

The critical mass for the unconfined vapour cloud explosion of compressed and liquid hydrogen

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Abstract

The (unconfined) vapour cloud explosion (VCE) is a dramatic phenomenon that generates a severe pressure wave with a high potential to damage assets and produce injuries in the far field. This definition applies also to hydrogen. Nevertheless, no clear tools and methodology have been so far developed and tested for this highly reactive gas, and even advanced numerical simulations lack validation and suffer from large uncertainties. In this view, the comprehension of the physics which subtends this dramatic phenomenon for the specific case of hydrogen is still a central issue. This paper revises some of the most adopted theories on VCE based on classical acoustic theory and models for pressure wave propagation and provides a consequence-based, threshold (minimum) value for the critical mass of hydrogen m_f^{crit} (4.0 kg) which is needed—at a stoichiometric concentration in air—for a vapour cloud to behave as a VCE. To this regard, any non-stoichiometric hydrogen concentration in air or lower amount of hydrogen would decrease either the flame Mach number M_f or the total energy, thus resulting in negligible overpressure. In this sense, the effects of buoyancy, diffusivity, and weather conditions on the dispersion of hydrogen should be taken into account. The results are valid either for compressed or cryogenic liquid tanks and can be adopted for the sake of distinction between hydrogen flash fire and VCE; for the hazard analysis of hydrogen production and storage; and more in general for the risk assessment of hydrogen systems.

KEYWORDS

critical mass, deflagration, detonation, energy-scaled plot, hydrogen, vapour cloud explosion

1 | INTRODUCTION

A vapour cloud explosion (VCE) may be defined as the explosion resulting from igniting a large cloud of flammable vapour, gas, or mist in which flame speeds accelerate to sufficiently high velocities to produce significant

overpressure in the far field with respect to the source region. The VCE is often considered a rare event, albeit with dramatic consequences on industrial assets, buildings, and the population living in the surroundings.

The prediction of the likelihood and severity of the unconfined or partially confined explosion of hydrogen

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clouds is under debate.^[1] Indeed, either the hazard assessment or the analysis of the consequences suffers from the lack of experiments and poor knowledge of the physics of dispersion, ignition, and the explosion of hydrogen in open areas. Besides, not differently from other hydrocarbons, the intrinsic difficulties of testing large-scale explosions have forced scientists and technologists to adopt poorly validated numerical approaches, such as computational fluid dynamics (CFD), for the analysis of the dispersion and the prediction of the pressure wave generated by the flame propagation. Nevertheless, due to the centrality of the near-future hydrogen economy and due to the relevance of hydrogen safety, literally hundreds of papers and technological reports have addressed the numerical analysis (rather than experimental analysis) of hydrogen-air explosions in unconfined, confined, or partially confined systems in the last decade. Most of the studies focus on small-scale domains because of the difficulties of reproducing hydrogen combustion in detail. Quite obviously, large uncertainties are associated with these estimations, making the obtained results loosely applied for the prediction of the actual large-scale explosion of the unconfined VCE, which is characterized by highly turbulent combustion, high energy density, and dramatic effects on structures and people in the far-field.

This work analyzes the state of the art and the gaps in the knowledge for the VCE, with specific reference to the hydrogen release in the gas (compressed hydrogen, CH₂) and liquid phase (liquefied hydrogen, LH₂), providing possible directions for future experiments and some hints for numerical models, either integral or CFD. Hence, the critical mass of hydrogen for the actual definition of VCE according to its definition is proposed, giving a useful threshold value for the mass flow rate of hydrogen release, for the inclusion of this accidental phenomenon in the event tree analysis, for the definition of barriers, and more in general for the aims of the risk assessment.

2 | THE VAPOUR CLOUD EXPLOSION (VCE)

From a physical perspective, the phenomenon of the VCE is quite complex because it includes the analysis of the formation of the cloud (which includes the fuel source and its dispersion in the atmosphere), the analysis of the turbulent and laminar combustion phenomena, the physics of waves, and other fundamental sciences. Indeed, large uncertainties are still carried out by numerical analysis even for common, well-known hydrocarbons.

