HYDROGEN RELEASE AND ATMOSPHERIC DISPERSION: EXPERIMENTAL STUDIES AND COMPARISON WITH PARAMETRIC SIMULATIONS

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ABSTRACT

In our society the use of hydrogen is continually growing and there will be a widespread installation of plants with high capacity storages in our towns as automotive refueling stations. For this reason, it is necessary to make accurate studies on the safety of these kinds of plants to protect our town inhabitants Moreover, hydrogen is a highly flammable chemical that can be particularly dangerous in case of release since its mixing with air in the presence of an ignition source, could lead to fires or explosions. Generally most simulation models, whether or not concerned with fluid dynamics, used in safety and risk studies are not validated for hydrogen use. This aspect may imply that the results of studies on safety cannot be too accurate and realistic. This paper introduces an experimental activity which was performed by the Department of Energetics of Politecnico of Torino with the collaboration of the University of Pisa. Accidental hydrogen release and dispersion were studied in order to acquire a set of experimental data to validate simulation models for such studies. At the laboratories of the Department of Mechanical, Nuclear and Production Engineering of the University of Pisa a pilot plant called Hydrogen Pipe Break Test was built. The apparatus consisted of a 12 m^3 tank which was fed by high pressure cylinders. A 50 m long pipe moved from the tank to an open space and at the far end of the pipe there was an automatic release system that could be operated by remote control. During the experimental activity, data was acquired regarding hydrogen concentration as a function of distance from the release hole, also lengthwise and vertically. In this paper some of the experimental data acquired during the activity have been compared with the integral models, Effects and Phast. In the future, experimental results will be used to calibrate a more sophisticated model to atmospheric dispersion studies.

1.0 INTRODUCTION

The growing use of hydrogen in our society requires a scientific and suitable basis for the evaluation of credible safety issues. This aspect will become progressively very important because of the widespread installation of plants with high capacity storages in our towns as automotive refueling stations. Moreover as we all know, hydrogen is a highly flammable chemical and in case of fire or explosion the consequences can become serious under certain conditions. Risk and safety analyses can be used to evaluate and investigate hazards of a hydrogen plant. In these similar studies the focus is the scenario of accidental release. After release, in the presence of an ignition source, a jet fire could be verified from the leak until the supply is controlled or exhausted, otherwise the gas will evolve in a flammable cloud and in case of ignition a flash fire or an explosion could result. For these reasons, in case of immediate ignition of the jet, it is important to know the flame length and thermal radiation heat flux distribution, whereas in case of delayed ignition a very important and fundamental issue is the study of the spatial behavior of the hydrogen jet concentration in the surrounding air and the determination of locations where the concentration falls below the lower flammability limit. Generally, accidental releases can originate from small and large holes in pipes, from high-pressure storage tanks or from flanges and gaskets of components, like compressors, electrolysis systems, etc.

In safety studies about hydrogen systems, the scientific community has identified the characterization of unintended release and subsequent dispersion as the most important problem. In fact all over the world many researchers are investigating on possible hydrogen accidents, especially on atmospheric dispersion phenomena, in order to determine protection measures from hydrogen fires and explosions. Unfortunately most simulation models, whether or not concerned with fluid dynamics used in studies on safety and risk are not validated for hydrogen use. This aspect may imply that results of safety studies cannot be too accurate and realistic. In fact the current trend in the research studies is to test several CFD models for various types of hydrogen releases [1-7]. Detailed and reliable experimental database to validate these models are increasingly necessary.

In this paper some experimental measurements are reported. The Department of Energetics of the Politecnico of Torino has performed a wide safety research programs on the employment of hydrogen in refueling stations. Accidental hydrogen release and dispersion phenomena have also been studied with the collaboration of the University of Pisa in order to acquire a set of experimental data to validate simulation models for such studies. Two objectives are achieved in this paper: the experimental characterization of hydrogen release from a low pressure system and a preliminary comparison of experimental data with two integral models in order to examine their capacity to handle hydrogen release. At the laboratories of the Department of Mechanical, Nuclear and Production Engineering (DIMNP) of the University of Pisa a pilot plant called Hydrogen Pipe Break Test (HPBT) was built. The apparatus consisted of a 12 m^3 tank which was fed by high pressure cylinders. The maximum internal pressure was 1 MPa. A 50 m long pipe moved from the tank to an open space and the far end of the pipe had an automatic release system that could be operated by remote control. During the experimental activity, data was acquired regarding the hydrogen concentration as a function of distance from the release hole, also lengthwise and vertically in order to determine the extent of the flammable cloud generated. Meteorological data was also acquired continuously by means of an anemometer localized near the source of release. In this paper some of the experimental data acquired during the activity are compared with the integral simulation models, Effects and Phast, to verify the behavior of the two models as regards hydrogen release and dispersion simulations.

In the future, experimental results will be used to calibrate more sophisticated models for atmospheric dispersion studies, such as a Lagrangian particle model for hydrogen atmospheric dispersion.

