HYDROGEN-AIR EXPLOSIVE ENVELOPE BEHAVIOR IN CONFINED SPACE AT DIFFERENT LEAK VELOCITIES

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ABSTRACT

The report summarizes experimental results on the mechanisms and kinetics of hydrogen-air flammable gas cloud formation and evolution due to foreseeable (less than 10⁻³ kg/sec) hydrogen leaks into confined spaces with different shapes, sizes and boundary conditions. The goals were - 1) to obtain qualitative information on the basic gas-dynamic patterns of flammable cloud formation at different leak velocities (between 9,35 and 905 m/sec) for a fixed leak flowrate and 2) to collect quantitative data on spatial and temporal characteristics of the revealed patterns. Data acquisition was performed using a spatially distributed, reconfigurable net of 24 hydrogen gauges with short response time. This experimental innovation permits to study spatial features of flammable cloud evolution in detail, which previously was attainable only from CFD computations. Two qualitatively different gasdynamic patterns were documented for the same leak flowrate. In one limiting case (sufficiently low speed of leak), the overall gas-dynamic pattern can be described by the well-known "filling box" model. In another limited case (high velocity of leak), it is proposed to describe the peculiarities of gas-dynamic behavior of flammable cloud by the term of a "fading up box" model. From the safety view point, the "fading up box" case is more hazardous than the "filling box" case. Differences in macroscopic and kinetic behavior, which are essential for safety provision, are presented. Empirical non-dimensional criterion for discrimination of the two revealed basic patterns for hydrogen leaks into confined spaces with comparable length scale is proposed. The importance of the revealed "fading up box" gas-dynamic pattern is discussed for development of an advanced hydrogen gauges system design and safety criteria.

1.0 INTRODUCTION

Mechanisms and kinetics of hydrogen-air flammable gas cloud formation and evolution due to foreseeable (less then 10⁻³ kg/sec) hydrogen leaks into confined spaces is an important issue for the development and design of hydrogen safety systems, based on hydrogen gauges. In order to provide an enhanced and assured level of protection against hypothetical hydrogen releases/leaks clear answers to the following questions are required: What is the most probable gas-dynamic pattern (geometrical form and its macroscopic evolution) of a flammable cloud formation for a given set of the initial/boundary conditions ? How quickly is a potentially flammable (explosive) cloud forming for a given gas-dynamic pattern ? How large (in volume or in mass) will the flammable cloud be, when it can be detected by available hydrogen gauges ? How quickly will the flammable cloud pose a real threat to the surrounding enclosure ?

Information on mechanisms and kinetics of flammable clouds formation and evolution is presented in various sources. Comprehensive information sources for heavier than air gases (mainly, gaseous hydrocarbons and for outdoor, free jets conditions) are summarized in [1]. Information on methane behavior in enclosures is given in [2]. An excellent introductory review, specifically targeted on the hydrogen safety problems in enclosures, is made in [3].

The overall long-term goal of our research efforts [4] is to build a database of consistent, accurate and validated experimental data on hydrogen leaks into confined spaces with different shapes, sizes and boundary conditions. The database is intended to be used for 1) development of the performance-based requirements for allocation and actuation of hydrogen gauges, 2) robust validation of CFD codes.

Specifically, this report summarizes experimental results on the mechanisms and kinetics of hydrogenair flammable gas cloud formation and evolution for foreseeable (less then 10^{-3} kg/sec) hydrogen leaks into confined spaces with comparable scales (length, width, height). Experiments were performed in a fixed flow rate and varied outflow velocities.

2.0 EXPERIMENTS

2.1 Measurement and data acquisition system

Data acquisition was performed using a spatially distributed, reconfigurable net of 24 hydrogen gauges (hydrogen concentration + temperature) with a short response time. The following considerations have been taken into account during the in-house acquisition system development: high noise immunity, simplicity of assembling new connecting sensors, possibility to simultaneously interrogate from 24 to 32 separate sensors, high response speed of data acquisition system – sampling time should be less than the response speed of sensor. The time of overall data collection from 24 sensors is less than 300 ms. This experimental innovation permits to study spatial features of flammable cloud evolution in detail, which previously was attainable only from CFD computations. Details of data acquisition system are given in [5, 6].

