HYDROGEN STRATIFICATION IN ENCLOSURES IN DEPENDENCE OF THE GAS RELEASE MOMENTUM

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ABSTRACT

The hydrogen dispersion phenomenon in an enclosure depends on the ratio of the gas buoyancy-induced momentum. Random diffusive motions of individual gas particles become dominative when the release momentum is low. Then a uniform hydrogen concentration appears in the enclosure instead of the gas stratification below the ceiling. The paper justifies this hypothesis by demonstrating full-scale experimental results of hydrogen dispersion within a confined space under six different release variations. During the experiments, hydrogen was released into the test room of 60 m^3 volume in two methods: through a nozzle and through 21 points evenly distributed on the emission box cover (multi-point release). Each release method was tested with three different hydrogen volume flow rates ($3.17 \cdot 10^{-3}$ m^3/s, $1.63 \cdot 10^{-3}$ m^3/s, $3.34 \cdot 10^{-4}$ m^3/s). The tests confirm the increase of hydrogen convective upward flow and its stratification tendency relative to increased volume flow. A tendency of more uniform hydrogen cloud distribution when Mach, Reynolds, and Froud number values decreased was demonstrated. Because the hydrogen dispersion phenomena impact fire and explosive hazards, the presented experimental results could help fire protection systems be in an enclosure designed, allowing their effectiveness optimization.

1.0 INTRODUCTION

Industrial uses for hydrogen have been recently expanded due to the element's impressive potential as a source of energy with minimal environmental impact. This has been driven by regulations concerning climate change and the desire to reduce reliance on fossil fuels [1]. Hydrogen is considered a particularly clean and ecological fuel, mainly when it is produced from renewable sources [2]. It could be received, for example, as hydrogen-rich syngas from palm oil mill effluent [3] or from reforming methane [4]. Beneficial physical properties of hydrogen include its density, which is one-tenth (1:10) of natural gas, or the energy capacity per unit of mass, which is three times higher than that of gasoline. This makes it more attractive than other fuels available on the market [5].
Growth in the use of hydrogen fuels is noted in transportation, where hydrogen fuel cell vehicles (FCVs) are introduced dynamically [6]. It is forecasted that, in 10 years, the FCV market would become significant. The benefits of using hydrogen as a vehicle fuel and supporting regulations make it an attractive option, leading to an increase in the number of FCVs. However, despite the benefits, it has to be recognized that hydrogen is also a hazardous gas [7]. This is mainly due to its flammable properties. It is reported that the increasing usage of alternative fuel vehicles (electricity, hydrogen, LNG, LPG, etc.) has significantly altered vehicle fire characteristics [8]. For example, hydrogen ignition energy is ten times lower than methane, and its flame velocity is eight times higher than that of methane [2]. Moreover, its lower flammable limit (LFL), in which the volumetric fraction is equal 4%, is much lower than other flammable gases.
Additionally, hydrogen is an odorless, colorless, and tasteless gas. Human senses are not able to detect it [9]. Consequently, ensuring a safe infrastructure for such vehicles becomes an urgent goal. This goal could be reached by doing research on hydrogen dispersive, burning, and explosive phenomena and protective measures mitigating hazards relative to FCVs using.

Most FCVs are fueled with gaseous hydrogen [10]. It was found that the release of hydrogen from the onboard storage tank, say due to a collision, would be a major risk, especially when initiated by an external fire. Such accidents could create different hazardous outcomes, depending on varying factors. The most dangerous situations would appear when the release happens in an enclosed space, such as a garage. Then the most significant elements would be the volume of the enclosure, its ventilation, and the hydrogen release rate spread from the vehicle [10], [11], [12], [13].

In the case of accidental hydrogen release from the vehicle, several scenarios are possible. These can be captured in an event tree, as shown in Fig. 1. As an initiating event, the release of hydrogen from the car's system is taken into account, with ten different outcomes considered [14]. There are several possibilities of hydrogen release from the FCVs. The first possible scenario is the hydrogen permeation through the tank walls; however, this phenomenon is prolonged and happens mainly in vehicles fueled with liquid hydrogen, which is not popular, as was mentioned earlier [10]. Another scenario could be the hydrogen release from the car's pipe installation when the tank stays closed by a valve. In this case, the gas spread could be much more rapid than previously, but its volume would be limited to the volume of the pipe installation. The third scenario, which is taken into account most often in accidental hydrogen release analyses, is the gas spread directly from a tank through a thermally activated pressure relief device (TPRD), an obligatory element of the VCV's installation [15]. At the same time, this is the most dangerous scenario and could happen in a case of the TPRD failure or a matter of fire when the tank pressure increases due to the surrounding temperature increase [16].

The consequences of the scenarios described above could be more or less severe. It depends mostly on the ignition sources existing in the enclosure, hydrogen detection, and ventilation systems. The most dangerous outcome event could be a hydrogen explosion, which could be a deflagration, detonation, or detonation-to-deflagration transition (DDT) [17], [18], [19]. This scenario could happen when the hydrogen cloud stays at an enclosure in a concentration at least above the lower flammable limit (LFL), and ignition sours appear. It can occur when a ventilation system does not work (is not efficient enough/doesn't exist/is not activated by the detection system) [10]. Another severe outcome event would be a pressure peaking phenomenon (PPP), which happens in enclosures under intensive light gas emission into the heavier gas, under minor ventilation conditions [15], [19], [20]. Another outcome event presented in the event tree (Fig. 1) is the hydrogen gas cumulation in the enclosure. This could be a reason for another event. However, the most probable final event would be a slow gas spread outside of the enclosure. Another event, which is gas dilution and/or evacuation, doesn't create dangerous situations in the enclosure and would happen when the protective measures such as detection and ventilation system would be effective enough, and/or the hydrogen release would be not rapid.