2.0 EXPERIMETAL APPARATUS

The experimental apparatus HPBT was installed within the Laboratory “Scalbatraio” of the University of Pisa. This apparatus was used to investigate the behavior of hydrogen leakages from pipelines; it was able to simulate a real, low pressure hydrogen release into free air. The purpose of the University of Pisa’s activity was to generate data that could help develop an Italian regulation for the transport of hydrogen through pipelines. Unfortunately the apparatus was not designed to investigate on high pressure and high capacity storage systems, which falls under the scope of the Politecnico of Torino unit. For this reason the plant was utilized by the Torino unit only in its most dangerous configuration. The pressure system was designed to have a maximum working pressure of 1 MPa. Discharge orifices of varying diameters and discharge pressure were changed to study different accidental conditions. The supply system used (four storage tanks of 3 m^3 each) ,with the largest orifice (0,011 m) allowed the maximum discharge pressure to be maintained only for about one minute before the pressure began to drop below 0.7 MPa. Below this value the recharge of experimental apparatus became too expensive and the jet length too small for the scope of the research. All the releases were directed horizontally, 0.9 m above the test ground. The experimental activity also addressed the acquisition of data that could be useful to validate computational codes like CFD models. The Torino unit would have needed a higher pressure and capacity, but a bigger economic investment was not available. Despite these problems, results of the Torino unit were very reasonable.

During the experimental series a total of 22 tests were performed. The conditions that changed during the tests were: hole diameter (0.0025 m; 0.005 m; 0.011 m) and internal pressure (0.2 MPa; 0.5 MPa; 1 MPa). Only two of all tests performed are reported and analyzed in this paper, both performed with D=0,011 m and P=1 MPa [8].

2.1 Experimental apparatus layout

The layout of the HPBT apparatus can be divided into four ideal parts [9]:

1. Hydrogen and nitrogen storage. There were two gas boxes: the first housing the hydrogen banks; the second containing the nitrogen banks used for cleaning to remove air inside the apparatus and to remove the residual hydrogen at the end of experiments. Each bank consisted of 25 cylinders with an initial pressure of 20 MPa;

2. Gas reservoir (test pressure). Composed of four storage tanks 3 m^3 each with a maximum working pressure of 1 MPa; making it possible to store up to 130 Nm^3 of hydrogen. The reservoir was connected to the banks by a pipe of 2 in (0.0508 m) internal diameter. The reservoir delivered hydrogen to the pipeline system by a discharge manifold. Furthermore it was also directly connected to an emergency vent line;

3. Pipeline system. A pipe of 4 in (0.102 m) internal diameter and 50 m long leading from the gas reservoir to an automatic release system (ARS) where the hydrogen leakage took place in an open field. When the ARS was turned on, it opened in about ten seconds and could be closed on command. The ARS consisted of two different valves in series: the first was a pneumatic ball valve which opened in few seconds, the second was a pneumatic fast opening butterfly valve. The line length allowed the simulation of a real pipeline and also guaranteed a safety distance between the gas storage and the release point. The far end of the pipe was connected to the vent line by a pipe of 2 in (0.0508 m) in internal diameter, to allow the removal of hydrogen when necessary;

4. Vent line. A 6 m high pipe of 2 in (0.0508 m) internal diameter that was able to vent the gas when necessary. It was a system used in the event of a malfunctioning to remove residual hydrogen from the tanks, but it was also used when compressed nitrogen was fed to the apparatus in order to leave inert gas inside the plant.

3.0 DATA ACQUISITION SYSTEM

In this paragraph the acquisition system set only for the two tests here reported, is described [9]. During the tests the following data were acquired: oxygen concentration, internal pressure, internal temperature and wind intensity and direction. The pressure and the temperature of the hydrogen close to the release nozzle and in the storage tanks were recorded during each test in order to control the release. The air temperature, wind intensity and direction were measured continuously near the release point using an anemometer and a thermocouple.

3.1 Anemometer

Wind was monitored continuously at about 0.9 m above the ground and near the source of the release. It was far from obstacles that could create turbulence. The instrument used was an anemometer MODEL N°1086 LTD by Gill Instruments Ltd (Lymington Hampshire – England).

3.2 Thermocouples and pressure transducers

Temperature and pressure were measured in three different points: inside tank 1, inside tank 4 and next to the release nozzle. The instruments used were commercial thermocouples type “K” and Druck pressure sensor type PTX1400.

3.3 Concentration acquisition system

Unfortunately hydrogen sensors available for the tests do not work properly in free air when analyzing range concentration between 0% and 100% in volume. Therefore in order to have data on hydrogen concentration in free air, oxygen concentration was acquired in eight different points. The data on the concentration of hydrogen was obtained by measuring the oxygen concentration assuming that any decrease in the concentration of oxygen was caused by displacement of oxygen by hydrogen gas. The sensors used were SMART3 CC-CD (NET/x) by SENSITRON S.r.l. (Milano – Italy). In order to connect the points where the samples were placed to the sensors, eight rylsan pipes (6x4mm) were used. A vacuum pump model TIPO BS V3 was used to suck the samples in the sensors. The flow rate was regulated through each pipe by asameters model TECMA FLUSSIMETRO SERIE 1900.

