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DEVELOPMENT OF A REALISTIC HYDROGEN FLAMMABLE ATMOSPHERE INSIDE A 4-M^3 ENCLOSURE

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ABSTRACT

To define a strategy of mitigation for containerized hydrogen systems (fuel cells for example) against explosion, the main characteristics of flammable atmosphere (size, concentration, turbulence…) shall be well-known. This article presents an experimental study on accidental hydrogen releases and dispersion into an enclosure of 4 m^3 (2 m x 2 m x 1 m). Different release points are studied: two circular releases of 1 and 3 mm and a system to create ring-shaped releases. The releases are operated with a pressure between 10 and 40 bars in order to be close to the process conditions. Different positions of the release inside the enclosure i.e. centred on the floor or along a wall are also studied. A specific effort is made to characterize the turbulence in the enclosure during the releases. The objectives of the experimental study are to understand and quantify the mechanisms of formation of the explosive atmosphere taking into account the geometry and position of the release point and the confinement. Those experimental data are analyzed and compared with existing models and could bring some new elements to improve them.

1.0 INTRODUCTION

Since a few years, hydrogen appears as a credible energy-vector and some of those hydrogen applications can be containerized. However, hydrogen applications are still considered dangerous, indeed hazardous events like explosion could occur if a hydrogen-air mixture comes to be formed; and some accidents involving hydrogen, like Hindenburg disaster or more recently Fukushima Daiichi nuclear disaster, are still in the minds. And it should be recognised that hydrogen leaks can produce extended explosive clouds because of the broad flammability range, and that hydrogen-air mixtures ignite extremely easily and burn/explode fast and violently [1], [2].

That's why to define a strategy of mitigation for containerized hydrogen systems (fuel cells for example) against explosion, the main characteristics of flammable atmosphere (size, concentration, turbulence…) should be well-known. This article presents an experimental study designed to understand and quantify the mechanisms of accidental hydrogen release and dispersion into a 4 m^3 enclosure (2 m x 2 m x 1 m). A specific effort is made to characterize the turbulence (turbulent intensity and integral scale) in the enclosure during the releases.

1.1 Description of the existing experimental data of hydrogen dispersion

The GARAGE facility [3] is representative of a realistic single vehicle private garage, i.e. a rectangle with interior dimensions of 5.76 m (length) x 2.96 m (width) x 2.42 m (height). The GARAGE facility was equipped with two small openings at the rear near the door. Both are 200mm diameter openings and are equipped with a cap.

The leakage source is upwards, centred in the middle of the GARAGE floor and at a height of 220 mm from the floor. The release diameter was 70 mm. Either the upper or the lower vent is open and

the other vent is closed or both vents were open. The flowrate was from 0.1 to 18 Nl/min when the lower vent or the upper vent is open and was from 10 to 300 Nl/min when the both vents were open.

The GAMELAN enclosure [4] is a parallelepipede with a square base of 0.93m by 0.93m and 1.26m high. The vent is located on a side wall, near the ceiling (the upper side of the vent is 20 mm below the ceiling). Three vents were used; the larger is a rectangle vent of 90cm by 18cm; the two other vents have approximately the same area, the first is a square vent of 18cm by 18cm and the another is rectangle vent of 90cm by 3.5cm. Helium is injected from the bottom of the enclosure through a tube of 5mm or 20mm diameter, centred in the horizontal section and directed upward. The outlet of the injection tube is at 21cm from the floor. Several volumetric flow rates of Helium were tested from 1Nl/min to 300Nl/min.

Merilo et al. [5] studied hydrogen releases into a real scale garage. The dimensions of the facility were (H2.72xW3.63xL6.10) m3. The front face was made with a thin plastic sheet on which two openings were perforated. A rectangular lower vent (L1.22xH0.09) m² located near the floor and a circular vent (0.37 m diameter) at 2.42 m from the floor. Those two vents had the same area (0.11 m²). The injection diameter was 7.75 mm. The results of the tests 1 and 2 at around 9 kg/h showed a uniform ceiling layer, the momentum-induced forces dominate the buoyancy forces, i.e. the jet length Lm is greater than the height of the enclosure for those both tests. While the results of the test 3 at 0.9 kg/h showed a vertical stratification, in this case the momentum forces are not dominant, i.e. the jet length is smaller than the height of the enclosure, upper than 1.8m the release is a pure plume.

