehicle Testing |

Table 3. Adiabatic blow-down times calculated for different leak diameters for on-board storage tanks of vehicles 1 - 4

| Leakage diameter [mm] | Blow-down time vehicle 1 (s) | Blow-down time vehicle 2 (s) | Blow-down time vehicle 3 (s) | Blow-down time vehicle 4 [s] |
|---|---|---|---|---|
| 2 | 427 | 226 | 502 | 260 |
| 3 | 189 | 100 | 222 | 115 |
| 4 | 105 | 56 | 124 | 64 |
| 5 | 67 | 35 | 79 | 41 |
| 6 | 46 | 24 | 54 | 28 |

Figure 8 demonstrates how the pressure reduces inside the storage tanks during the blow-down. As you can see the drop in the pressure is significant: it reduces to atmospheric values rapidly.

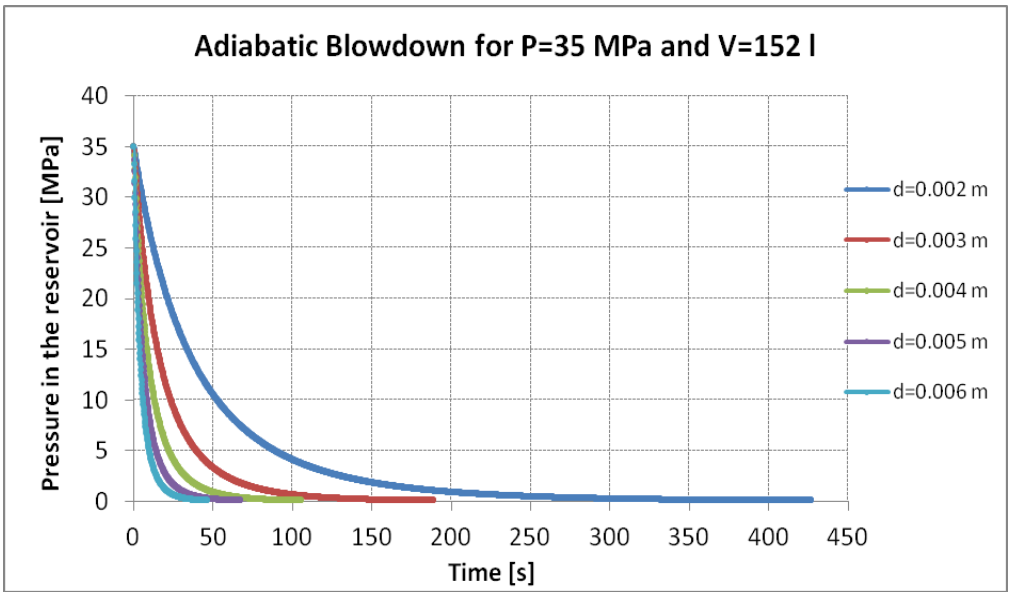



Figure 8. A pressure drop inside a hydrogen storage tank as a function of time for different leak diameters (vehicle 1).

A CFD study of unignited hydrogen releases from on-board hydrogen storage of a FC car was carried out at HySAFER centre of Ulster University by Li et al. [19] in 2015, that considered an accident scenario involving a vertical downward release of hydrogen from a thermally activated pressure relief device (TPRD) under a FC car located outdoors (i.e. in the open atmosphere). An unignited release of hydrogen may occur due to an unexpected initiation a faulty TPRD, for example following a severe car crash. TPRDs are usually located underneath the FC vehicle (Figure 8), and their vents orientated vertically downwards, therefore the release of hydrogen will impinge the ground [19]. The released hydrogen can form a flammable cloud that has the potential to result in a flash fire.

The dimensions of a typical salon car used in [19] are shown in Figure 9. The authors assumed a full on-board hydrogen tank with a volume of 171 litres, adopted from the Honda FCX specification [20]. Two storage pressures were considered: 35 and 70 MPa. The TPRD is assumed to be located near the rear wheel under the vehicle as shown in Figure 9 and its orifice diameter is 4.2 mm [21]. Other assumptions include: the ambient pressure and temperature at 1 atm and 20 °C, respectively; no wind in the environment [19].

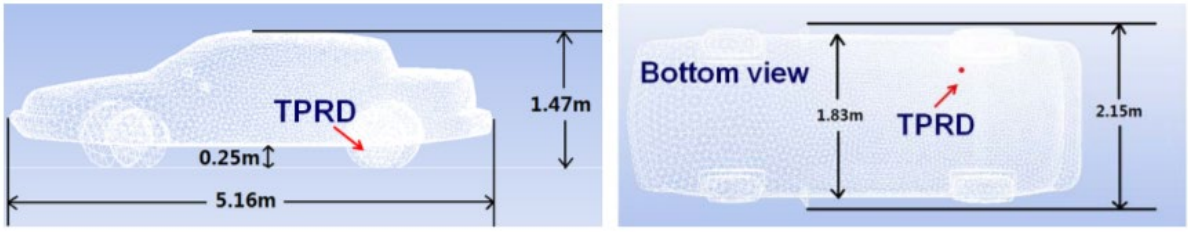



Figure 9. Geometry of a FC car and location of a TPRD [19].

In this study, the conservative value of hydrogen concentration equal to 4 vol. % is applied as the harm criterion for the general public, while 8 vol. % - as the harm criterion for Responders

who are not equipped with thermal protective clothing, such as policemen and emergency medical personnel. For the firefighters having a bunker gear, the hydrogen flammable cloud will be harmless as the gear will protect them from the potential hydrogen flash fire. In addition to the harm criteria to people, 4 vol. % hydrogen concentration was selected as the damage criterion to the air intake of buildings. If the flammable envelope of 4 vol. % hydrogen concentration reaches the location of an air intake into a high-rise building, then the consequences for both the occupants and the structure of the building could be catastrophic [19].