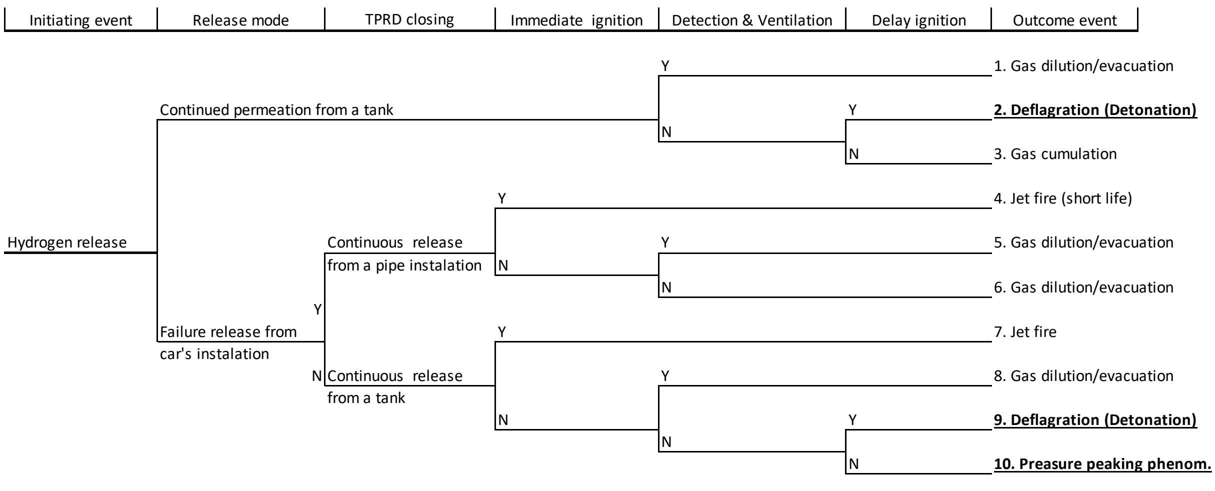


Figure 1. Event tree of hydrogen release incident scenarios

As can be observed in the event tree (Fig. 1), the serious outcome events, which are deflagration (detonation) and PPP proceed in three scenarios, and they will be a subject of the considerations presented in this article. It is demonstrated that those events could happen in the case of hydrogen permeation and continuous release, and their appearance depends on the protective measures applied.

There are several methods for protective measures design. The most important seems to be, in this case, passive ventilation systems. However, their parameters are usually based on the assumption of uniform hydrogen distribution in the enclosure [12], [14]. This assumption is taken due to the time to create a flammable mixture at LFL level, which would be reduced if the uniformity of hydrogen distribution was not valid and permeated hydrogen would gather in a layer [10]. However, the hydrogen stratification would create layers over 4% volumetric concentration, with volumes exceeding the upper limits for hydrogen inventory to prevent damage in a case of explosion [18].

The question is when a uniform or layered hydrogen distribution in an enclosure could be expected? It is known that by adjusting the hydrogen jet velocity at the nozzle exit point, a momentum-controlled or buoyancy-controlled jet could be created. In this case, convective movement can dominate, and gas stratification could be observed. On the other hand, a very slow hydrogen release would be dominated by diffusive motion and lead to the uniformity of the gas distribution [10].

The conducted experiments represent both scenarios presented in Fig. 1 – continued permeation from the tank (multi-point release scenario) and failure release (nozzle release scenario). They confirmed that the method of hydrogen release would have a significant influence on the flammable mixture formation and demonstrate the gas concentration distribution under its emission stage.

2.0 MATERIALS AND METHODS

Measurements were carried out in a confined room of 60 m^3 volume, which detailed parameters are presented in Fig. 2. Hydrogen was released from a cylinder (0.8 MPa) and supplied to the room through a box connected with a cylinder through a 10 m long pipe of Φ =4 mm. The cover of the box with dimensions of 1.0 m × 0.5 m, had 21 openings of Φ =6 mm. The surface also had a space for the pipe of Φ =4 mm distribution for the nozzle release experiments. The hydrogen concentration in the enclosure under experimental conditions, due to safety concerns, was kept below the lower flammable limit (LFL). The temperature in the experimental box was 10ºC. Hydrogen concentration was recorded continuously in six measurement points with sensor VQ-21, located at the heights presented in Fig. 2.

The experiments were undertaken over three stages, with different hydrogen volume outflows: $3.17 \cdot 10^{-3}$, $1.63 \cdot 10^{-3}$, $3.34 \cdot 10^{-4}$ m^3/s. Hydrogen was released upward through a nozzle (the pipe of Φ=4 mm) or through 21 release points in the cover of the box (multi-point release) alternatively. The realized experimental test list is presented in Table 1. Additionally, the Reynolds (*Re*) and Froud (*Fr*) were calculated, which allowed further discussion of the experimental results [21].

Table 1. The realized experimental tests.

Test no.	**Hydrogen release method**	**Hydrogen release outflow [m^3/s]**	**Opening area [m^2]**	**Velocity [m/s]**	***M***	***Re***	***Fr***
Test 1	Nozzle release	$3.17 \cdot 10^{-3}$	$1.256 \cdot 10^{-5}$	252.39	7.42×10^{-1}	9914	1.62×10^{6}
Test 2		$1.63 \cdot 10^{-3}$		129.78	3.81×10^{-1}	5097	4.29×10^{5}
Test 3		$3.34 \cdot 10^{-4}$		26.59	0.78×10^{-1}	1044	1.80×10^{4}
Test 4	Multi-point release	$3.17 \cdot 10^{-3}$	$5.935 \cdot 10^{-4}$	5.34	0.16×10^{-1}	1415	1.05×10^{2}
Test 5		$1.63 \cdot 10^{-3}$		2.75	0.80×10^{-2}	729	2.80×10^{1}
Test 6		$3.34 \cdot 10^{-4}$		0.56	0.20×10^{-2}	148	1.16×10^{0}