Test points were chosen both in planar and spatial configurations in order to study jet shapes and wind influence.

4.0 EXPERIMENTAL RESULTS

Only the results of Test 2 and Test 3 are reported in this paper:. They were chosen in order to consider the most critical release conditions realized during the experimental studies and to make the comparison with the two integral Effect and Phast models.

4.1 Meteorological data

The acquisition of data by the anemometer was started at the beginning of the experimental day. In this way a continuous measure of the three components of the wind was available. The anemometer was oriented northward by means of a compass. The instrument was positioned near the source of release at altitude of 0.90 m. For each test the acquired data of the anemometer was averaged on three temporal intervals (see Fig. 1):

- T1= Includes the duration time of the test and two minutes before and after the test. It was selected in order to have a best statistic average of meteorological wind status during the test;

- T2= Includes the duration time of the test and a brief period before (about 200 seconds);

- T3= Includes only the duration time of the effective release (about 70-80 seconds).

The meteorological data was evaluated at the previous three different temporal intervals in order to support a possible simulation activity where an estimation of the error in wind data (velocity and direction) would be considered. The T1 interval is advisable because it describes a larger average than the others and considers a possible synchronism error between wind and concentrations logging.

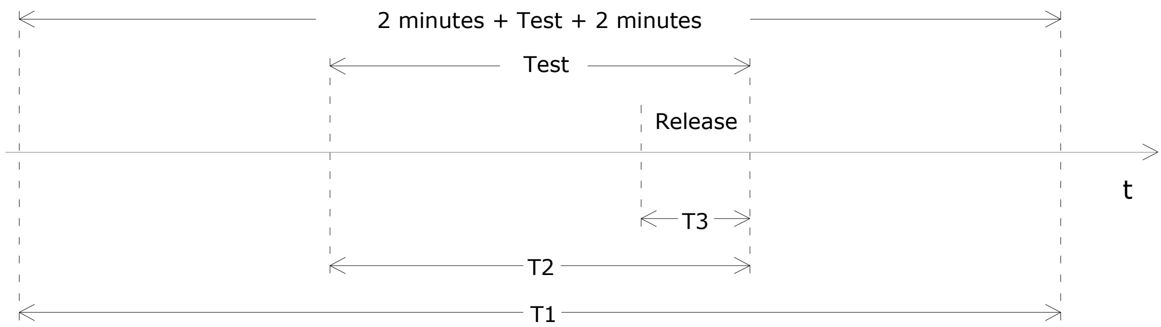
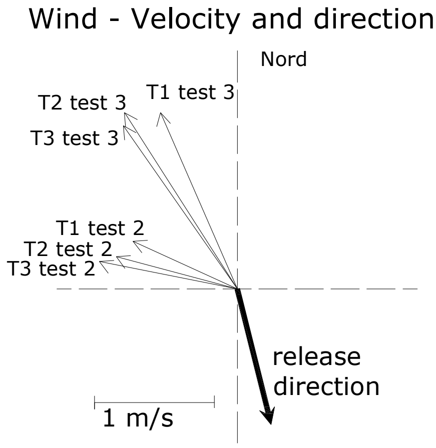


Figure 1. Temporal evaluation intervals of meteorological data

As shown in Fig 2, during Tests 2 and 3 a wind with opposing direction and intensity from 1 m/s to 1,5 m/s was verified. The results of the three temporal intervals allow to quantify an error in the direction of about 12° and on intensity of about 0.2 m/s. By acquired data it was also possible determine some turbulent parameters, such as standard deviations for the three wind components.



Test	Intensity [m/s]	Direction from Nord clockwise
T1 test2	0.96	294°
T2 test2	1.04	285°
T3 test2	1.18	281°
T1 test3	1.61	337°
T2 test3	1.75	328°
T3 test3	1.67	325°

Figure 2. Wind direction and intensity for each temporal interval

4.2 Hydrogen concentration data

Experimental tests were performed with release pressure of 1 MPa and from 0.011 m hole diameter; the direction was approximately horizontal because of a slight upward inclination which was observed to be about 4°, due to an erroneous installation. The release of each test lasted about 70-80 seconds. The following reference system (right-handed Cartesian system) should be considered: the center of coordinates is the release point; the X axis represents the horizontal direction following the release; the Y axis is the horizontal cross-direction; and the Z axis is the vertical direction. The position of the samplers during Tests 2 and 3 are reported in Tab 1.