In real-world conditions, a hydrogen release from a high-pressure tank is not a steady-state release but a blow-down process with pressure reducing in the reservoir until the tank is empty. The notional nozzle model mentioned earlier can be applied to simulate the pressure dynamics in the hydrogen storage tank during an under-expanded jet. The Cyber Laboratory tool 'Adiabatic blow-down of storage tank' was used. The obtained results are shown in Figure 10. For the release from the 4.2 mm orifice, 171 L tank at 35 MPa, the total blow-down time is less than 110 seconds (< 2 minutes), and the transition from an under-expanded to an expanded jet occurs at 85 s. For the release from the 4.2 mm orifice, at 70 MPa with identical mass, the total blow-down time is less than 75 s, with the transition from an *under-expanded* to an *expanded jet* occurring at 58 s [19].

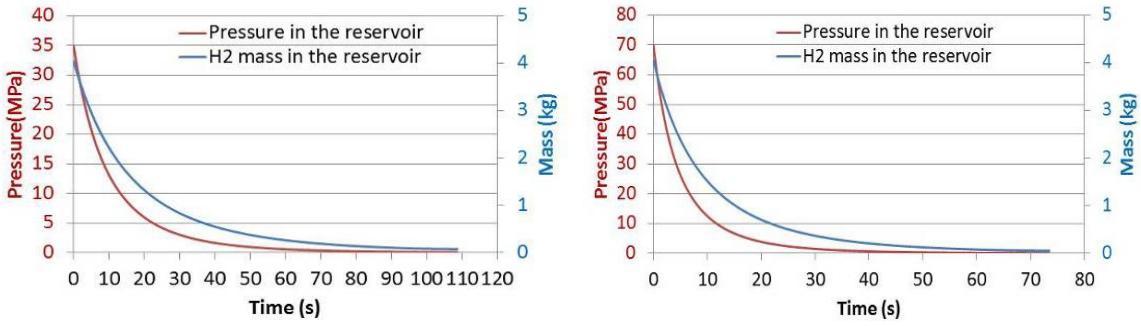
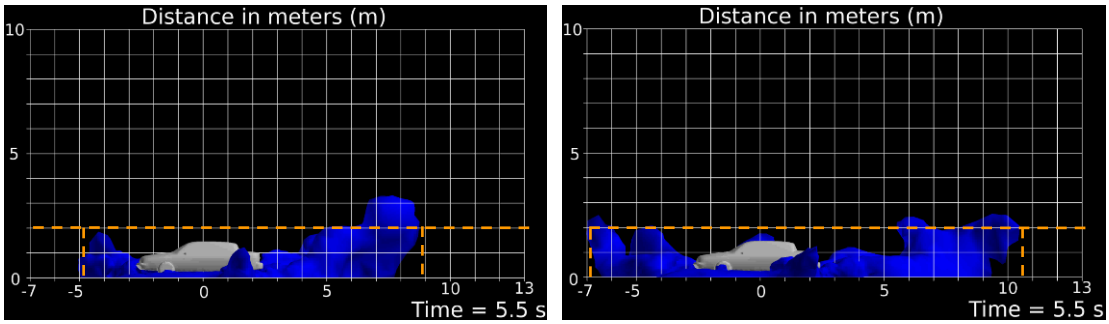


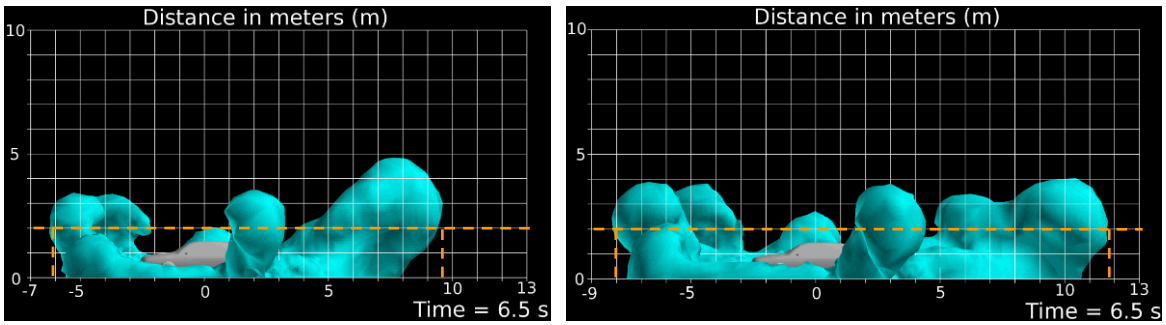

Figure 10. Adiabatic blow-downs (reduction of pressure and hydrogen mass) of storage tanks at 35 MPa (a) and 70 MPa (b) (orifice diameter - 4.2 mm) [19].

As you can see from Figure 11 for the Responders without thermal protective clothing, the longest hazard distance from the point of the release is 8.8 m and 10.5 m, for 35 MPa and 70 MPa storage pressures, respectively. The hazard distance for the release from hydrogen storage at 70 MPa is nearly 20% higher compared to 35 MPa storage pressure. In both cases the largest sizes of 8 vol. % hydrogen envelopes near the ground were formed at 5.5 s after the opening of the TPRD. Afterwards, the envelopes will shrink, and the hazard distances decrease.