2.2 Description of experimental setups

Three sets of experiments have been carried out to investigate hydrogen (or helium as a hydrogen surrogate) releases into the air confined by enclosure.

Unventilated barrel (December 2006 – October 2007)

The first set of experiments has been carried out in a metal 4 m^3 unventilated cylindrical barrel, placed horizontally with two semispherical covers [4]. The internal view of the experimental chamber and the schematic of the spatial allocation of the sensors and hydrogen source are presented at Figure 1.



Figure 1. The internal view (left) of the experimental chamber and the schematic of the spatial allocation of the sensors (right) [5].

The experiments have been carried out at hydrogen flow rate $0,48*10^{-4}$ kg/s (0,53 l/s), the output nozzle can be directed vertically upward, vertically downward and horizontally. In the experiments the stratification of hydrogen distribution from top to bottom was observed. The stratification value depended on the direction of initial hydrogen release.

Ventilated hydrogen facility at UNIPI (10-19 March 2008)

The second set of experiments has been carried out in the laboratory of Prof. Carcassi at UNIPI (University of Pisa). The experimental chamber was a cube with transparent walls made from organic glass and metal frame, volume of the cube is 25 m^3 .



Figure 2. General view (left) of the experimental chamber of UNIPI and the schematic of spatial allocation of the sensors (right) [6].

The experimental chamber was equipped with special square windows for ventilation. The general view of the experimental chamber and the schematic of the spatial allocation of the sensors, hydrogen source and ventilation windows are presented on Figure 2. Hydrogen was released vertically upward and horizontally sideways with flow rate $0.57*10^{-4}$ kg/s (0.63 l/s) through a tube (nozzle) with 0,001 m diameter. In these experiments a sharp distinction (in comparison with the first set of experiments) in the hydrogen flow pattern was observed – namely there was no evidence of stratification along the vertical axis. As we assumed later, it was likely to be connected with essential differences in outflow velocities during release from the nozzles of different diameters at about the same hydrogen mass flow rate.

"Surrogate garage" (May 2008 – January 2009)

The third set of experiments has been carried out for a more detailed study of the effect of hydrogen release speed on basic gas-dynamic patterns. In these targeted experiments helium (as hydrogen surrogate) was used.



Figure 3. Internal view (left) of the experimental chamber and the representative schematic of spatial allocation of the sensors (right). Helium source coordinates: x = 1,16 m, y = 0,49 m.

The experimental chamber was a parallelepiped with height 2,02 m, length 2,32 m and depth 1,9 m. The walls of the surrogate "garage" were made from fiber boards. The joints were sealed. This

prevents an essential gas leakage from the experimental volume. The internal view of the "garage" and the schematic of the spatial allocation of the sensors are presented in Figure 3. Helium from the gas vessel was supplied into the experimental chamber through a pipe and was released through tubular nozzles of different diameters (from 0,0006 to 0,008 m) in controlled conditions. The value of the release gauge pressure varied from 0,00008 to 2,19 bar. The flow rate of helium release was fixed at $8,4*10^{-5}$ kg/s (or 0,47 l/s) level. Tubular nozzles were placed exactly at the centre of the experimental chamber, the distance between the gas release point and the floor was 0,49 m. The value of gas flow rate was selected so, that to create large volumes of explosive mixtures with concentration higher than 4% (volume) in reasonable time (about 15 - 20 min). Data records were made synchronously from 22 sensors with the rate of two readings per second.

3.0 EXPERIMENTAL RESULTS

Two basic gas-dynamic patterns: "filling box" and "fading up box"

Two qualitatively different gas-dynamic patterns were documented for the same leak flow rate.