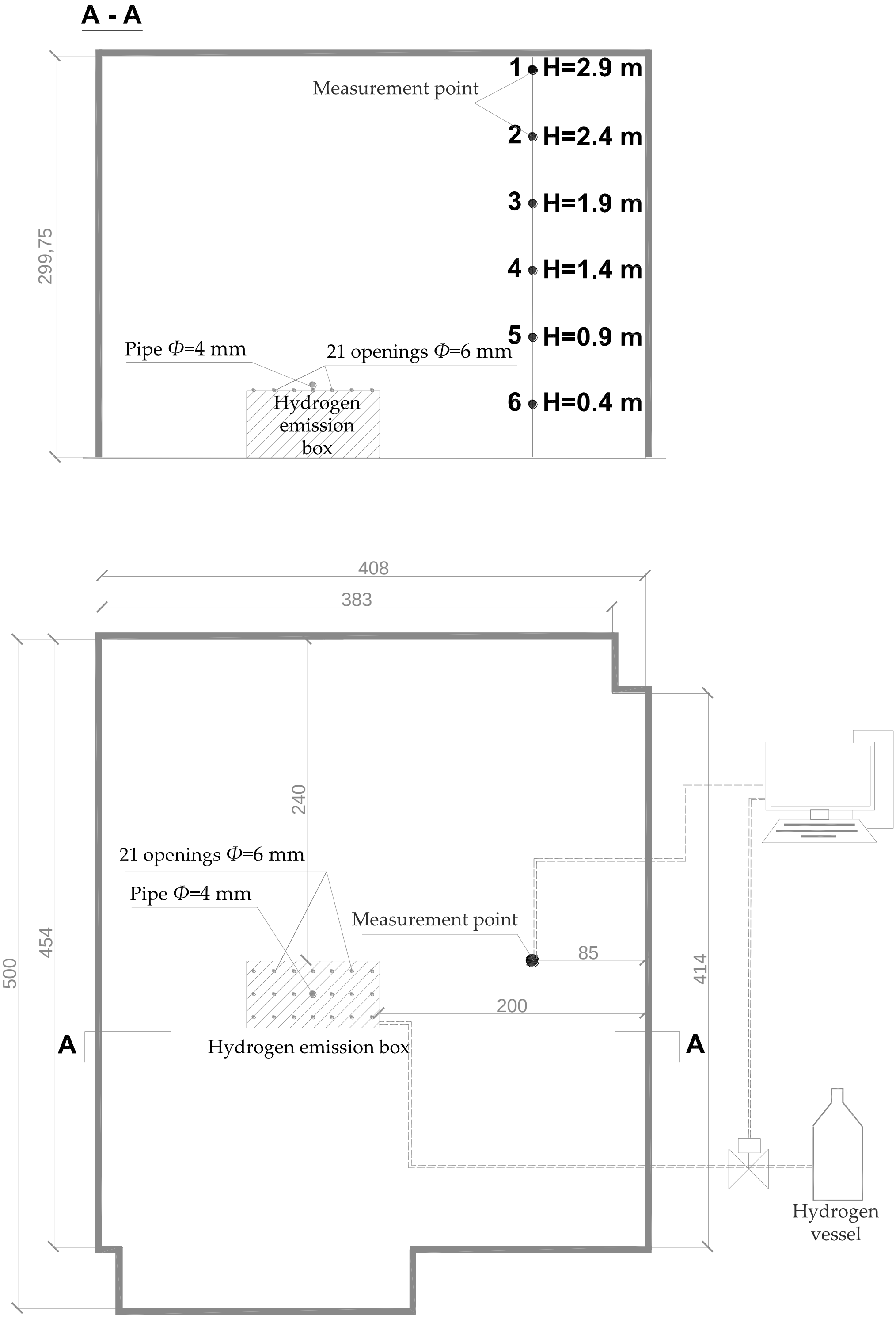
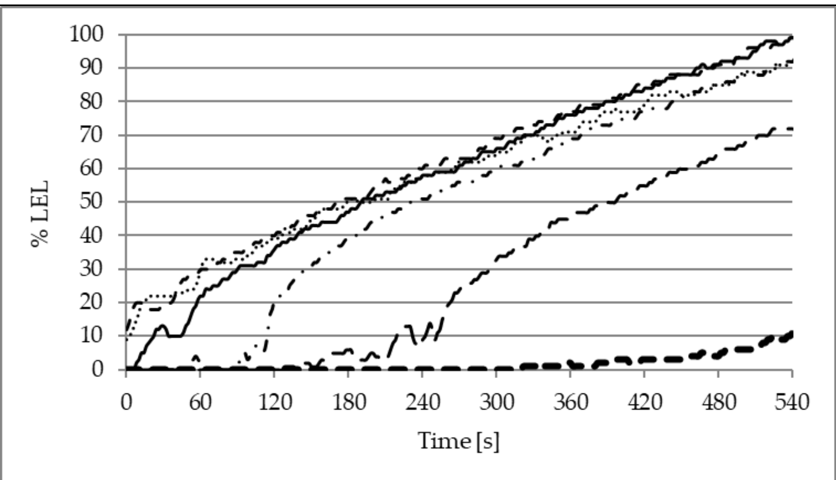
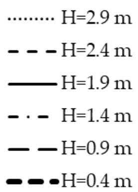


Figure 2. Hydrogen release measurement layout.

3.0 RESULTS

In Tables 2 and 3, the experimental results are presented. The hydrogen concentration in the enclosure under the nozzle and multipoint release is shown in time in several measurement points.

Table 2. Measurement results for hydrogen release from the nozzle.