(a) (b)

Figure 11. 8 vol. % hydrogen envelopes registered at 5.5 s after start of release: a) for 35 MPa storage pressure, b) for 70 MPa storage pressure (longest hazard distance extend assuming Responders height cut-off at 2 m) [19].

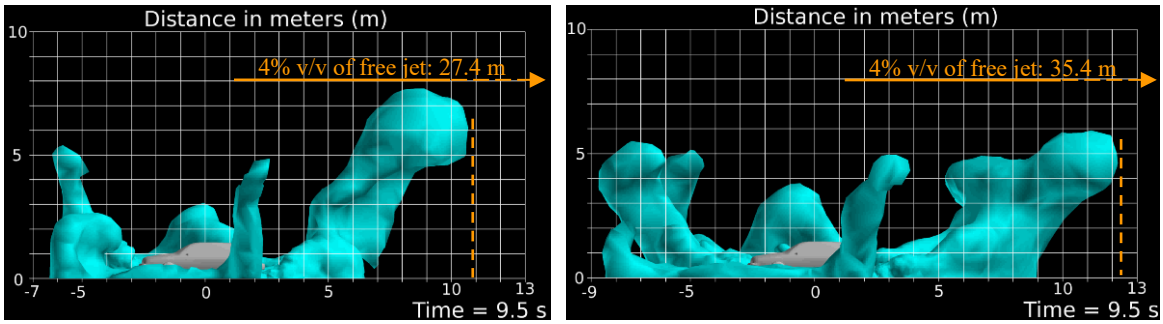



(a) (b)

Figure 12. 4 vol. % hydrogen envelopes registered at 6.5 s after start of release: a) for 35 MPa storage pressure, b) for 70 MPa storage pressure (longest hazard distance extend assuming Responders height cut-off at 2 m) [19].

As it shown in Figure 12 for the general public on the ground, the longest hazard distances for 35 and 70 MPa releases are 9.4 and 11.8 m, respectively. In both cases the largest 4 vol. % hydrogen envelopes near the ground were registered at 6.5 seconds after the initiation of the TPRD. Afterwards, the envelopes as well as the hazard distances will reduce. Figure 12 only represents 4 vol. % envelopes for the height lower than 2 m (orange cut-off lines). The largest flammable envelopes for the total domain are shown in Figure 13, from which the hazard distances for the surroundings can be evaluated [19].

It can be seen from Figure 13 that for a building air intake, the longest hazard distances are equal to 10.7 m and 12.3 m, for 35 MPa and 70 MPa releases, respectively. The hazard distance for the 70 MPa release is increased by 15% compared to the 35 MPa release. In both cases the largest 4 vol. % hydrogen envelopes occur at 9.5 seconds after the opening of the TPRD. Afterwards, the envelopes will shrink, and the hazard distances will decrease. If the hazard distances from these impinged jets are compared with the hazard distances calculated for free jets (27.4 m for 35 MPa storage pressure and 35.4 m for 70 MPa storage pressures) , the results show a significant reduction of more than 60% and 65%, for the 35 MPa and 70 MPa releases respectively, as shown in Figure 13.




(a) (b)

Figure 13. The extent of 4 vol. % hydrogen envelopes for unignited releases from storage tanks: a) pressurised to 35 MPa, b) pressurised to 70 MPa [19].

Thus, to sum up, the CFD study carried out by Li et al. [19] demonstrated that:

- for unignited hydrogen releases from storage tanks pressurised to 35 MPa and 70 MPa, the longest hazard distances occur within 10 s after the opening of the TPRD and the duration of the hazards associated with the release of hydrogen is less than 2 min;
- the deterministic hazard distances for unignited hydrogen releases from a TPRD, orientated vertically downwards under a FC car, are significantly shorter than those of free jets;
- for both the members of the general public and Responders not equipped with the thermal protective clothing maximum hazard distance from unignited release ranges from 8 to 12 m depending on storage pressure;
- to ensure that the concentration of hydrogen is always less than LFL (4 vol. %) at the location of the air intake of buildings, the hazard distance should be at least 11 m for 35 MPa releases and 13 m for 70 MPa releases.

Hydrogen unignited releases indoors will be discussed later in the Lecture ‘Confined spaces’.

Generally, the main safety strategy how to deal with a hydrogen leak is to minimize its mass flow rate, for example by reducing the size of any potential leak by keeping the pipe diameters as small as possible and to ‘let it go’ to prevent hydrogen accumulation to a hazardous level when a flammable hydrogen-air mixture represents unacceptable risks [1]. The prevention and mitigation of unignited hydrogen leaks are discussed below.

## 8. Prevention of hydrogen leaks

The initial steps, prior to the detection and mitigation of hydrogen releases, are the preventive measures to control the associated risks. Prevention measures are the safety measures that can act on the combination of causes of a hazardous event to prevent it from happening. These measures reduce the likelihood of the event. They aim to prevent a loss of tightness and an equipment failure. They include but not limited to the following measures:

- Prevent the formation of flammable atmospheres through an inherently safer design of FCH systems;
- Carefully select the materials for FCH systems to avoid hydrogen embrittlement;
- Minimize the quantity of hydrogen (and potentially oxygen) that is stored and involved in an operation;
- Equipment validation;
- Physical protection;
- Periodic leak checks;
- Periodic equipment inspection.

Prevention measures related to the mechanical integrity of hydrogen systems involve:

- Validating equipment design through performance testing;
- Periodical checks of the equipment once it has been installed.