In one limiting case (sufficiently low speed of leak – less than 150 m/s), the overall gas-dynamic pattern can be described by a well-known "filling box" model [7]. Here, hydrogen (helium) jet, released from a nozzle, is transformed into plume before reaching the "garage" ceiling. In this case hydrogen is first accumulated under the ceiling of the room and then propagates down slowly, and here the main "driver" of hydrogen-air mixing is hydrogen plume.

In the other limiting case (high velocity of leak), it was proposed to describe the peculiarities of gasdynamic behaviour of flammable cloud as a "fading up box" model. Here the hydrogen (helium) jet, released from a nozzle, "touches" the ceiling without transforming into a plume. In this case hydrogen (helium) concentration increases practically uniformly through the whole free volume above the nozzle, and here the main "driver" of hydrogen-air mixing is hydrogen jet.

Macroscopic differences between "filling box" and "fading up box" cases

Distinguished features of these two limiting experimentally documented regimes (at the same mass flow rate) are the following:

1. **Macroscopic kinetics of explosive cloud formation**. Sensible volumes of hydrogen-air flammable clouds in the "filling box" regime are formed several times (see Figure 4 below) faster than in the "fading up box" regime, however in so doing the rate of explosive cloud increase is relatively low. The situation for the "fading up box" regime is reverse – explosive mixture was created with noticeable delay in comparison with the first case, but the rate of explosive cloud increase was very high.

From the safety point of view, one of the most important characteristics of explosive cloud (hydrogen concentration range is from 4 vol.% to 74 vol.%) is growth. Explosion cloud growth rate value defines the available time for emergency response. Actuation of alarm, protective and/or mitigation systems shall be made before the explosive cloud attains a size (potentially harmful), which will intolerably threaten the enclosure or the equipment within it.

The existence of sufficiently dense mesh of gas gauges allows to approximate the value of helium concentration in every point of the room except the area in the direct vicinity of the jet. Thus, it is possible to estimate the "explosive" mixture envelope at each moment of the experiment by summing products of W*L* Δz (W – room width; L – room length) along the room height (in conditions that helium concentration at z height is within 4% – 74% range at each moment) and neglecting the volume of the "explosive" mixture in the direct vicinity of the jet. The results of these calculations when helium release lasts 25 minutes for minimal (D₀ = 0,0006 m) and maximal (D₀ = 0,008 m) nozzle diameters are presented in Figure 4.



Figure 4. Time evolution of "explosive" mixture envelope for two different regimes of gas outflow.

As it can be seen at Figure 4, in case of large diameter of release ($D_0 = 0,008$ m) (plume formation) the "explosive" mixture envelope in the upper part of the room appears at 7-th minute and grows relatively slowly – 50% of the room is filled up by "explosive" mixture within approximately 8 min. In the opposite case of a small diameter of release ($D_0 = 0,0006$ m) with the same helium flow rate the "explosive" mixture envelope appears at 19-th minute only, but its growth rate is very high (about 6 times higher than in the "filling box" case) – 50% of the room is filled up by "explosive" mixture within 1,4 min after the flammable cloud formation.

2. **Level of vertical stratification**. In the "filling box" case, the average gradient (see Figure 5) of hydrogen concentration at two thirds (see details at Figure 6 and 7) of "source-ceiling" distance is about ten times higher than the average concentration gradient in the "fading up box" case.



Figure 5. Time dependence of vertical helium concentration gradient on 2/3 length of gas propagation from ceiling to floor for 0,0006 m and 0,008 m nozzle diameters; helium flow rate is $8,4*10^{-5}$ kg/s.

It is obvious that the average helium concentration gradient calculated for 2/3 length of gas propagation for these two extreme cases of outflow differs dramatically (10 times).



Figure 6. Time dependence of helium concentration at vertical line of sensors (NN 17 - 7 - 4 - 9 - 15 - 21 at Fig.3) (0,35 m apart from the jet axis) at "filling box" regime (0,008 m nozzle - left) and at "fading up box" regime (0,0006 m nozzle - right).