<table>
<tr><th>Test no.</th><th>Experimental results</th><th>Legend</th></tr>
<tr><td>Test 1</td><td>
</td><td rowspan="3">
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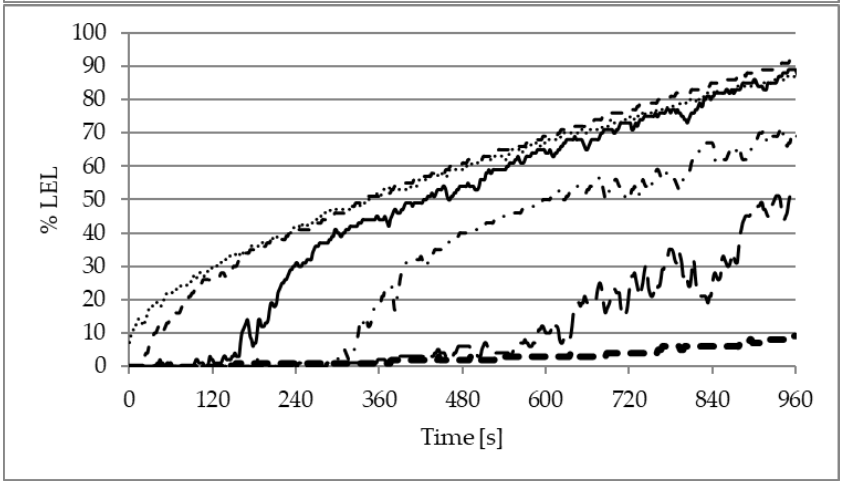
<tr><td>Test 2</td><td>
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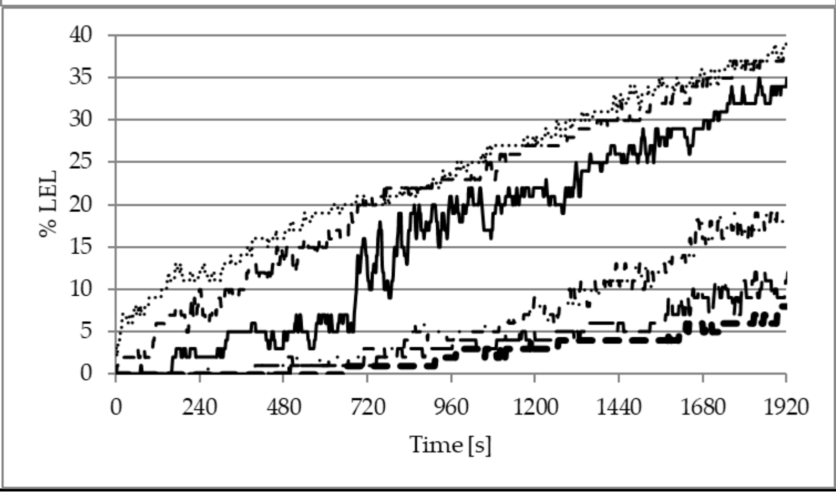
<tr><td>Test 3</td><td>
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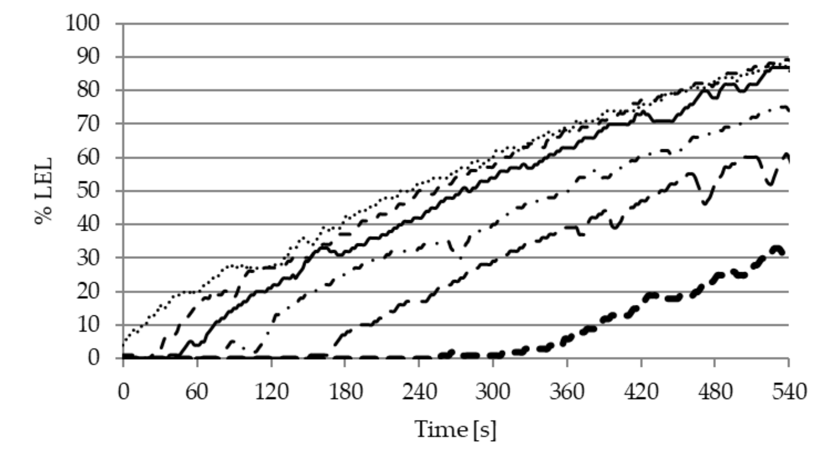
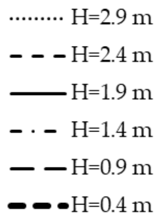
</table>

Table 3. Measurement results for hydrogen release from the multi-release point.