The equipment can be removed from service at the end of the specified lifetime [22]. Design testing and validation aim to prove that the equipment suits the application, for which it is going to be used. Resistance and endurance tests can also be applied. Resistance tests are basic tests, when all the relevant characteristics of the system are tested (e.g. hydraulic pressure test). Endurance tests are more elaborate, when the system is subjected to cyclic loading, and the time until it fails characterises its endurance. A factor is applied to establish the service life, for example, a requirement to withstands 50,000 test cycles for a specified service life of 10,000 cycles [22].

The equipment subjected to mechanical integrity testing is listed below:

- Gaseous hydrogen container,
- Container valves,
- Flexible hoses,
- Quick connection devices.

Inherently safer design is an approach that focuses on reducing or eliminating hazards associated with the product or the process. Let us consider how safety of a fuel cell (FC) system could be improved by reducing hazards without the interfering the technology itself. Unfortunately, current FC systems are often designed using piping diameters of 5 - 15 mm and pressures of 0.5 - 1.5 MPa without consideration of hazards. The mass flow through a 5 mm diameter orifice at storage pressure of 0.5 MPa can be calculated using the *under-expanded jet theory*, and is about 6 g/s. For a pipe of 15 mm diameter and pressure 1.5 MPa the mass flow rate is 170 g/s [1].

Now let us estimate the mass flow rate for a 50 kW FC system, which provides energy to large facilities such as hotels, hospitals, office buildings, and multi-family dwellings. Assuming that an electrical efficiency of a FC is 45%, and the upper heat of reaction (combustion) of hydrogen with air is [(286.1 kJ/mol)/(2.016 g/mol)] = 141.92 kJ/g, the mass flow rate for the FC functioning at maximum power can be calculated as (50 kW)/0.45/(141.92 kJ/g) = 0.78 g/s. For example, this mass flow rate can be provided at a pressure 0.5 MPa through a restrictor in the storage or piping system with an orifice diameter of only about 1.8 mm, or at pressure 0.2 MPa through a diameter orifice of about 2.9 mm [1].

As it is shown earlier in this lecture, the hazard distance for an unignited release is proportional to a nozzle diameter and square root from a storage pressure. Thus, the decrease of a pipe diameter from 15 to 2.9 mm and reduction of pressure from 1.5 to 0.2 MPa could decrease the hazard distance by more than 14 times [1].

Further analysis can be performed to compare hazard distances for these two options:

- option 1 – pressure 0.5 MPa and pipe diameter 1.8 mm,
- option 2 – pressure 0.2 MPa and pipe diameter 2.9 mm.

The ratio of hazard distances for unignited releases in option 1 and 2 in assumption of full-bore rupture can be estimated as 0.98, i.e. they are practically the same. These examples clearly demonstrate the advantages of the science-informed safety design of hydrogen and fuel cell systems to essentially reduce hazard distances without affecting the performance parameters of FC [1].

## 9. Mitigation measures for unignited releases

Once the prevention steps have been taken thought can be given to detection and mitigation measures. The safety measures aimed at mitigating or reducing the severity of the consequences of the hazardous event are called mitigation measures [22]. In terms of mitigating the risk the number of people at risk should be reduced as much as possible and an explosion relief, suppression or containment can be provided. For detection of a leak, the gas sensors may be used if applicable. Flowmeters, IR detectors, etc. are also relevant. Some mitigations measures are listed below [22, 23]:

- reduce the size of a leak, for example through the use of hydrogen flow restrictors;
- isolate hydrogen from oxidizers, hazardous materials and dangerous equipment;
- use of alarms and/or warning devices (including hydrogen sensors and fire detectors);
- use of shut-down devices and systems;
- identify and, if possible, separate or eliminate potential ignition sources;
- prevention flammable atmospheres forming, e.g. by ventilation (natural and active);
- elevation of hydrogen systems, use of flow meters, etc.;

- use outdoor location when possible;
- avoid congestion;
- implement hazard distances;
- practice good housekeeping, such as keeping access and evacuation routes clear and keeping weeds and other debris away from hydrogen systems;
- provision of the emergency response.

## 10. Detection of hydrogen leaks

Because hydrogen is colourless and odourless, sensors will be key safety equipment for safe refuelling stations and other hydrogen facilities. The addition of an odorant to hydrogen would ease the detection of small leaks. However, this is not practicable in most situations, e.g. this would poison an expensive catalyst in the fuel cells. Detectors/sensors can be used to detect hydrogen releases, to automatically shut down systems, to activate the alarms, and to notify Responders. Hydrogen sensors are the subject of ongoing research worldwide.

The following criteria are applied to the selection of hydrogen sensors:

- durability and reliability;
- sensitivity to cross-contamination from e.g. hydrocarbons;
- sensitivity to humidity;
- response time;
- whether they will give false positive readings;
- sensitivity to temperature extremes;
- maintenance requirements;
- measurement range;
- an accuracy.

Focus areas addressed by FCTO (fuel cell technologies office) are summarized in the Hydrogen, Fuel Cells & Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan (the Multi-Year Project Plan, or MYPP), which was published in 2007 [24] and updated in 2012 [25]. A short list of critical hydrogen sensor parameters with target specifications was identified in 2007. The table summarizing these metrics and corresponding specifications is reproduced in Figure 14. A nearly identical table was included in the 2012 MYPP [25].