Figure 7. Helium concentration profiles at "filling box" (left) and "fading up box" (right) regimes from consequently 0,008 m and 0,0006 m nozzles, data were collected every 2 min.

It can be seen at Figure 7 that at low outflow velocities ($D_0 = 0,008$ m) helium (hydrogen) is accumulated under the ceiling of the room and then propagates down slowly to the nozzle (source). At fast outflow velocities ($D_0 = 0,0006$ m) helium (hydrogen) concentration increases practically uniformly in the free volume above the nozzle. The gas-dynamic patterns for intermediate nozzle diameters are of transitional character between the two limiting types ("filling box" and "fading up box").

4.0 DISCUSSION

Gas-dynamic pattern discrimination criterion – Morton number

We propose to characterize the difference between the two revealed cases by a non-dimensional ratio of plume formation distance (L_i) (see definition in [8]) and distance from release point to ceiling (Z_r).

In our experiments, for all situations where this ratio was small - $L_j/Z_r \langle \langle 1, a | arge inhomogeneity (helium stratification along the height) was observed (see Table) inside the experimental volume. At the same time when the ratio <math>L_j/Z_r \rangle \rangle 1$, helium mixed well with air in the whole free space above the release point.

N⁰	Release diameter D_0 , m	Release gauge pressure, bar	Initial velocity w ₀ , m/s	Plume formation distance L _i , m	Morton number Mo = L_j/Z_r	Type of release regime inside the enclosure
1	0,0006	2,19	905*	7,3	4,8	Jet
2	0,001	0,35	461*	4,8	3,1	Jet
3	0,002	0,020	151	2,23	1,5	Transition
4	0,004	0,0012	37,4	0,78	0,5	Plume
5	0,008	0,00008	9,35	0,28	0,18	Plume

Table 1. Characteristic parameters for different release diameters (initial velocity of gas stream)

* taking into account the value of helium release gauge pressure.

Analysis of the data from Table 1 showed that, in the first limiting case ("filling box"), in accordance with our assumption, the hydrogen (helium) jet, released from the nozzle, is transformed into a plume before reaching the room ceiling. In the second case ("fading up box") the hydrogen (helium) jet, released from the nozzle, is not transformed into a plume before it reached the room ceiling, therefore the vertical jet strikes the ceiling with non-zero momentum.

In order to obtain an additional argument for our hypothesis, we made targeted experimental measurements of helium concentration drop along the jet/plume axis. In accordance with the theory [8], the plume and jet flows are characterized by different dependencies of concentration drop along the jet axis. For jets, the following correlation takes place - 1/z (concentration is inversely proportional to the distance from source - z).For plumes, another correlation is valid - $1/z^{5/3}$.

The measured dependencies of helium concentration upon distance are presented at Figures 8 for different outflow regimes at the first minute of gas release (when helium cloud moving down from the ceiling does not strongly disturb the jet flow field).

In case of outflow from a small diameter nozzle ($D_0 = 0,0006$ m) factors of power-law dependence are close to -1 (averaged amount along the whole distance is -0,9). It means that gas-dynamic flow is jet-like.

In case of outflow from a large diameter nozzle ($D_0 = 0,008$ m) at the distance from the nozzle about 0,2 m an incline of the curve is practically equal to -1, that means that in this area jet flow prevails. At the longer distance the power-law factor increases up to -1.6, so plum flow prevails there (averaged amount of power-law factor along the distance is -1,32). Thus, it is clear that at the distance from 0,2 to 0,4 m (in case of outflow from the 0,008 m nozzle) the jet released from the nozzle is transformed into a plume, which fully corresponds to the length of plume formation calculated for this case and presented in Table 1 (0,28 m).



Figure 8. Dependencies of helium concentration upon distance from the release point at jet axis for different outflow regimes (k - factors of power-law dependence for helium concentration on the distance).

So, both numerical assessments of plume formation length and direct experimental measurements of concentration drop upon distance along axis prove, that in the experiments under discussion variation of the gas outflow velocity (via variation of release diameter under fixed flow rate) results in variation in dominant flow regime. In low velocity regimes (large diameter) a plume-like gas-dynamic flow and "filling box" mixing take place. In high velocity regimes (small diameter), a jet-like gas-dynamic flow and "fading up box" mixing are dominant.