As mentioned before, during the tests oxygen volume concentrations were acquired and opportunely converted into hydrogen volume concentrations considering an average oxygen volume concentration in the atmosphere of 20.6%. Consequently in the following figures negative hydrogen volume concentrations are relative to an oxygen concentration higher than the atmospheric average. The concentrations captured foresaw three different periods, as described below [10]:

- First period: about 120 seconds of data captured before the release. During this period the samplers recorded the hydrogen (in reality, oxygen) concentration in the atmosphere. As shown in Fig 3 and 5 (there is the same result for each test), the hydrogen concentration was not zero, but varied from -5% to 5%. This result allows evaluation of the calibration error of each sampler and the ground noise due to acquisition method. At the end of this phase the release started.

- Second period: about 50-60 seconds after the first period. During this phase the samplers started to capture the hydrogen jet. The data acquired contains a first period of about 15 seconds of strong instability due to internal dynamics of the samplers as a consequence of the jet impact. After a second period of about 35-45 seconds, the acquisition system slowly achieved a stationary status.

- Third period: about 20 seconds after the second period. During this phase most samplers achieved the stationary status. At the end of this period the release ended because internal pressure in tanks dropped down 0.7 MPa.

In Fig 3, 4, 5 and 6 the experimental measurements of hydrogen concentrations for Tests 2 and 3 are reported.

The results of measured concentrations are summarized and elaborated in Figs 7 and 8. These figures provide an immediate visualization of the hydrogen jet shapes in all the directions: for each test there is one graph on XY plane and another one on the XZ plane. The numbers reported in the graphs represent the distances in centimeter from the source or the horizontal axis, while the blue bar charts (on the left) are representative of the hydrogen volume concentrations measured in the last 20 seconds of the acquisition and red bar charts (on the right) are representative of the 10% error in volume concentration. The error was evaluated considering all sources of uncertainty: instruments and procedures. The numerical value of the concentrations is reported in Tab. 1.

The following figures necessarily point out that, after the analysis of all the tests achieved during the same day, the sampler X10 systematically overestimated the hydrogen concentration. Due to this, its measurements are not to be taken into account.