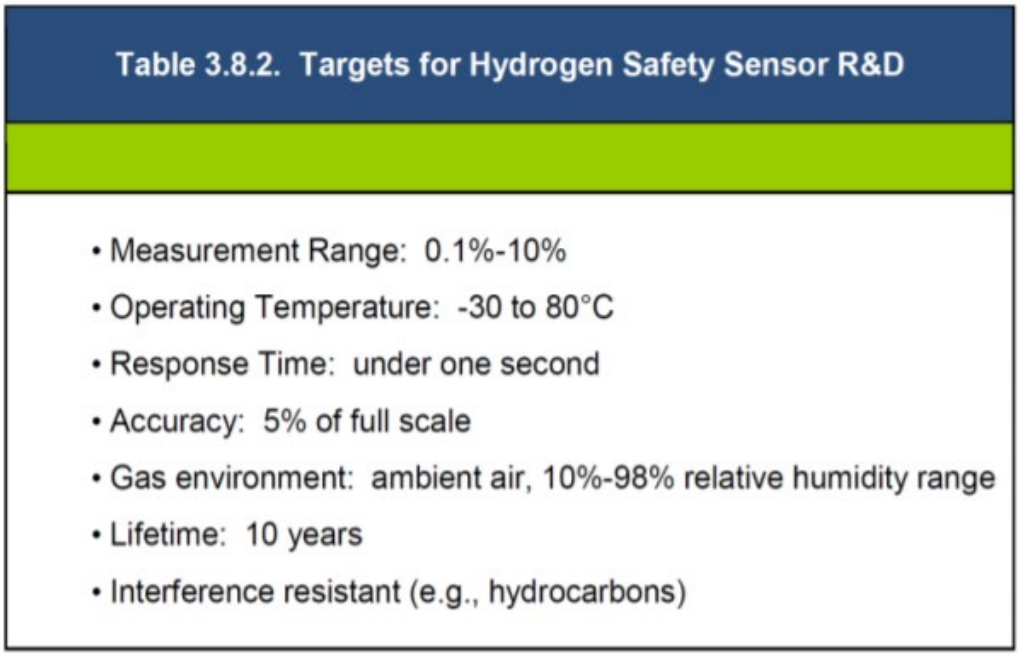



Figure 14. Targets for hydrogen sensors outline by US DoE [26].

It is also important to know that in hydrogen safety technologies sensors do not provide a complete detection strategy due to the buoyancy and diffusivity of hydrogen. For example, a hydrogen sensor will be of little use in a large enclosure or outdoors. The placement of sensors should be carefully considered and tools such as CFD may be used to simulate leak scenarios to provide insight into sensor positioning. Both fixed location and personal/hand-held monitors are necessary for the protection of personnel and facilities.

The suggested positioning of hydrogen sensors is detailed below:

- locations where hydrogen leaks or spills are possible;
- at hydrogen connections that are routinely separated (for example, hydrogen refuelling ports);
- locations where hydrogen could accumulate;
- in building air intake ducts, if hydrogen could be carried into the building;
- in building exhaust ducts if hydrogen could be released inside the building.

There are also requirements to fit hydrogen sensors on FC vehicles to warn about potential leaks. Hydrogen detectors locations for Fuel Cell Electric Vehicle (FCEV) are marked as blue dots on Figure 15 and include [27]:

- exhaust pipe (process control) ;
- passenger cabin (safety) ;
- engine (safety) ;
- fuel Cell stack (safety).

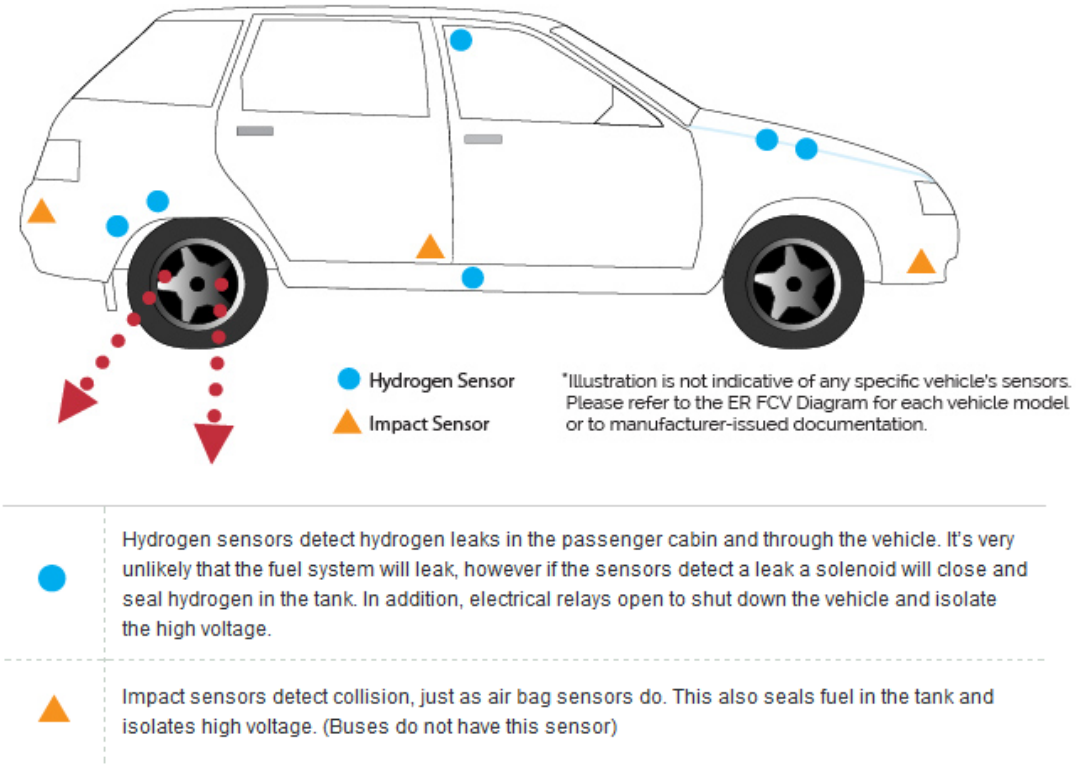



Figure 15. Possible location of hydrogen sensors in a FCEV [27].