<table>
<tr><th>Test no.</th><th>Experimental results</th><th>Legend</th></tr>
<tr><td>Test 4</td><td>
</td><td rowspan="3">
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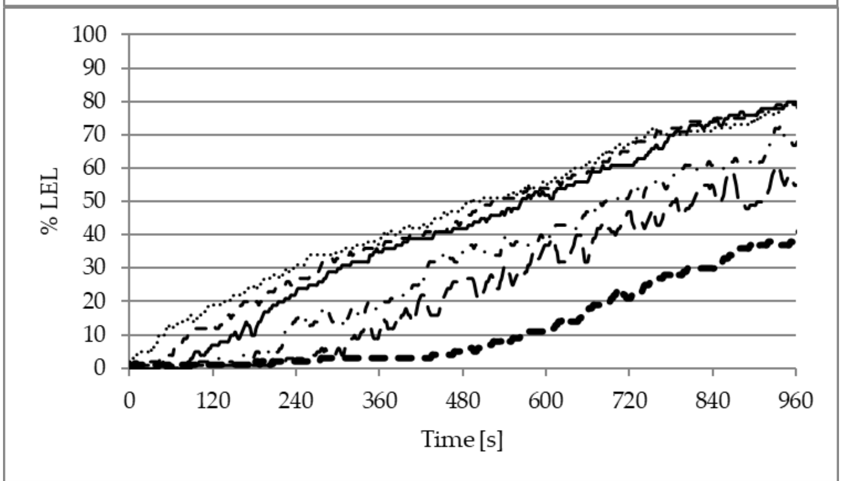
<tr><td>Test 5</td><td>
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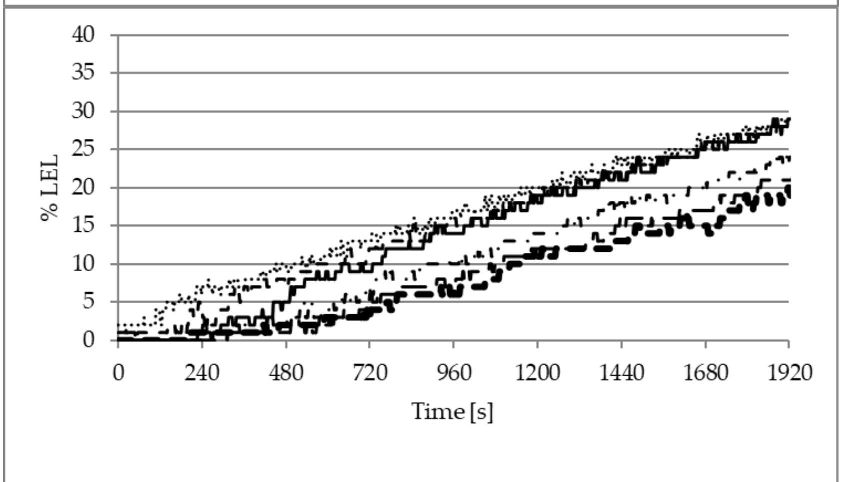
<tr><td>Test 6</td><td>
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4.0 DISCUSSION

The full-scale experiments of hydrogen release and dispersion in the confined space were undertaken as described in Section 3. Hydrogen gas was released into the room in two ways: through a nozzle (T1-T3) and through 21 points evenly distributed over the surface of the emission box (multi-point release Tests 4-6). The experiments assessed three different hydrogen release outflows ($3.17 \cdot 10^{-3}$, $1.63 \cdot 10^{-3}$, $3.34 \cdot 10^{-4}$ m^3/s). In total, six experimental tests listed in Table 1 were undertaken.

The experimental results confirmed that all tests were realized in the subsonic regime (M<1) (Table 1). Tests 1 and 2 discovered with the single nozzle hydrogen release appeared to create strongly turbulent, momentum-dominated jets (Re>2000), when Test 3 and multi-point release tests (4-6) appeared to represent buoyancy-controlled jets. It was also confirmed that the lower the Froud number, the lower the buoyancy force influences the hydrogen dispersion. The Froud numbers received from the experiments confirm their downward trend in subsequent tests (around one order of magnitude), relevant to the decreased buoyancy confirmed. Especially, test T6, in which the Froud number is of the order of one, proves the significantly lower stratification obtained in it than in the other tests, as evidenced by the low ΔH_2 value shown in Figure 4. The presented results confirm that together with the release velocity decreasing, all dimensionless numbers decreased too, affecting a more uniform hydrogen cloud distribution in the compartment.

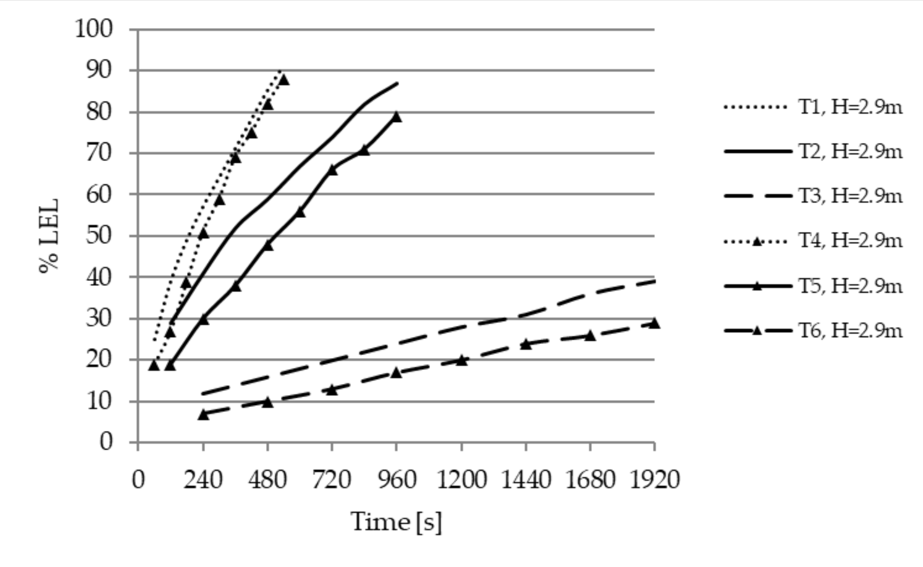
The experimental results demonstrate a significant influence on the dispersion phenomenon of the hydrogen release method and the volume outflow. The most general observation is that, together with the increase of the hydrogen outflow area, the hydrogen's convective upward flow decreases. Consequently, much greater hydrogen stratification in the room is observed under nozzle release conditions than in the case of the multi-release point. This phenomenon can be followed by comparing the experimental results of the same volume outflows but with different release methods (Tables 2, 3). The difference between hydrogen concentration near the ceiling (H=2.9 m) and near the floor (H=0.4) at the end of each experiment are presented in Table 4. It is noted that in T1 and T2, when a relatively big hydrogen outflow was realized through the nozzle, the concentration under the ceiling was nearly ten times higher than near the floor. This concentration ratio decreased with a decrease in hydrogen volume outflow and an increase in the number of openings, reaching below 1.5 times lower concentration under the ceiling than near the floor, as covered in T6.