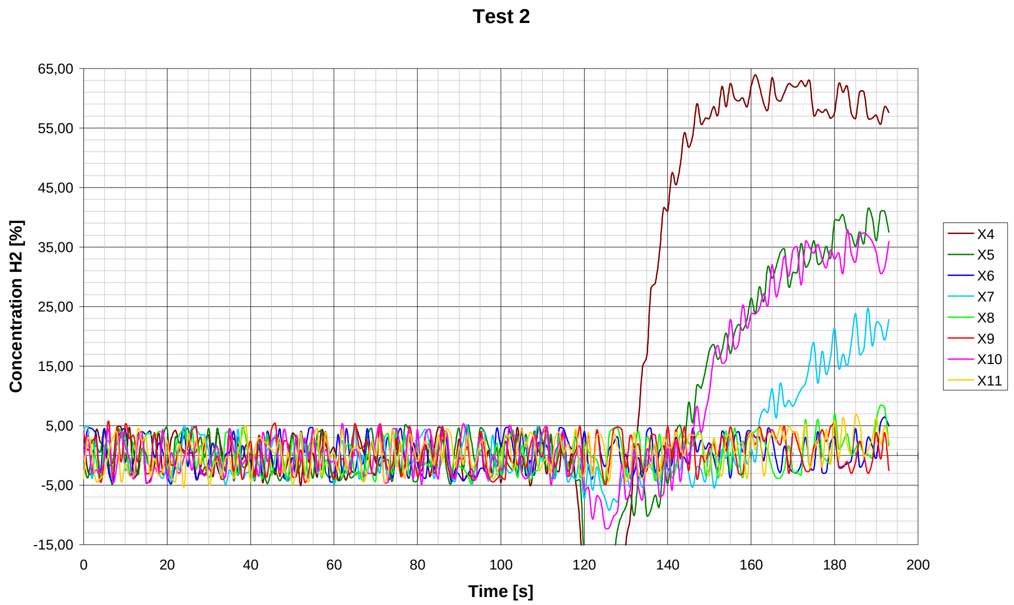


Figure 3. Hydrogen concentration in Test 2

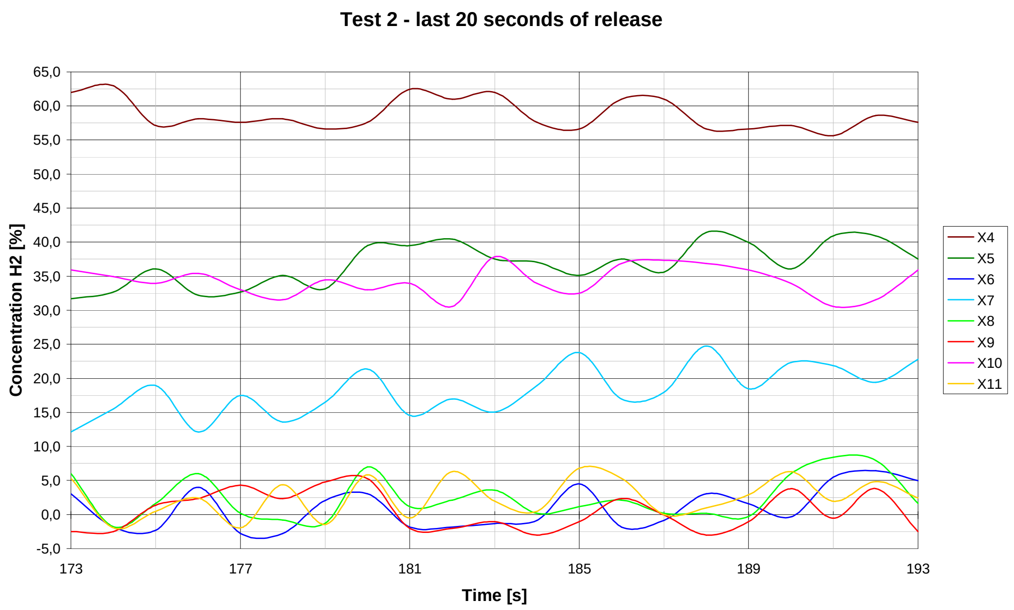


Figure 4. Hydrogen concentration in the last 20 seconds of Test 2

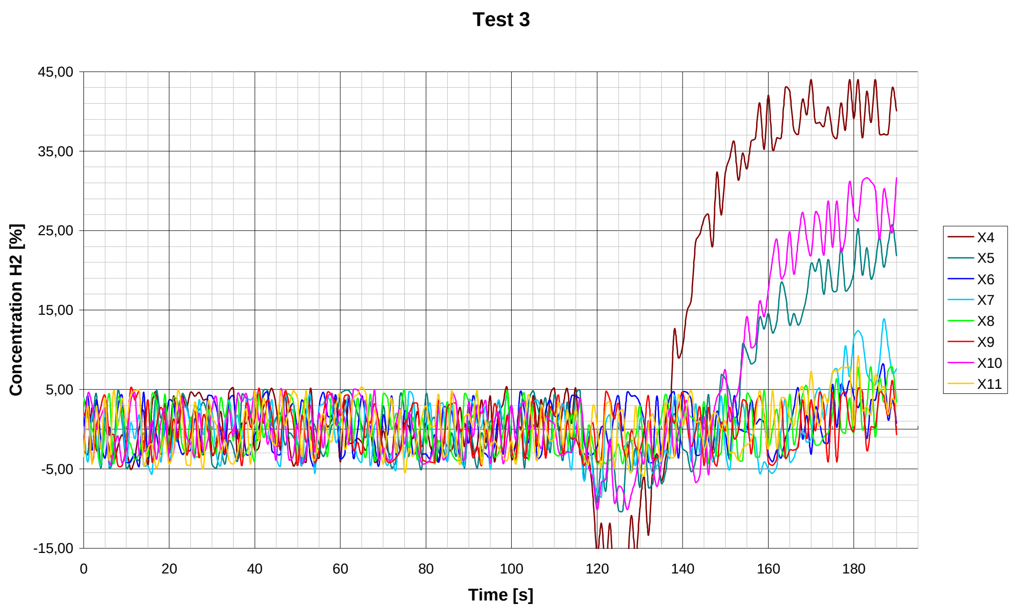


Figure 5. Hydrogen concentration in Test 3

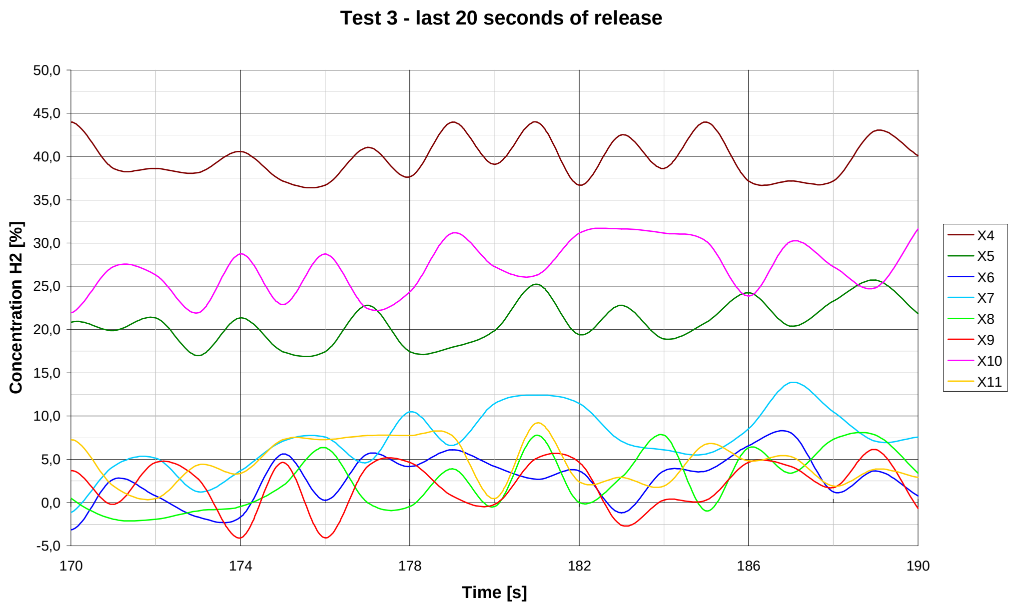


Figure 6. Hydrogen concentration in the last 20 seconds of Test 3

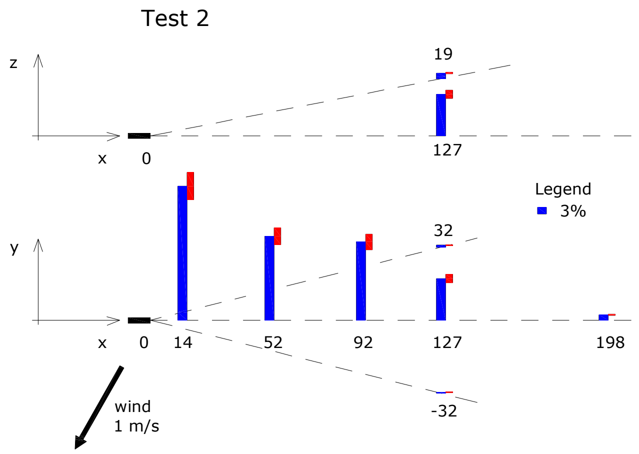


Figure 7. Hydrogen jet shape in Test 2 (X10 readings are erroneous)

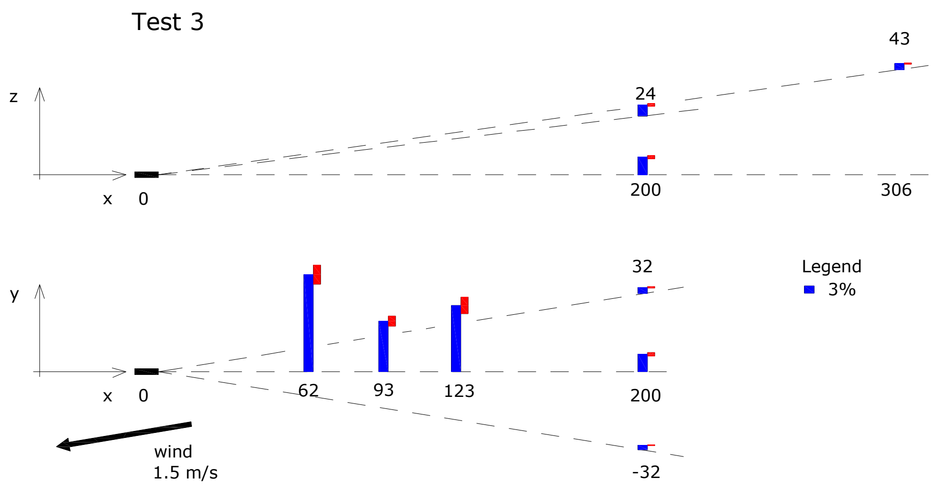


Figure 8. Hydrogen jet shape in Test 3 (X10 readings are erroneous)

Table 1. Position of samplers and mean hydrogen concentration in Tests 2 and 3.

	Test 2		**Test 3**	
Sampler	**Position [cm,cm,cm]**	**Mean concentration [%]**	**Position [cm,cm,cm]**	**Mean concentration [%]**
X4	(14,0,0)	58.8	(62,0,0)	39.8
X5	(52,0,0)	36.8	(93,0,0)	20.8
X6	(127,32,0)	0.9	(200,32,0)	2.6
X7	(127,0,0)	18.2	(200,0,0)	7.2
X8	(198,0,0)	2.4	(306,5,43)	2.5
X9	(127,-32,0)	0.4	(200,-32,0)	19
X10	(92,0,0)	34.3	(123,0,0)	27.2
X11	(127,0,19)	2.5	(200,0,24)	4.6

5.0 COMPARISON WITH INTERGRAL MODELS

The comparison of the experimental measurements in Tests 2 and 3 and the integral models Effects 4.0 and Phast 6.3 is proposed in this paper. Currently these models are the most used in industrial risk analysis. Effects was developed by the TNO Institute of Netherlands whereas Phast was developed by the DNV of Norway. Both models provide integral models to study accidental sequences from the moment the substance is released to the explosion phenomena and/or toxic dispersion, considering different types of chemicals. The scope of the comparison is to verify the behavior of the two models as regards hydrogen release and dispersion simulations. A considerable overestimation of the concentration trend versus the distance is expected [10]. Integral models cannot account for release direction not aligned with wind direction. In this case the use of a CFD is adviced. However in this paper, a first exam of the tests results are reported.