A commonly used concentration level for main alarm is 1 vol. % of hydrogen in air, which is equivalent to 25% of the LFL. This level normally should provide an adequate time to respond in an appropriate manner, such as system shutdown, evacuation of personnel, or other measures, as necessary. A warning may be given earlier. More than one sensor platform necessary to reach all target specifications and a combination of sensor platforms shows the best results.

The types of commercially available hydrogen sensors/detectors are:

- electrochemical detectors;
- metal oxides detectors;
- thermal conductivity detectors ;
- field effect gas detectors (FED);
- resistance-based palladium thin film;
- catalytic detectors;
- micro electro mechanic systems (MEMS);
- optical devices;
- research is on-going.

The following factors should be considered while selecting the detectors [28]:

- accuracy (1-10%);
- reliability;
- maintainability;
- calibration;
- detection limits (high and low);
- response time (<10 s);
- recovering or non-recovering in time;
- long life time (more than 5 years);
- low energy consumption (< 10 MW);
- simple system integration.

In 2009 INERIS (France) conducted a test programme within the HYPER project, 2008 [29] based on the international standard parts IEC 61779-1&4, 1998 [30] and aimed at assessing the performance of commercially available hydrogen detectors. These devices were of electrochemical and catalytic types, i.e. the two types most often used in industry. The catalytic sensor was 5 times faster than the electrochemical one to respond to a sudden exposure to hydrogen. However, the response time was approximately 10 s for the catalytic sensor and 50 s for the electrochemical sensor. These figures also apply for the recovery time. In many practical scenarios this long time is hardly acceptable [1].

Catalytic detectors studied within the HYPER project were also prone to loss the sensitivity and drift of zero after a prolonged exposure to hydrogen. This emphasizes the need to regularly calibrate these devices. Higher humidity tended to increase the reading of the catalytic detector for constant hydrogen content. The catalytic detector was very sensitive to the presence of carbon monoxide (CO) but the interfering was only temporary, i.e. when the CO exposure ceases the detector behaves in an ordinary way [1].

Research carried out at the Joint Research Centre, Institute for Energy, The European Commission within the HYPER project demonstrated that the time required by the electrochemical sensor to respond to hydrogen exposure of known concentration becomes longer when the gas flow rate is reduced, i.e. it could be twice longer by solely reducing the flow rate from 100 to 30 ml/min. This finding is particularly important when the sensor is intended to control the formation of an explosive atmosphere within a FC cabinet [1].

There is another issue related to faster catalytic sensors that is not yet sufficiently addressed in the literature. This is a potential to ignite hydrogen-air mixture with high concentrations of hydrogen by the sensor. The ignition of hydrogen-air mixtures with high content of hydrogen by recombiners was already observed [31].

A variety of methods and sensors types are commercially available to detect the presence of hydrogen [23]. Many of these detectors are suitable for the use in automatic warning and operating systems, see for example ISO 26142:2010 [32] for details concerning stationary systems.

# 11. e-Laboratory tools for relevant phenomena

The introduction of e-Laboratory has been given in Lecutre 1. Users will need to log in e-Laboratory through https://hyresponder.eu/e-platform/e-laboratory/ and select the relevant tools from the list. In this section, the tools of 'Jet parameters model', 'Similarity law for concentration decay and hazard distance' and 'Buoyancy calculation' were discussed in details.

## 11.1 Jet parameters model

This model describes the parameters in either an expanded or an under-expanded jet. As shown in Figure 16 is a simple schematic of an 'expanded' (red lines) and an 'under-expanded' (black lines) jet, in comparison with Figure 4.

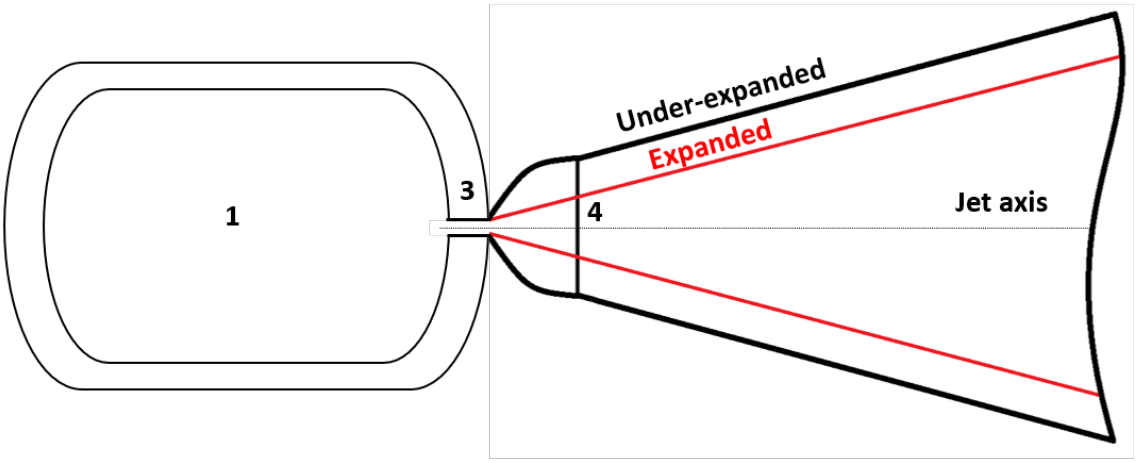



Figure 16. Simple schematic of 'expanded' (red lines) and 'under-expanded' (black lines) jet. 1 – Reservoir (high-pressure vessel), 3 – Nozzle or orifice exit (notional entrance), 4 – notional nozzle/orifice exit (where pressure and velocity is equal to ambient and local speed of sound, respectively). Note that exit 4 is only for under-expanded jet.