In restricting the analysis to the actual explosion phenomenon, thus neglecting the source and the

dispersion phase, the determination of the pressure wave produced in free space by the combustion of a spherical vapour cloud was first analyzed by Strehlow et al.,^[2] starting from the approach of Lighthill,^[3] under acoustic approximation, that is:

$$\frac{\rho - \rho_0}{\rho_0} \ll 1 \quad (1)$$

where ρ and ρ_0 are respectively the density of the compressed air (due to pressure wave) and the ambient density. Lighthill states that at any distance r from the source point, a source of mass $\dot{m}(t)$ can generate an isotropic sound wave characterized by an over-pressure $P - P_0$, where P_0 is the ambient pressure, according to the following equation:

$$\frac{P - P_0}{P_0} = \frac{\gamma}{\rho_0 c_0^2} \frac{\dot{m}(t)}{4\pi r} \quad (2)$$

where $t = t' - r/c_0$ (t' is the actual time), because the wave is propagating away from the source region at the velocity of sound c_0 at the conditions of the undisturbed atmosphere, and γ is the specific heat ratio. This equation is only valid in the far-field (point source approximation), that is, if the frequency ω of the explosion is negligible for the sound speed c_0 , or:

$$\left(T \frac{c_0}{r}\right)^2 \ll 1 \quad (3)$$

where T is the characteristic period of the phenomenon.

According to Strehlow et al.,^[2] for a combustion phenomenon, the term $\dot{m}(t)$ corresponds to the expansion of the hot combustion products after the flame propagation through the unburned gas mixture, which can be approximated—for slow deflagration—with the variation in the time of the overall rate of combustion $\dot{m}_c(t)$. Based on the nature of the scenario, different phenomena having significantly different characteristic times can be distinguished with obvious implications on the overall rate. Indeed, considering a purely diffusive scenario (e.g., jet fire or pool fire), $\dot{m}_c(t)$ is strongly limited by the mixing/diffusive phase of the fuel in the air, hence the pressure wave generated by the explosion is far weaker than in the case of pre-mixed combustion. In this last case, the term $\dot{m}_c(t)$ can be correlated to the mass of combustion products m_b and a flame propagation travelling at the flame speed $S_f(t)$ from the ignition point can be observed after ignition. Hence, Equation (2) can be expressed as follows, assuming a constant density of the unburned gas ρ_b and a volume $\ddot{V}_c(t)$.

$$\dot{m}_c(t) = \frac{\partial^2 m_b}{\partial t^2} = \rho_b \cdot \ddot{V}_c(t) \quad (4)$$

Introducing the flame area $A_f(t)$, the volume change $\ddot{V}(t)$ can be also defined as reported in Equation (5). Besides, by considering the expansion ratio as the ratio of the density of burned gas ρ_b (at the adiabatic flame temperature) and unburned gas at ambient temperature ρ_u , S_f can be related to the effective normal burning velocity S_u .

$$\ddot{V}(t) = \frac{d}{dt}[S_f \cdot A_f] \quad (5)$$

$$S_f = \frac{\rho_u}{\rho_b} S_u \quad (6)$$

Eventually, being $\rho_u = \rho_o$, Equation 2 can be modified as follows:

$$\frac{P - P_o}{P_o} = \frac{1}{4\pi r} \cdot \frac{\gamma}{c_o^2} \cdot \frac{d}{dt}[S_u \cdot A_f] \quad (7)$$

This essential equation states that the maximum pressure of the wave generated in the combustion domain by a monopole source (the flame propagation from the ignition point to the cloud border) is generated at the flame front as the pressure is inversely proportional with r . Also, the same equation states that at any distance from the source point, the maximum pressure is directly dependent on the specific energy added (which is correlated to ρ_b through the enthalpy of combustion at standard state ΔH_c^o , as for the Hugoniot equation), and the variation of the burning velocity and the variation of the flame area provided acoustic behaviour in the far-field assumption. The burning velocity is an intrinsic characteristic of fuel-oxidant mixtures at any given temperature and pressure, which can be measured or estimated through different techniques.^[4,5] In the following paragraph, Equation (7) is declined for different flame speeds as for slow deflagration, fast deflagration, and detonation, aiming at predicting the pressure intensity at any distance from the explosion point.