Grand Gamelan [6] facility is a 2-m3 enclosure which internal dimensions are (W0.96xL0.96xH2.1) m3. Two openings are located on the same face of the enclosure one at the top and the other at the bottom. Their dimensions are (H19xW90) cm². The injection point is a circular tube of either 27.2-mm diameter or 4-mm diameter located at 28 cm from the floor. The release was centred on the floor and directed upwards. The flowrate was tested from 5 to 210 Nl/min.

The dimensions of the 1 m3 enclosure [6] are (W0.995xL0.995xH1.0) m3, similar to the GAMELAN facility but equipped with two vents. Two openings are located on the opposite faces of the enclosure one at the top and the other at the bottom. Their dimensions are (H18xW96) cm². The injection point is a circular tube of either 27.2-mm diameter or 4-mm diameter located at 8 cm from the floor. The release was centred on the floor and directed upwards. The flowrate was tested from 10 to 210 Nl/min.

1.2 Description of the existing models of dispersion

Because the enclosure doesn't have an open event in all cases but isn't purely a closed enclosure (there is some distributed porosities), three different models will be used: the model of Linden; the model of Molkov and the closed enclosure model or Model of Cleaver. For both Linden and Molkov models, the vent is installed on a lateral wall on its upper part. Moreover, a pure plume is considered and the hydrogen fraction into the enclosure is considered uniform.

<u>Model of Linden</u> [7]

An opening is located on the high part of an enclosure, h is its height and A its area. If the enclosure is filled with a light gas, the difference of density between the interior and the exterior of the enclosure will lead to the ventilation of the enclosure through the vent. The gaseous release is supposed to be a pure plume. When the steady state is reached, the concentration into the enclosure is given as follows:

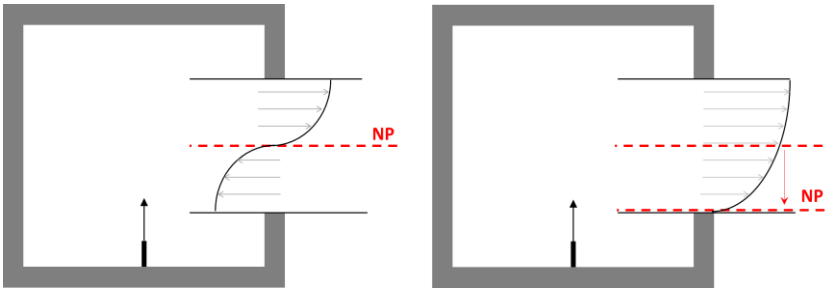
$$X = \left[\frac{Q_0}{C_D A (g' h)^{1/2}}\right]^{2/3}, \quad (1)$$

where Q_0 – injected flow rate, m³/s; A – vent area, m²; h – vent height, m; g' – reduced gravity acceleration defined as $g' = g * (\rho_{air} - \rho_{H_2}) / \rho_{air}$, m/s²; C_D – discharge coefficient, C_D=0.25.

Model of Molkov [8]

Molkov developed a model of ventilation partly based on the model of Linden; called passive ventilation. The two distinctive features are firstly, the use of a more usual coefficient C_D equal to 0.6 (C_D is equal to 0.25 in the Linden Model) and secondly, the introduction of passive ventilation. For the passive ventilation, the concept of the neutral plan is introduced and defined as "the height of vent where the pressure differences across the opening will be zero". Consequently, the gases will flow out above the neutral plan and flow in below it.

For the Molkov model and so a passive ventilation, the neutral plan can be positioned anywhere below the half of the vent height while for the natural ventilation (Linden Model) the neutral plan is always positioned at the half of the vent height, as showed in the figure below. In the equation (2) the first term reflects the modification of the neutral plan position.



a. Linden Model b. Molkov Model

Figure 1. Different positions of the neutral plan NP (in red) for natural ventilation (Linden Model) and passive ventilation (Molkov Model)

In the case of natural ventilation, an assumption is made: the out-coming and incoming volumetric flow rates are equal. Finally, the volume concentration in the steady state is as follows:

$$X = \left(\frac{9}{8}\right)^{1/3} . \left\{\left[1 - X\left(1 - \frac{\rho_{H_2}}{\rho_{air}}\right)\right]^{1/3} + (1 - X)^{2/3}\right\} . \left[\frac{Q_0}{C_D A (g_0' h)^{1/2}}\right]^{2/3}, \quad (2)$$

where Q_0 – injected flow rate, m³/s; A – vent area, m²; h – vent height, m; g' – reduced gravity acceleration, m/s²; C_D – discharge coefficient, C_D=0.6; ρ_{H2} – hydrogen density, kg/m³ and ρ_{air} – ambient air density, kg/m³.