The critical pressure ratio for the transition to sonic flow for hydrogen is approximately 1.9. If the critical pressure ratio is smaller than 1.9, the flow is subsonic and expanded, if it is larger than 1.9, the exit velocity remains locally sonic and the jet is under expanded. The exit pressure rises above ambient with the result that expansion, down to ambient pressure, $p_{atm}$, takes place outside of the real nozzle at location downstream of the Mach disc, called the "notional" nozzle or effective diameter.

The critical pressure ratio across the leak, $p^*$, determines whether the flow is subsonic or sonic/supersonic.

$$p^* = \frac{p_1}{p_{atm}} = \left(\frac{(\gamma+1)}{2}\right)^{\gamma/(\gamma-1)} \quad (6)$$

Therefore, different equation sets are solved depending on whether the flow is expanded or under-expanded. The equation set relating to an under-expanded jet is detailed in Molkov [11] and is based on the Abel-Noble equation of state for hydrogen; conservation equations for mass and energy; assumption that at state (4) (so-called "effective nozzle diameter") pressure is equal to ambient and velocity is equal to the local sound speed.

The expanded jet equation set utilises the isentropic pressure and density relationships.

The input parameters include orifice diameter $d_3$, ambient pressure $p_4$, hydrogen pressure in reservoir $p_1$, and hydrogen temperature in reservoir $T_1$. Once the above parameters were defined, the values of a set of calculated parameters are obtained, as shown in Table 4.

Table 4 The input and output parameters of the jet parameters model

| Name | Symbol | Value | Unit |
|---|---|---|---|
| Orifice diameter | $d_3$ | 0.0095 | m |
| Ambient pressure | $p_4$ | 1.01325e+5 | Pa |
| H2 pressure in reservoir | $p_1$ | 2.05e+7 | Pa |
| H2 temperature in reservoir | $T_1$ | 288 | K |
| H2 density in reservoir | $\rho_1$ | 15.2366 | kg/m³ |
| H2 density at orifice | $\rho_3$ | 9.48882 | kg/m³ |
| Pressure in orifice | $p_3$ | 9.83938e+6 | Pa |
| Velocity in orifice | $V_3$ | 1253 | m/s |
| Temperature at the orifice | $T_3$ | 233 | K |
| Diameter of effective nozzle exit | $d_4$ | 0.0942914 | m |
| Density in effective nozzle exit | $\rho_4$ | 0.1025 | kg/m³ |
| Velocity in effective nozzle exit | $V_4$ | 1178 | m/s |
| Temperature in effective nozzle exit | $T_4$ | 239.501 | K |
| Mass flow rate | $\dot{m}$ | 0.84318 | kg/s |

## 11.2 Similarity law for concentration decay and hazard distance

This model allows for prediction of axial concentration decay of a leaking gas for sub-sonic, sonic, and super-sonic jets. The similarity law is shown to be valid in a wide range of conditions from expanded to highly under-expanded jets. The model is used for calculation of hazard distances formed by the size of the flammable envelope.

Analysis of experimental data has demonstrated that the longest, and therefore most hazardous, flames can be expected to be formed from under-expanded jets. With this in mind, the similarity law [33] for axial concentration decay in round unignited, expanded, momentum-dominated jets, as suggested by Chen and Rodi [34], has now been expanded and validated for under-expanded hydrogen jets [1].

The equation implemented to calculate the axial distance from the nozzle, to a prescribed concentration of hydrogen can be written as:

$$x = \frac{5.4 \times D}{C_{ax}} \cdot \sqrt{\frac{\rho_N}{\rho_S}} \quad (7)$$

where $x$ is the axial distance from the nozzle, $D$ is nozzle diameter, $C_{ax}$ is mass fraction of hydrogen at axial distance $x$, $\rho_N$ is density at nozzle exit, and $\rho_S$ is air density.

When considering under-expanded jets knowledge of density in the nozzle, $\rho_N$, (as a function of storage pressure and losses in the leak pathway) is required. This density is calculated using the 'hydrogen jet parameters' engineering tool which was described in Section 10.1. For expanded jets, calculations are based on isentropic pressure and density relationships (this equation set is described in more detail in the description of the 'flame length and separation distance for jet fires' engineering tool).

A correlation between the location of the hydrogen flame jet tip and the location of three different hydrogen concentrations (8%, 11% and 16%) in an unignited jet emanating from the same leak source.

This engineering tool calculates the axial distance from the nozzle corresponding to the location of various pre-selected hydrogen concentrations, namely 4%, 8%, 11%, 16% and 29.5% by volume, in unignited jets. This model also allows the 'user' to choose their own concentration for analysis, within the hydrogen flammability limits of 4% to 75% by volume.

An example of the input and output parameters and their values of this tool are shown in Table 5.