5.1 Phast 6.3 [11]

Phast is a software that collects different integral models that allow the study of an accidental sequence, from the release to the explosion and/or toxic dispersion of a chemical. As to its use in risk analysis, an important quality of Phast is the low setting and calculation times that are very brief and compatible with the requirements for studies on risk and safety. Regarding its use in this particular hydrogen study, a limitation of the model was that it was not possible to define wind direction of the wind respect to the release direction, since both wind and release must have the same direction. For a comparison with the experimental tests it would be important to have the possibility of considering wind direction and intensity. To remedy this problem, the lowest intensity wind was defined in Phast in order to minimize the effect of the wind on the dispersion. Two stability atmospheric classes were also considered: the neutral class D and the very stable class F. Another limit of the model was the setting of the outlet's hydrogen velocity, because there was a higher limit of 500 m/s. Instead in the tests' release conditions, assuming the mass and momentum conservation [12], the velocity after the complete expansion in the atmosphere resulted to 1848 m/s, whereas in correspondence the outlet section was about 1300 m/s (sonic velocity for the hydrogen at atmospheric temperature).

In order to consider the pressure variation during the release, two different simulations were set: the first considered the mass flow at the initial conditions, the second the final conditions of the real release (see Tab. 2).

Table 2. Initial release condition in Phast

Data	**Mass flow [kg/s]**
Pressure of 10 bara	0.059
Temperature of 25 °C	
Hole diameter of 11 mm	
Pressure of 7 bara	0.041
Temperature of 25 °C	
Hole diameter of 11 mm	

Figs 9 and 10 show the results that were obtained using the Phast UDM dispersion model.

In Tabs 3 and 4 there is the comparison between the volume concentrations in the experiments and by Phast.

The comparison shows that:

- At a distance of 14 cm from the source the concentration is overestimated by about 10%;
- Between 52 cm and 62 cm from the source, the concentrations are very similar;
- At a distance of 92-93 cm the concentration is overestimated by about 10%;
- At a distance of 123-127 cm the concentration is overestimated by about 8%;

- At a distance of 198-200 cm the concentration is overestimated above at 10%; it was considered a unrealistic estimation;

- As regards the samplers positioned above the jet axis, the concentrations are all exceedingly overestimated.

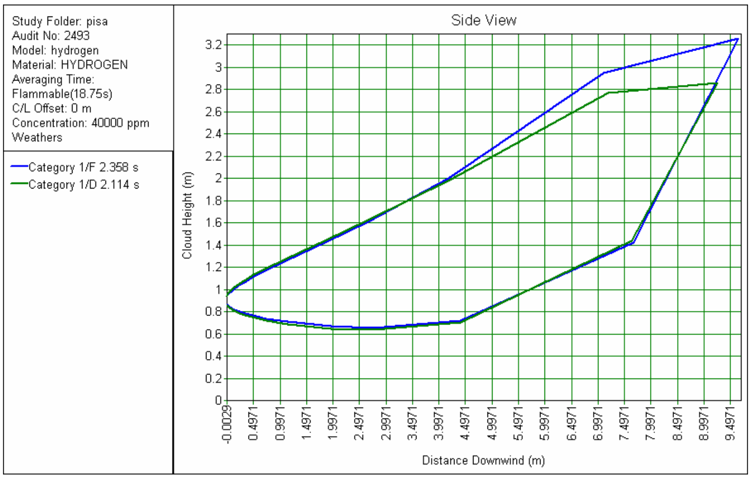


Figure 9 Hydrogen jet shape relative to a volume concentration of 4%

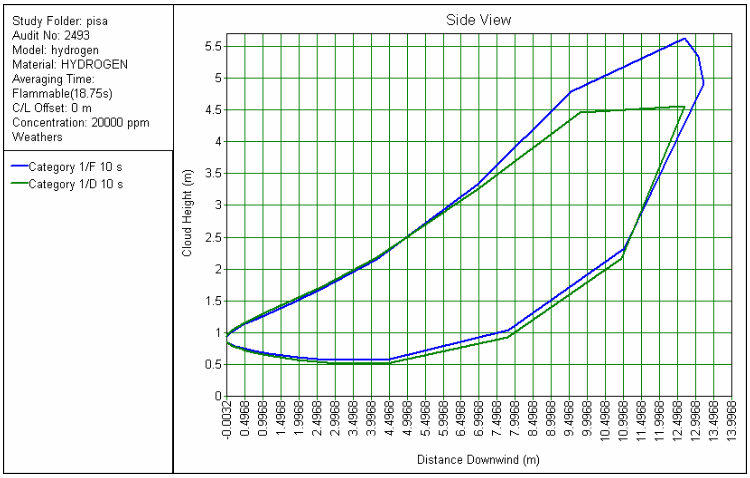


Figure 10. Hydrogen jet shape relative to a volume concentration of 2%

Table 3. Comparison between experimental measurements in test 2 and Phast

Test 2		**Concentration in [% volume] by Phast 6.3**				**Error [%]**
Sampler and coordinates	**Measured concentration**	**Mass flow of 0.059 kg/s**		**Mass flow of 0.041 kg/s**		
[cm,cm,cm]	**[%]**	**D**	**F**	**D**	**F**	**Min/max**
X4 (14,0,0)	58.8	71.4	72.4	68.1	69.3	+14/+19
X5 (52,0,0)	36.8	44.3	46.1	40.3	42.2	+9/+20
X7 (127,0,0)	18.2	25.9	27.6	22.4	24.0	+19/+34
X8 (198,0,0)	2.4	17.7	19.1	15,.39	16.5	+84/+87
X11 (127,0,19)	2.5	25.5	27.4	22.0	23.7	+89/+91