Closed enclosure model or Model of Cleaver [9]

A closed enclosure model is also presented to be as complete as possible and not to ignore a possible way.

$$X_{H2} = \frac{m_{H2} . R . T}{P . V . M_{H2}}, \quad (3)$$

where m_{H2} – injected hydrogen mass in the enclosure, kg; R – ideal gas constant, J/K/mol; T – temperature, K; P – pressure, Pa; V – volume enclosure, m³ and M_{H2} – hydrogen molar mass, kg/mol

To compare with and obtain additional flow information, an experimental parametric study is presented in the follow part of this article about the dynamics of hydrogen releases and dispersion and several parameters are studied.

2.0 EXPERIMENTAL SETUP

The experimental installation (Figure 2) is composed by:

- A 4-m3 enclosure
- A tank of 50 L equipped with two electro-pneumatic valves; one for the gas supply and the other for the purge
- A pipe for the tank purge (internal diameter 8 mm)
- A pipe for the injection of the gas in the 4-m3 enclosure (internal diameter 8 mm)
- An isolation valve to isolate the tank and the enclosure
- A seeding system i.e. before the injection in the enclosure, the hydrogen goes through two sections of plenum equipped with an entrance for the reagents – hydrochloric acid and ammonia – designed to seed the flow
- A leak system in the enclosure
- An electro-pneumatic valve for ventilating the enclosure with compressed air

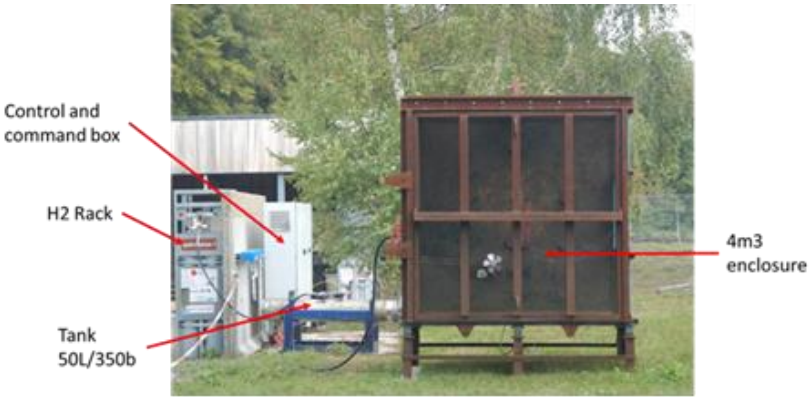


Figure 2. Illustration of the experimental set-up

The mock-up is a 4-m3 enclosure with a 2-m length, a 2-m height and a 1-m width. It is composed by three transparent walls, a vent located on a lateral wall and centred regarding the horizontal axe. The built of the mock-up consist on a machine-welded structure made with 50-mm T-iron and I-iron that support the transparent faces in 2-cm-thickness PMMA. The other walls (not transparent) are 5-mm-thickness plates of steel mechanically reinforced by 5-mm I-iron (Figure 3).

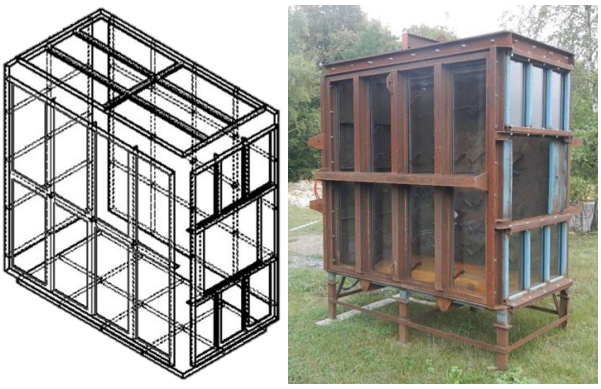


Figure 3. Assembly and picture of the experimental mock-up [10]

The tank (Figure 4) used is a 50-l tank made in 316L stainless steel (adapted to hydrogen) and withstands a pressure up to 350 bars. It's equipped with three electro-pneumatic valves; for the gas supply, for the purge and for the injection in the enclosure. The tank is also equipped with a thermocouple and a pressure sensor.