Table 5 Input and output parameters of this tool

| Name | Symbol | Value | Unit |
|---|---|---|---|
| H2 pressure in reservoir | $p_1$ | 3.5e+7 | Pa |
| H2 temperature in reservoir | $T_1$ | 293 | K |
| Orifice diameter | $d_3$ | 0.005 | m |
| Ambient pressure | $p_4$ | 1.01325e+5 | Pa |
| Ambient temperature | $T_{atm}$ | 293 | K |
| Axial distance from nozzle to 4% by vol. H2 | $X_{4\%,H_2}$ | 32.6212 | m |
| Axial distance from nozzle to 8% by vol. H2 | $X_{8\%,H_2}$ | 15.6793 | m |
| Axial distance from nozzle to 11% by vol. H2 | $X_{11\%,H_2}$ | 11.0593 | m |
| Axial distance from nozzle to 16% by vol. H2 | $X_{16\%,H_2}$ | 7.20885 | m |
| Axial distance from nozzle to 29.5% by vol. H2 | $X_{29.5\%,H_2}$ | 3.33269 | m |
| Axial distance from nozzle to ?% by vol. H2 | $X_{?\%,H_2}$ | 5.50167 | m |

## 11.3 Buoyancy calculation

This tool shows effect of the hazard distance decrease due to buoyancy in unignited releases. It allows to calculate the distance from the release nozzle to the concentration at which release changes from momentum-controlled regime to buoyancy–controlled, as well as the hazard distance to concentration of 4% by volume of hydrogen.

All jets can be divided into three types based on the effect of buoyancy. These jet types are shown schematically in 错误!未找到引用源。 for a horizontal jet. Fully momentum-controlled jets are not affected by buoyancy. Fully buoyancy-controlled jets are quickly diverted from the horizontal to vertical flow direction. Third type of jets that can be called transitional have momentum-dominated part closer to the nozzle and buoyancy-controlled flow further downstream when jet velocity drops and jet diameter increases. For hydrogen safety engineering it is important to know when this transition takes place. This has direct implication on the separation distance and thus safety and costs of hydrogen system or infrastructure.

The engineering tool presented here to qualify a hydrogen jet (both expanded and under-expanded) or its part as momentum-controlled, and the rest of the jet downstream as buoyancy controlled, is based on the work by [17, 18] that was carried out with expanded jets only. The dependence of the distance to nozzle diameter ratio (x/D) on the Froude number (Fr), for a particular concentration of hydrogen in air, is expressed Eq. (5). For under-expanded jets, the notional nozzle exit diameter and velocity at the notional nozzle exit were calculated by the under-expanded jet theory [11]. Both expanded and under-expanded jets obey the same functional dependence with accuracy 20% acceptable for engineering applications.

Five theoretical curves (solid lines) and experimental data for expanded jets [17, 18], and data of other researchers for under-expanded jets are. Practically all under-expanded jets in hydrogen incidents/accidents will be in the momentum-controlled regime. Four of five theoretical curves in the graph are related to hydrogen concentrations of 4%, 17%, 30%, and 60% by volume respectively. Each of these four curves has an ascending buoyant part and a momentum “plateau” part.

It is important to stress that the Froude number at transition from the buoyant part of the curve to the momentum part depends on the concentration of hydrogen under consideration. For example, at axial concentration of hydrogen 60% by volume the jet stays in momentum-controlled regime until Log (Fr) > 3.5, while for the jet to be in the momentum-controlled regime at axial location where hydrogen concentration drops to 4% by volume (LFL) the nozzle exit Froude number has to be much higher, i.e. Log (Fr )> 6.5 (three orders of magnitude higher).

The curve “Downward jets” is of special interest for hydrogen safety engineering. It gives for a jet directed vertically downward a dimensionless distance from the nozzle to the turning point, where the jet changes direction of flow from downward to upward. To calculate the distance to turning point only knowledge of the Froude number at the nozzle (notional nozzle) is required. The fifth curve intersects each of the four other curves in the region of transition from momentum-dominated to buoyancy-controlled flow as expected.

There are two sub-tools about the effect of buoyancy on decrease of hazard distance for unignited releases in e-Laboratory to answer the following two questions.

- Q1: for horizontal release the distance and concentration at which release becomes buoyant and hazard distance to 4% v/v along the jetcentral line.
- Q2: for vertical downward release the distance and concentration at which flow start to go up due to buoyancy.

The input parameters of this tool are the hydrogen pressure in reservoir $p_1$, hydrogen temperature in reservoir $T_1$, orifice diameter $d_3$ and ambient pressure $p_4$. As an example, the output parameters of the tool for Q1 are listed in Table 6.

Table 6 An example of the input and output parameters of this tool

| Name | Symbol | Value | Unit |
|---|---|---|---|
| H2 pressure in reservoir | $p_1$ | 7e+6 | Pa |
| H2 temperature in reservoir | $T_1$ | 288 | K |
| Orifice diameter | $d_3$ | 0.02 | m |
| Ambient pressure | $p_4$ | 1.01325e+5 | Pa |
| Axial distance from nozzle to $C_B$% by vol. $H_2$ where release becomes buoyant. | X | 35.6746 | m |
| Axial distance from nozzle to 4% by vol. $H_2$ | $X_{4\%}$ | 44.2411 | m |
| Axial distance from nozzle to 17% by vol. $H_2$ | $X_{17\%}$ | 9.24329 | m |
| Axial distance from nozzle to 30% by vol. $H_2$ | $X_{30\%}$ | 4.5659 | m |
| Axial distance from nozzle to 40% by vol. $H_2$ | $X_{40\%}$ | 3.08551 | m |
| Axial distance from nozzle to 50% by vol. $H_2$ | $X_{50\%}$ | 2.43964 | m |
| Axial distance from nozzle to 60% by vol. $H_2$ | $X_{60\%}$ | 1.28028 | m |

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