Table 4. Hydrogen **c**oncentration ratio at heights of H=2.9 m and H=0.4 m.

Test no.	$H_{2,H=2.9\,m}$	$H_{2,H=0.4\,m}$	$\Delta H_2 = H_{2,H=2.9\,m}/H_{2,H=0.4\,m}$
T1	91.0	10.0	9.10
T2	87.0	9.0	9.66
T3	39.0	8.0	4.87
T4	88.0	31.0	2.84
T5	79.0	38.0	2.08
T6	29.0	20.0	1.45

In Tables 5 and 6, the experimental results are broken down into individual measurement heights. It is noted that, in the upper half of the room (Table 5), the concentration of hydrogen released through the nozzle (T1, T2, T3) is significantly higher than through the multi-point (T4, T5, T6). The difference between the concentrations for the experiments with the same volume outflow remain almost constant. This process changes in the lower half of the room (Table 6). At the height of 1.4 m, a significant influence of the volume outflow is observed. For the biggest outflow (T1, T4), the concentration of gas exiting through the nozzle remains higher than through the multi-point. However, this relationship begins to diverge for smaller outflows, giving almost the same concentrations in T2 and T4 and higher concentrations in T6 than T3. Continuation of the above can be observed at the height of 0.9 m, where the results of T1 and T4 are similar (the largest outflow), and T5 and T6 have given much higher results than T2 and T3, respectively. Finally, close to the floor (H=0.4 m), a uniformly higher concentration for tests with the multi-point release than with the nozzle was observed.

Table 5. Hydrogen **c**oncentration under the nozzle and multi-point release at heights of 2.9 m, 2.4 m, and 1.9 m.