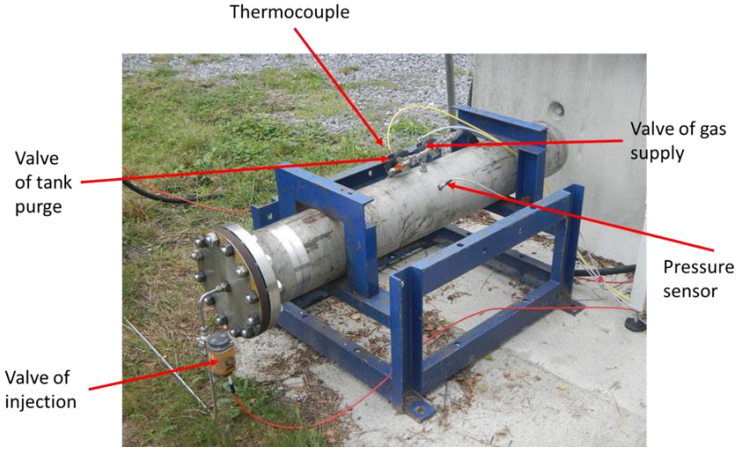


Figure 4. Experimental installation: Tank

2.1 Leakage system

In order to as close as possible to real industrial situations, two types of releases had been selected a circular bore release (Figure 5.a) corresponding to a puncture or a guillotine break and a ring-shaped release (Figure 5.b) corresponding to a leak on a joint for example. To create the ring-shaped release, a micrometric screw thread is welded to a circular flange. This flange blocks a circular release and create a "plane" jet.

a. b.
Figure 5. Example of a jet release and ring-shaped release

2.2 Instrumentation

Measurement of velocity and turbulence: The Pitot sensor linked with a differential pressure transducer (Figure 6.a), allows to measure the difference of the dynamic pressure and then to deduce the velocity of the flow and the fluctuations in the flow.

Measurement of concentration: To characterize the dispersion into the experimental chamber, concentrations measurement (Figure 6.b) is set up and 6 oxygen analysers will sample the atmosphere long the vertical axis each around 35 cm (O1=1.84m; O2=1.5m; O3=1.16m, O4=0.82m; O5=0.47m and O6=0.13m). The objectives of the concentration measurements are to be able to determine the concentration of the cloud generated by a small leak, its height if a gradient can appear inside the cloud.

Visualisation of release shape: The evolution of the release shape is estimated by filming the transparent wall of the enclosure with a camera. In order to see both the development of the release and the dispersion, the mixture injected is seeded/sowed with nanoparticles/micro-particles of ammonium chloride NH_4Cl. The ammonium chloride is a white ionic salt which melts at a

temperature around 340°C. The ammonium chloride particles are created by the interaction of vapors of hydrochloric acid and ammonia through the following reaction: $HCl + NH_3 \rightarrow NH_4Cl$. This technique doesn't modify the dispersion behaviour.

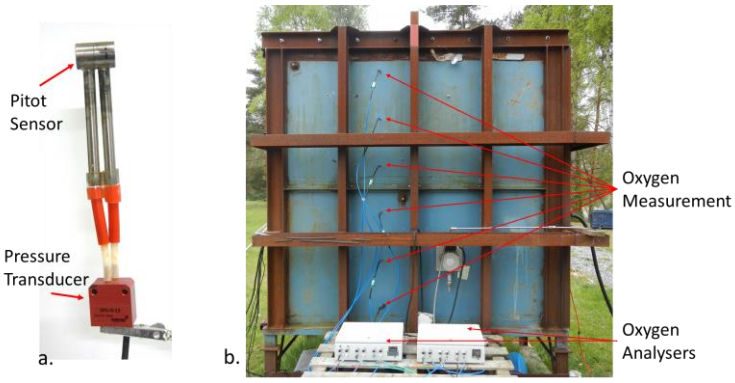


Figure 6. a. Picture of a pitot sensor linked with a pressure transducer; b. Picture of the back of the experimental installation showing the location of the oxygen measurements and the oxygen analysers

Measurement of integral scale: In order to measure the integral scale, the original pitot sensor had been replaced by bended capillaries (Figure 7) connected to the positive port of the differential pressure sensors. Those heads are less invasive than the pitot heads and allows the calculation of the integral length with the spatial correlation. To facilitate the measurements, all the second port of the differential pressure sensor are linked to the atmospheric pressure.