On the base of the results obtained, we suggest using ratio

$$Mo = L_{j}/Z_{r} \tag{1}$$

as a non-dimensional criterion, which indicates a relative role of "plume-drived" and "jet-drived" hydrogen-air mixing in enclosure with comparable dimensions. It will be reasonably to name this ratio as a Morton number - *Mo*.

In the formula for the Morton number, the following definitions are used:

Z_r – distance from gas release point to ceiling,

L_i - plume formation distance (see [8]),

$$L_{j} = 1.02 \frac{M_{0}^{3/4}}{F^{1/2}} = \frac{0.96w_{0}\sqrt{D_{0}}}{\sqrt{\frac{g(\rho_{a} - \rho)}{\rho_{a}}}}$$
(2)

$$F = w_0 g \frac{\rho_a - \rho}{\rho_a} \pi \frac{D_0^2}{4} - \text{buoyancy flux},$$

$$M_0 = \frac{w_0^2 \pi D_0^2}{4}$$
 - release momentum,

w₀ - initial velocity of hydrogen flow at release point,

 ρ_a – the ambient air density,

g – specific gravity,

 D_0 - nozzle diameter.

5.0 CONCLUSIONS

Analysis of the results of the experimental studies of the hydrogen explosive cloud envelope behavior, targeted on the foreseeable (< 10^{-3} kg/sec) hydrogen releases and performed by using a spatially distributed net of 24 hydrogen sensors at three experimental facilities with different boundary/initial conditions, results in the following conclusions:

1. **Phenomenology**. For a given value of hydrogen release flowrate, two basic patterns of hydrogen explosive envelope evolution inside a confined enclosure have been discriminated experimentally. In a "filling box" case (at a low speed of hydrogen outflow), the explosive cloud initially forms as a thin layer at the ceiling and then expands via concentration front downward. In a "fading up box" case (at a high speed of hydrogen outflow), the explosive cloud forms nearly uniformly throughout the whole volume above the discharge point.

2. **Discrimination criterion**. We propose to characterize the difference between the two revealed cases by a non-dimensional ratio of plume formation distance (L_j) and distance from the release point to the ceiling (Z_r) . Ratio Mo= L_j/Z_r can be named as a Morton number. According to our experimental database, for situations, where Mo < 1, a "filling box" case takes place, for Mo >> 1 a "fading up box" case occurs.

3. **Precaution for safety provisions**. Kinetic features of explosive cloud formation should be taken into account when designing hydrogen safety systems. The "filling box" case has a relatively short delay time (time before the first detection of residual concentrations of hydrogen) and a rather long (for actuation and timely response to hydrogen explosion threat) time for the hydrogen-air cloud to grow to a hazardous scale. Unlike the "filling box" case, the "fading up box" case has a long delay time and an extremely high development rate of explosive cloud for the same mass flow rate. It can be said that the explosive volume appears almost at once in the space between the hydrogen release point and the ceiling. In particular, in our experiments the time between the first alarm detection of hydrogen and the formation of the explosive cloud varies about 3 times for the two cases mentioned, and this difference does not practically depend on the sensitivity level of gas detectors. Due to the principal difference in patterns of the explosive cloud formation in "filling box" and "fading up box" cases, it is essentially important that this difference should be taken into account when special technical or organizational measures of hydrogen safety (ventilation, hydrogen detection system, recombiners allocation and so on) are developed and implemented.

4. **Next steps**. The majority of our experiments were performed in enclosures, whose characteristic scales (height, width, length) are comparable. So, the revealed features of explosive cloud formation and evolution are directly applicable to the examined geometry (boiler rooms, garages for single cars, etc.) only. Patterns of explosive cloud formation for the "channel" (car repair workshops) and "slab" (undeground parking for numerous cars) geometries should be studied separately.

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