<table>
<tr><th>Tests no.</th><th>Experimental results comparison</th></tr>
<tr><td>H=2.9 m</td><td>
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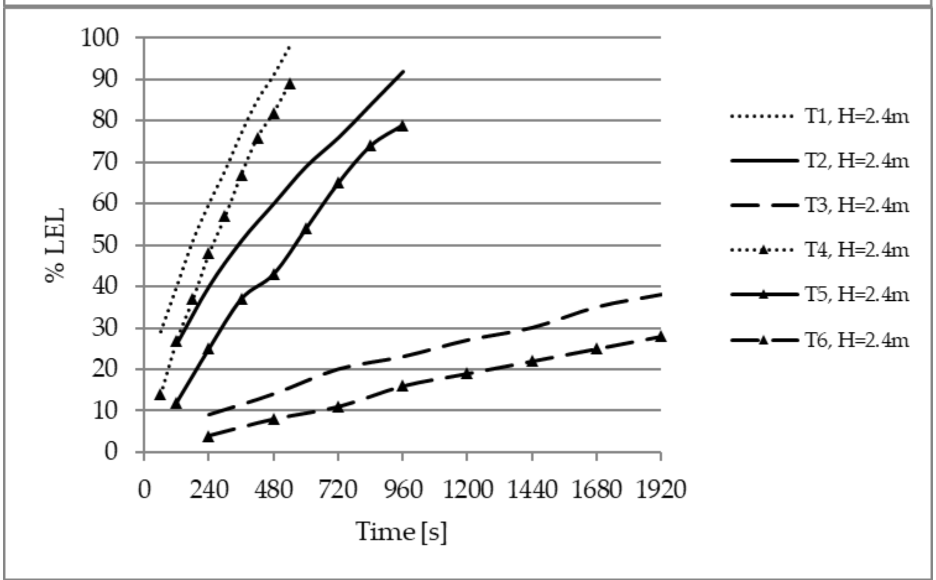
<tr><td>H=2.4 m</td><td>
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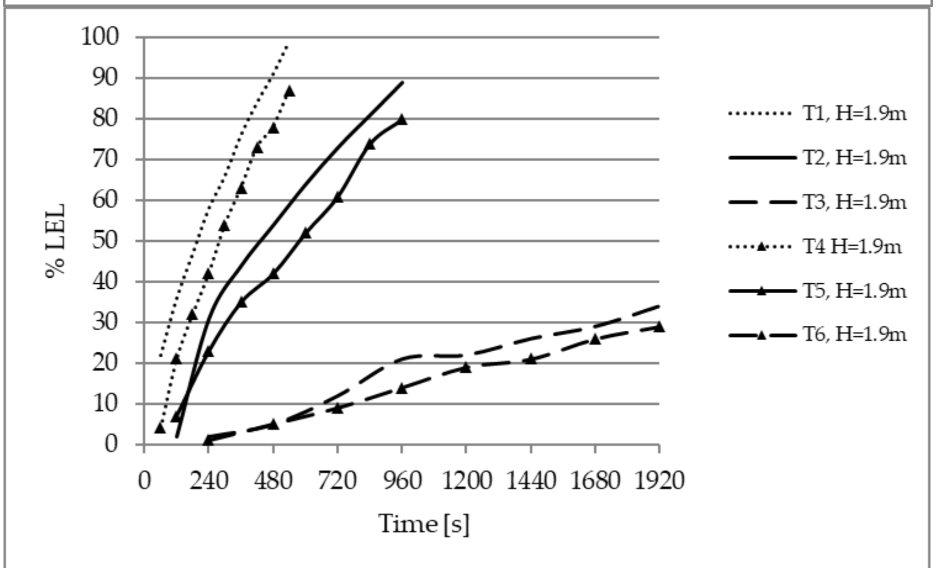
<tr><td>H=1.9 m</td><td>
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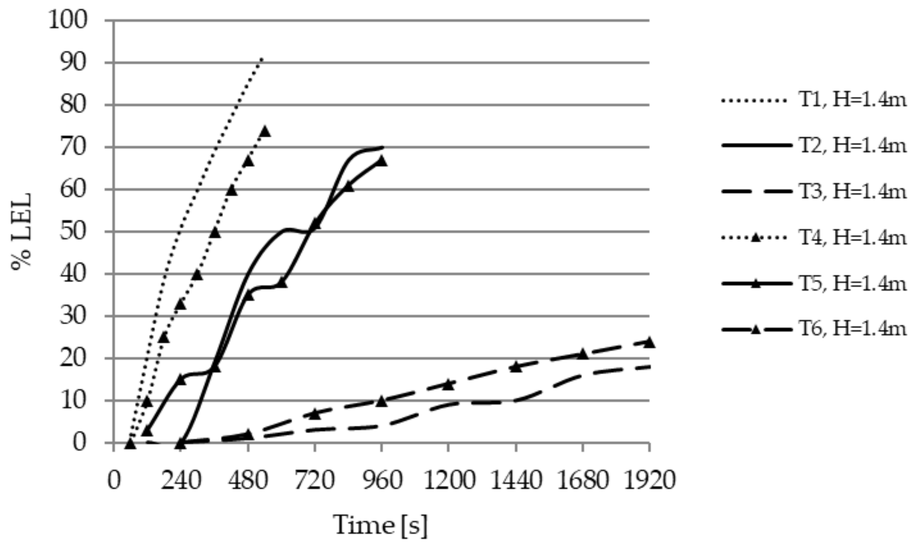
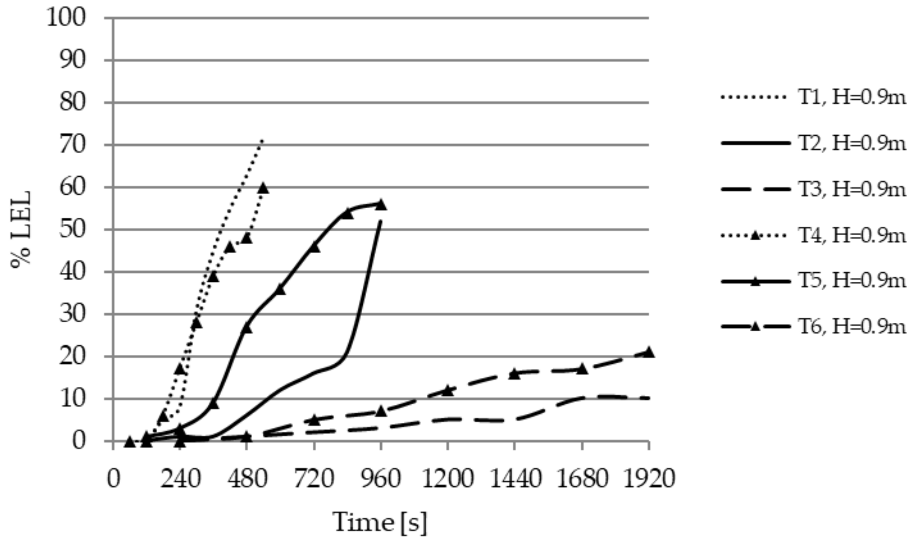
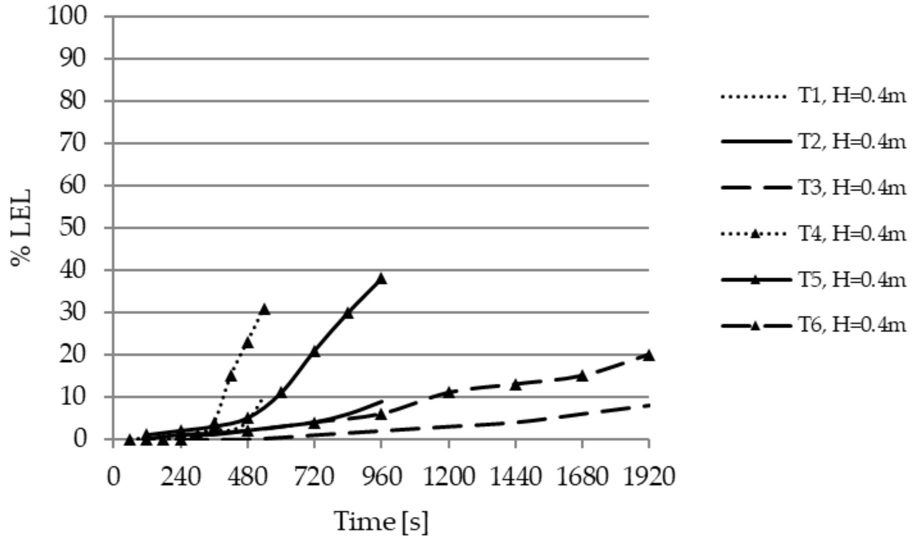
Table 6. Hydrogen concentration under the nozzle and multi-point release at heights of 1.4 m, 0.9 m, and 0.4 m.

Tests no.	Experimental results comparison
H=1.4 m	
H=0.9 m	
H=0.4 m	

4.0 CONCLUSIONS

Six tests of hydrogen dispersion in a confined space were undertaken, with a volume outflow over a range from $3.17 \cdot 10^{-3}$, $1.63 \cdot 10^{-3}$, $3.34 \cdot 10^{-4}$ m^3/s using both nozzle and multi-point release. The results confirm a significant influence on the hydrogen dispersion of both analyzed parameters. The most important observation is the increase of the hydrogen outflow area when the gas's convective upward flow decreases. At the same time, a greater hydrogen stratification is observed in the test room under nozzle outflow conditions, which causes higher gas concentration directly under the ceiling and lower concentration near the floor than in the case of the multi-point release.