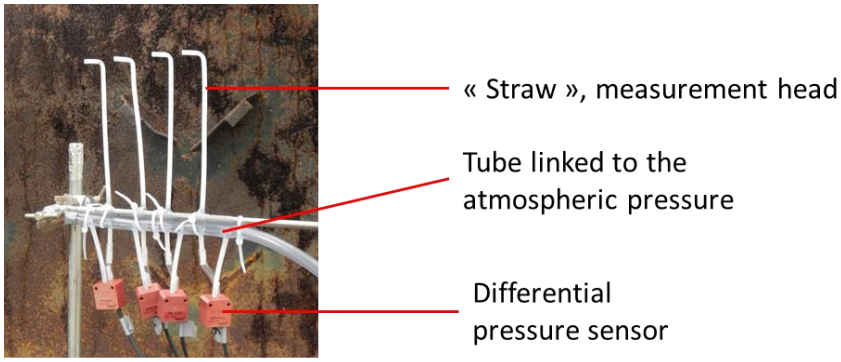


Figure 7. Integral scale sensors

3.0 RESULTS AND DISCUSSION

In function of the type of release the shape of the release is different (Figure 8), in case of a circular release, the jet hits the enclosure in the upper part then the layer created goes down and a uniform and turbulent layer is formed while in the case of a ring-shaped release the jet hits the enclosure in the lower part of the enclosure then the layer goes up in the enclosure and once again a uniform and turbulent layer is formed. Some experimental results are given in Table 1.

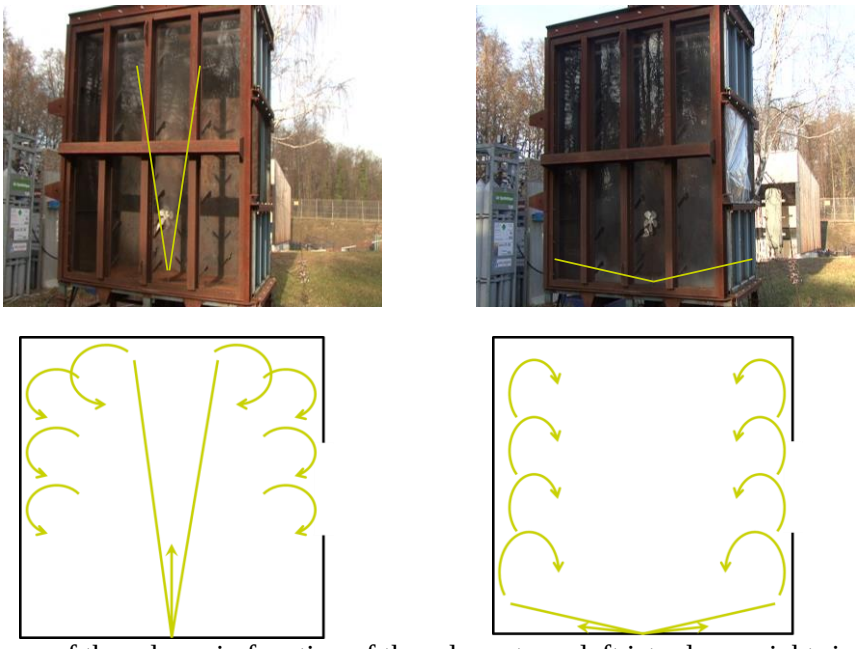


Figure 8. Shape of the release in function of the release type, left jet release, right ring-shaped release

For all the releases studied and whatever the pressures tested; 10, 20 and 40 bars, a uniform concentration is rapidly obtained in the enclosure due to the high release velocity around 1000m/s that implies a high momentum (Figure 9).

The discharge coefficient, calculated for all the experiments, is on average equal to 0.79 and takes also into account the length and the pressure loss of the pipe linking the reservoir to the enclosure.

Table 1. Recapitulation of the experiments.

Test n°	Tank Pressure bar	Leak diameter mm	Release type	Mass flow rate g/s	Volume flow rate Nl/min	H_2 fraction % vol.
32	9.8	1	Circular	0.45	205	9.1%
33	19.7	1	Circular	0.90	393	17.8%
34	40.1	1	Circular	1.74	782	32.6%
35	9.5	3	Circular	3.81	2108	10.0%
41	19.2	3	Circular	7.99	3421	18.8%
37	40.7	3	Circular	13.00	7057	33.6%
26	10.0	3.33	Ring-Shaped	0.87	110	8.4%
24	19.9	3.33	Ring-Shaped	1.14	559	15.4%
25	40.0	3.33	Ring-Shaped	3.85	1771	30.6%

All the tests had been doubled and shown a good reproducibility.