Table 4. Comparison between experimental measurements in Test 3 and Phast

Test 3		Concentration in [% volume] by Phast 6.3				Error [%]
Sampler and coordinates	**Measured concentration**	**Mass flow of 0.059 kg/s**		**Mass flow of 0.041 kg/s**		
[cm,cm,cm]	**[%]**	**D**	**F**	**D**	**F**	**Min/max**
X4 (62,0,0)	39.8	40.6	42.5	37.0	38.9	-8/+6
X5 (93,0,0)	20.8	32.1	34.0	28.1	29.9	+26/+39
X7 (200,0,0)	7.2	17.5	18.9	15.1	16.3	+52/+62
X8 (306,5,43)	2.5	14.3	15.4	12.6	13.6	+80/+84
X11 (200,0,24)	4,6	20.8	21.8	18.2	19.2	+75/+79

5.2 Effects 4.0 [13]

Effects is also a software that collects different integral models. In Effects 4.0 there are two models to simulate atmospheric dispersion: the neutral gas model and the turbulent free jet model (TFJ). The second was chosen because the hydrogen release is at high velocity. Fig 11 shows the results for 0.041 kg/s of mass flow rate; with a higher rate the result will be more critical. A concentration of 8% was obtained at centreline and distance from the source of 5 m. This result is extensively higher than the experimental measures.

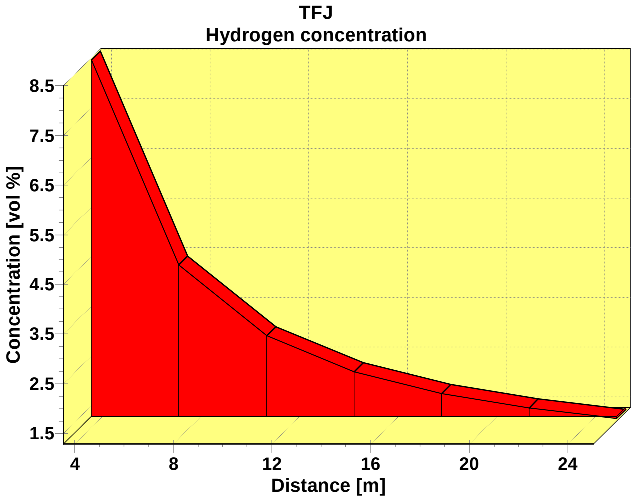


Figure 11. Hydrogen concentration with TFJ model in Effects with mass flow rate of 0.041 kg/s

6.0 CONCLUSION

The experimental experience in Pisa allowed the compilation of sets of experimental data about hydrogen release and atmospheric dispersion. The results also improve knowledge on the behavior of hydrogen jets in the atmosphere after an accidental release and contribute to making considerations on safety distances. Despite the small release pressure and storage capacity and the unfavorable meteorological conditions, the trend of hydrogen concentration measured during the tests were very realistic. The results also show an evident correlation between wind direction and intensity and hydrogen concentration as a function of distance from the release hole, also lengthwise and vertically. As the first purpose of the activity was to prepare data to be tested and validated by CFD models, it can be said that the objective has been achieved; models however should be able to consider opposite wind directions. The type of measurement of meteorological data achieved during the experiment is very important and useful in understanding hydrogen behavior in the atmosphere. Experimental data without this type of information should not be considered to calibrate dispersion models.

The reliability and reproducibility of experiments and data acquired is influenced by the method of acquisition: in particular the system used for gas concentration measurements was quite complex and could lead to errors in the experimental values that are difficult to estimate. In order to achieve more critical release conditions a higher pressure and volume storage of the gas is necessary, but this would require a bigger financial investment and the redesigning of the plant, in terms of pipes and safety equipment. The acquisition system should also be improved with more sophisticated and rapid hydrogen samplers.

A comparison with the two integral models Effects 4.0 and Phast 6.3 was developed. The models considered in this paper overestimated the hydrogen concentration measured during Tests 2 and 3. They also showed some difficulty in reproducing all the experimental conditions as to wind intensity and direction, and outflow velocity. Their use in the risk analysis could be advisable in the absence of more realistic models, but often results could be considered as greatly overestimating the real consequences in case of accidental release. Finally, particular and sometimes very interesting meteorological conditions cannot be simulated.

In the future a comparison with a more sophisticated model, such as a Lagrangian particle model for atmospheric dispersion, will be made in order to validate the model.

ACKNOWLEDGMENTS

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