The experiments also demonstrate that the increase of hydrogen volume outflow in the nozzle release causes an increase of the gas concentration rather than nearer the floor than under the ceiling. At the same time, the increase of hydrogen volume outflow in the case of multi-point release causes an increase of the gas concentration uniformly in the full space of the room. Mach and Reynolds number calculations allowed to indicate the regimes of the released jets when the Froud number Fr~1 predicted a decline of buoyancy motion in the hydrogen jet flow. A tendency of more uniform hydrogen cloud distribution when Mach, Reynolds, and Froud number values decreased was demonstrated.

The observations presented are important, in practice, for the design of protective measures against hydrogen explosion. Further analysis will include verification by CFD, modeling of hydrogen release, and dispersion phenomenon.

References

[1] V. V. Molkov, "Unified correlations for vent sizing of enclosures at atmospheric and elevated pressures," *J. Loss Prev. Process Ind.*, vol. 14, no. 6, pp. 567–574, 2001.

[2] R. Kaplan and M. Kopacz, "Economic Conditions for Developing Hydrogen Production Based on Coal Gasification with Carbon Capture and Storage in Poland," *Energies*, vol. 13, pp. 1–20, 2020.

[3] Y. W. Cheng, C. C. Chong, S. P. Lee, J. W. Lim, T. Y. Wu, and C. K. Cheng, "Syngas from palm oil mill effluent (POME) steam reforming over lanthanum cobaltite: Effects of net-basicity," *Renew. Energy*, vol. 148, pp. 349–362, 2020.

[4] C. C. Chong, Y. W. Cheng, H. D. Setiabudi, N. Ainirazali, D. V. N. Vo, and B. Abdullah, "Dry reforming of methane over Ni/dendritic fibrous SBA-15 (Ni/DFSBA-15): Optimization, mechanism, and regeneration studies," *Int. J. Hydrogen Energy*, vol. 45, no. 15, pp. 8507–8525, 2020.

[5] M. Molnarne and V. Schroeder, "Hazardous properties of hydrogen and hydrogen containing fuel gases," *Process Saf. Environ. Prot.*, vol. 130, pp. 1–5, 2019.

[6] Y. Tao, J. Qiu, S. Lai, X. Zhang, G. Wang, and P. Ev, "Collaborative Planning for Electricity Distribution Network and Transportation System Considering Hydrogen Fuel Cell Vehicles," *IEEE Trans. Transp. Electrif.*, vol. 6, no. 3, pp. 1211–1225, 2020.

[7] International Organization for Standardization, "ISO/TR 15916:2015 Basic considerations for the safety of hydrogen systems," United Kingdom, PD ISO/TR 15916:2015, 2015.

[8] M. Klassen and D. Ph, "Modern Vehicle Hazards in Parking Structures and Vehicle Carriers," 2020.

[9] D. Brzezińska, "Ventilation system influence on hydrogen explosion hazards in industrial lead-acid battery rooms," *Energies*, vol. 11, no. 8, pp. 1–11, 2018.

[10] V. Molkov, *Fundamentals of Hydrogen Safety Engineering I*, vol. 1. 2012 Vladimir Molkov & bookboon.com, 2012.

[11] S. L. Brennan, D. V. Makarov, and V. Molkov, "Safety assessment of unignited hydrogen discharge from onboard storage in garages with low levels of natural ventilation," *Int. J. Hydrogen Energy*, vol. 22, no. 3, pp. 353–359, 2009.

[12] V. Molkov, V. Shentsov, and J. Quintiere, "Passive ventilation of a sustained gaseous release in an enclosure with one vent," *Int. J. Hydrogen Energy*, vol. 39, no. 15, pp. 8158–8168, 2014.

[13] V. Molkov, V. Shentsov, S. Brennan, and D. Makarov, "Hydrogen non-premixed combustion

in enclosure with one vent and sustained release: Numerical experiments," *Int. J. Hydrogen Energy*, vol. 39, no. 20, pp. 10788–10801, 2014.

[14] S. Brennan and V. Molkov, "Safety assessment of unignited hydrogen discharge from onboard storage in garages with low levels of natural ventilation," *Int. J. Hydrogen Energy*, vol. 38, no. 19, pp. 8159–8166, 2013.

[15] D. Makarov, V. Shentsov, M. Kuznetsov, and V. Molkov, "Pressure peaking phenomenon: Model validation against unignited release and jet fire experiments," *Int. J. Hydrogen Energy*, vol. 43, no. 19, pp. 9454–9469, 2018.

[16] V. Molkov, V.; Shentsov, "Regimes of hydrogen jet fire in an enclosure with two vents," in *6th Int. Symp. Non-Equilib. Process. Plasma Combust. Atmospheric Phenom.*, 2016, pp. 129–136.

[17] V. Molkov, *Fundamentals of Hydrogen Safety Engineering II*. 2012 Vladimir Molkov & bookboon.com, 2012.

[18] D. Makarov, P. Hooker, M. Kuznetsov, and V. Molkov, "Deflagrations of localised homogeneous and inhomogeneous hydrogen-air mixtures in enclosures," *Int. J. Hydrogen Energy*, vol. 43, no. 20, pp. 9848–9869, 2018.

[19] V. Molkov and M. Bragin, "HYDROGEN-AIR DEFLAGRATIONS: VENT SIZING CORRELATION FOR LOW-STRENGTH EQUIPEMENT AND BUILDINGS," 2015.

[20] A. W. Lach, A. V. Gaathaug, and K. Vaagsaether, "Pressure peaking phenomena: Unignited hydrogen releases in confined spaces – Large-scale experiments," *Int. J. Hydrogen Energy*, vol. 45, no. 56, pp. 32702–32712, 2020.

[21] D. Brzezińska, "Hydrogen dispersion phenomenon in nominally closed spaces," *Int. J. Hydrogen Energy*, 2021.