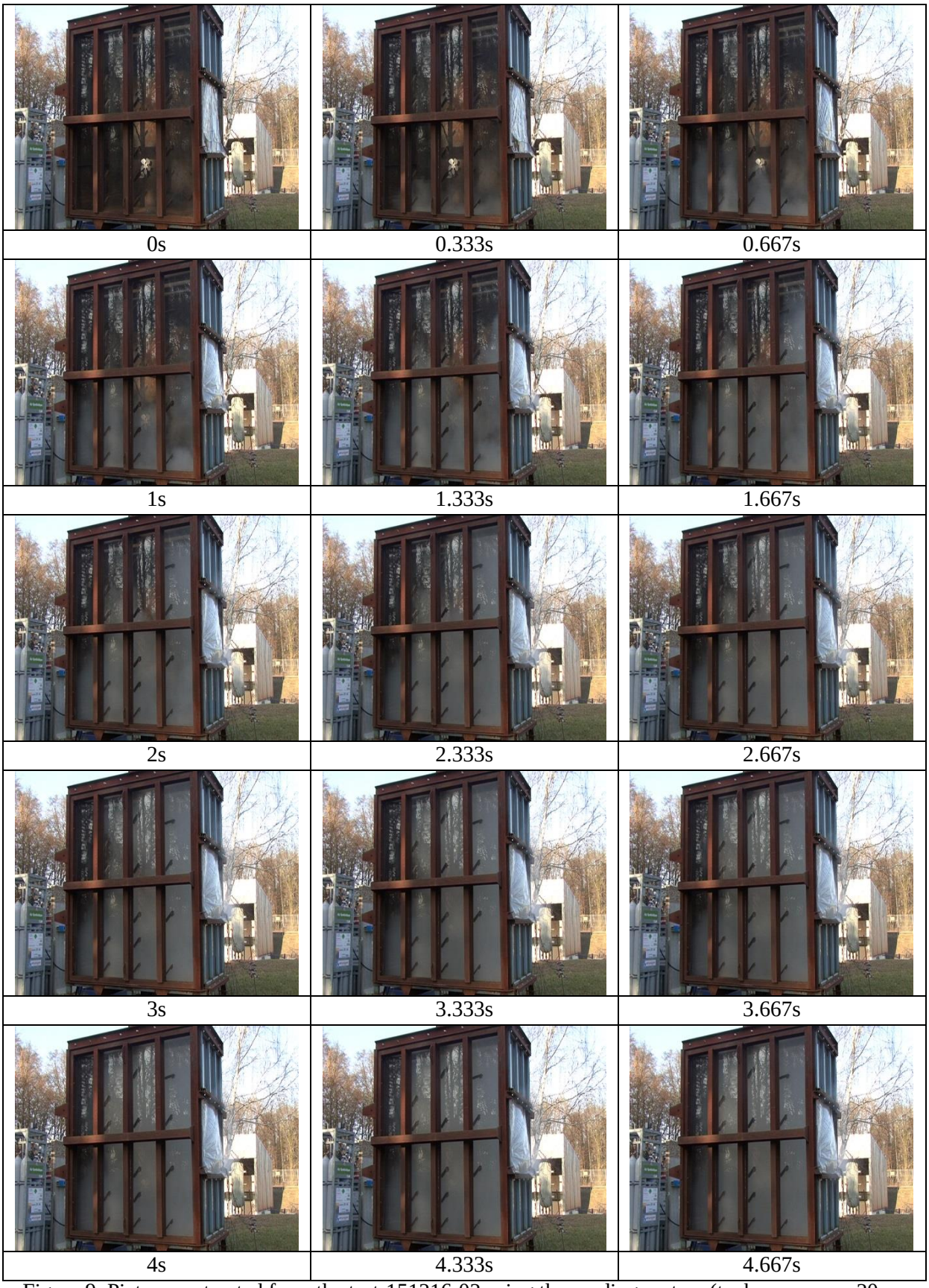


Figure 9. Pictures extracted from the test-151216-02 using the seeding system (tank pressure = 20 bars; ring shaped release)

3.1 Comparison with the existing models

The experimental hydrogen fraction is compared to the following models: Linden and Molkov models considering the presence of a vent and Cleaver model for closed but porous enclosure. An important disparity had been found (Table 2 and Figure 10). Note that the vent was considered fully open for the Linden and Molkov models which is not true. Indeed, the vent was partially covered with a thin plastic film during the experiments. The open area of the vent was around 0.004m². If this vent area is used in the Linden and Molkov models, the concentration calculated is greater than one which is impossible.

The models of Linden and Molkov are available/developed for pure plumes and centered releases on the floor and taken away from the side walls. The plume is also vertical and upwards, fully in line with the present configuration. About the opening the models consider that the vent is always on the high part of the wall. Those models are based on a mass balance between the inlets (release) and the outlets (vent).

The main difference between the models of Linden and of Molkov is the position of the neutral plan at the level of the vent. The model of Molkov takes into consideration the impact of the release flow rate on the position of this neutral plan; indeed, when the flowrate increases, the neutral plan tends to go down consequently through the vent, outlet flowrate is majority.

Those models predict the concentration at equilibrium at the level of the vent. This concentration corresponds to the maximal concentration into the enclosure. That's why users had to be careful if a stratified regime could occur.

Most of the release operated during the experiments presented above are centered on the floor, taken away from the walls and upwards. None of the presented models consider the volume of or the presence of congestion into the enclosure. Plus, into those models the layer is often considered uniform, the case of gradient is only approached by Worster and Huppert [11], available for closed enclosure only.

For all the models the maximal concentration was calculated using the initial flow rate which is also the maximum flow rate.

None of those models seem to be satisfactory.

Table 2. Comparison between the experiments and models.

Test n°	**H_2 fraction % vol. exp. final**	**% vol. Cleaver max**	**% vol. Linden max**	**% vol. Molkov max**
32	9.1%	12.0%	5.7%	6.4%
33	17.8%	24.0%	9.1%	10.0%
34	32.6%	48.2%	14.1%	15.1%
35	10.0%	11.7%	23.8%	24.2%
41	18.8%	23.4%	38.9%	36.6%
37	33.6%	48.9%	53.9%	46.8%
26	8.4%	12.2%	8.9%	9.8%
24	15.4%	24.2%	10.6%	11.6%
25	30.6%	48.0%	23.9%	24.4%

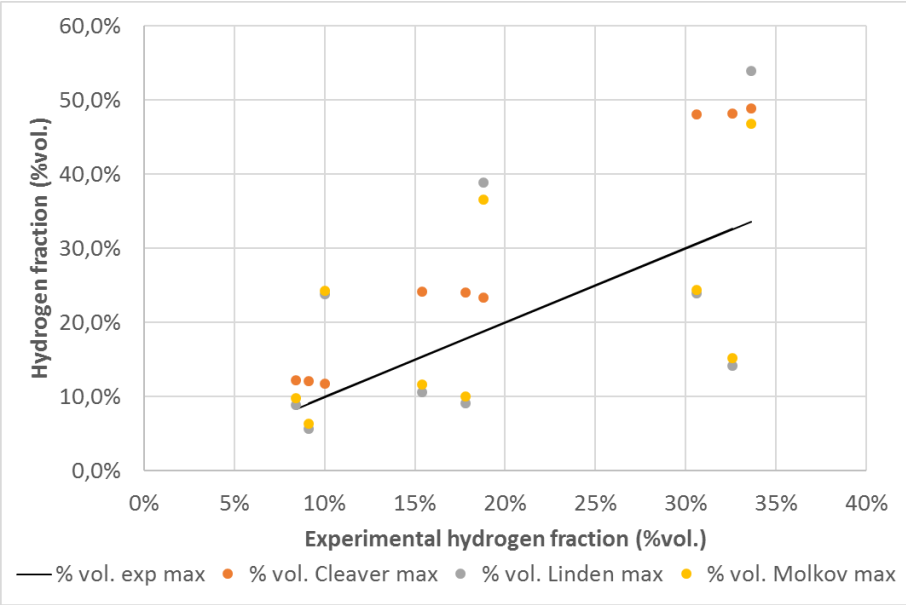


Figure 10. Comparison between the experimental hydrogen fraction and the existing models

3.2 Turbulence

Even if the release velocity is high. the jet velocity decreases rapidly and outside the jet the average velocity in the enclosure is only around 0.1 m/s while the turbulent intensity is from 1 m/s to around 7 m/s (Table 3). For all release diameters, the turbulent intensities u' increase with the tank pressure.

Table 3. Results of turbulent intensity u'.

Test n°	Tank Pressure bar	Leak diameter mm	Mass flow rate g/s	u' m/s
32	9.8	1	0.45	1.9
33	19.7	1	0.90	2.7
34	40.1	1	1.74	3.6
35	9.5	3	3.81	4.1
41	19.2	3	7.99	5.4
37	40.7	3	13.00	6.9
26	10.0	3.33	0.87	1.4
24	19.9	3.33	1.14	1.3
25	40.0	3.33	3.85	2.7

Figure 11 shows that the turbulent intensity in function of the time decreases with the same shape than the tank pressure drop and go back quickly around zero.

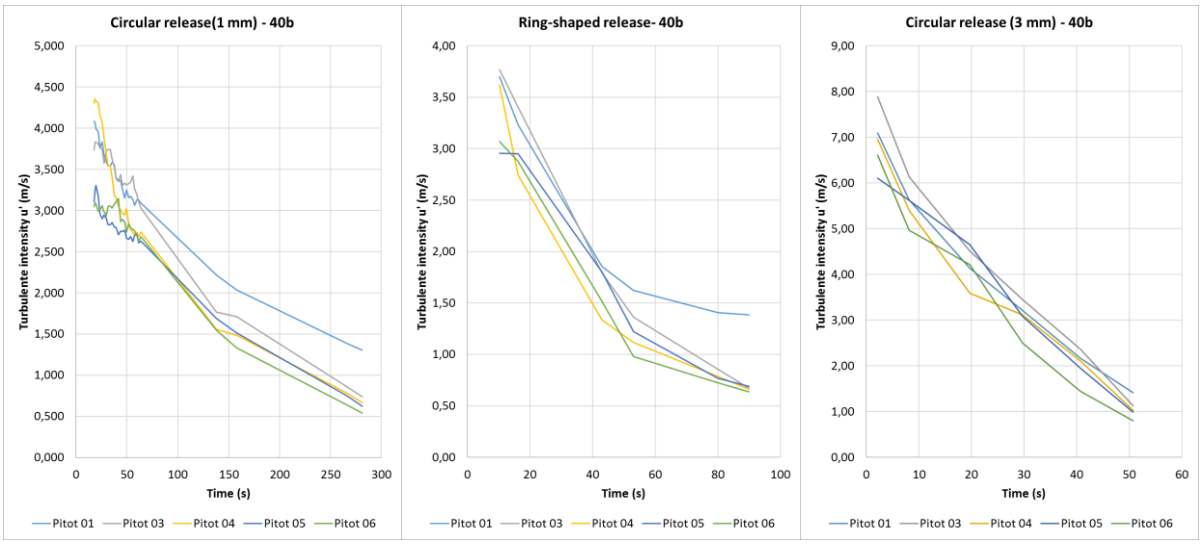


Figure 11. Turbulent intensity in function of the time for three tests: circular release Ø = 1 mm; ring shaped release Ø = 3.33 mm and circular release Ø = 3 mm at P = 40 bars

During our tests. we found that the Taylor "frozen turbulence" hypothesis was invalid. Indeed. the mean average into the enclosure is smaller (around 0.1 m/s) than the turbulent intensity which is the order of magnitude of several meters per second. We needed to change the instrumentation in order to be able to calculate the integral scale with a spatial correlation (Figure 7). The integral scale found is shown in Table 4.

Table 4. Results of the integral scale measurements

Test n°	Tank Pressure bar	Leak diameter mm	Release type	Lt cm
49	10.3	1	Circular	5.0
50	38.8	1	Circular	6.9
47	9.1	3	Circular	4.8
48	36.5	3	Circular	7.1
53	10.0	3.33	Ring-Shaped	3.0
54	40.2	3.33	Ring-Shaped	3.2

The integral scale seems to be more dependent of the type of release and of the tank pressure than of the release diameter. The results are of the same magnitude order than the Hinze [12] approximation (i.e. $L_T = 0.05\ (V_{enclosure})^{1/3}$) which is equal to 7.9 cm for the 4m^3 enclosure.

4.0 CONCLUSION

To define a strategy of mitigation for containerized hydrogen systems (fuel cells for example) against explosion. the main characteristics of a flammable atmosphere (size. concentration. turbulence…) shall be well-known.

This article presents an experimental study designed to understand and quantify the mechanisms of accidental hydrogen release and dispersion into a 4 m3 enclosure (2 m x 2 m x 1 m).

Hydrogen releases of 1mm and 3 mm diameter and ring-shaped of equivalent 3.33mm diameter orifices had been investigated at different positions inside the enclosure. A specific effort is made to characterize the turbulence in the enclosure during the releases. The results shown that even for the smallest mass flow rates (around 0.1 g/s) a uniform and turbulent atmosphere is formed in the enclosure with concentrations higher than the lower flammable limit. Such a mass flow rate represents a seaming default at (10b and an area of 0.2mm²) or (40b and an area of 0.05mm²).

The hydrogen dispersion is a vast topic and further experiments are needed in order to understand all the phenomenon imply during a release following by a dispersion.

The experimental data is analyzed and compared with existing engineering models. None of the presented models gave satisfactory results. An extra work is ongoing in order to consider all the situations studied in the paper and also releases near the ceiling or side walls that will study later and could lead to a stratification of the explosive atmosphere.

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