2.1 | Slow deflagration

If a gas or vapour release occurs in the open—hence without any physical obstruction and at constant atmospheric pressure and temperature—the gas gradually mixes with air unless, or until, ignition. In this case, the VCE develops as a sub-sonic weak phenomenon (a deflagration), with a low, approximately constant

burning velocity, and the generated pressure (Equation (7)) is only dependent on the flame area variation with time and not on the flame acceleration per se. However, provided the presence of a homogeneous, purely unconfined fuel-air mixture, any flame is subjected to the intrinsic, hydro-dynamic instabilities as the Rayleigh–Taylor, Richtmyer–Meshkov, or the Kelvin–Helmholtz instabilities. These phenomena induce a turbulent fluid motion and accelerate the flame to values which can be multiple of the theoretical laminar burning velocity.^[6,7] Nevertheless, the flame acceleration may be only able to produce very little overpressure, which can be rarely sufficient to achieve appreciative levels of destruction in the far field. For constant (low) turbulent burning velocity S_T , it is then:

$$\frac{P - P_o}{P_o} = \frac{2r_f}{r} \cdot \frac{\gamma}{c_o^2} \cdot \left(\frac{\rho_u}{\rho_b}\right)^2 \cdot S_T^2 \quad (8)$$

According to this equation, the peak pressure measured at any distance r is now dependent on the absolute square value of the turbulent burning velocity and the dimension of the cloud r_f , which is directly correlated to the total energy produced by the combustion reaction.

This analysis first originated the idea of a critical mass needed for a destructive VCE and—in other words—to the delayed ignition criteria when event trees are defined for the VCE likelihood. Indeed, only large, homogeneous (premixed) clouds can generate high pressure if the flame propagates at a relatively low speed (slow deflagration characterized by laminar, quasi-laminar, or low turbulent burning velocity), provided that no dramatic variation of the flame area with time can be predicted even for spherical or hemispherical clouds. And, quite clearly, the formation of a large premixed large cloud of fuel air within the flammability limits takes a long time (delayed ignition) and a large amount of fuel. Here it is also worth noting that, for the analysis of VCE, further considerations are needed because the impulse (i.e., the total duration of the explosion) is also relevant. This parameter (impulse) will be discussed in the following for the definition of the critical mass, with insights for the specific case of hydrogen.

2.2 | Fast deflagration

If the release occurs in an area containing physical obstructions (process piping, equipment, buildings, cars, or even vegetation such as brushes or trees) the turbulent mixing has two-fold effects. On one side, the mixing phase of the flammable gas with air is greatly enhanced as the release flows wrap and flow over several physical

objects (the 'late' ignition time is reduced). On the other hand, turbulent phenomena are affecting the burning velocity according to the Shchelkin-like effect, also known as the positive feedback of turbulence: the combustion rate affects the flow field, which in turn affects the turbulence intensity, which in turn affects the combustion rate. In this case, the generation of the pressure waves is largely dependent on flame acceleration, and the contribution due to the variation of the flame area is typically negligible. Hence, the concept of a critical mass loses the original meaning as defined above in the case of laminar or quasi-laminar phenomena. Indeed, even little clouds can generate high overpressure at the flame front. Here it is also worth noting that these effects are not predictable by simple integral models even if several equations and methodologies have been produced to keep into account the effects of geometry.^[8–10] Besides, CFD has been largely adopted in recent years, however with some uncertainties for the lack of a full representation of the turbulent burning velocity and for the overall complexity of reproducing a fast phenomenon in a large-scale domain.

Having said that, in the case of congested domains, the explosion phenomenon shows a transition from a weak to a fast deflagration, and the overall explosion yields a dramatic increase in peak overpressure (a shock wave), typically between 0.2 and 1.0 bar.^[11] These high pressures are capable of large destruction and characterize the actual VCE, either in the near field or in the far field.

2.3 | Deflagration to detonation transition

If the flame acceleration is further enhanced by induced turbulence, a detonation phenomenon can be abruptly achieved—a deflagration to detonation transition or DDT—which is an extremely catastrophic event. According to classic studies, in the case of detonation, the flame and shock wave are coupled and travel at sonic speed (at the temperature of a burnt product) or even at supersonic speed. This is the basis of the well-known Zel'dovich–von Neumann–Döring model (ZND) for the detonation process of an explosion.^[12] Decades of experimentation suggest that detonations are possible for nearly every flammable gas that the process industry is currently using, given certain physical or thermodynamic boundary conditions such as confinement or congestion. Besides, a common misconception is that detonation must yield tremendous damage indicators due to the high pressures generated. Indeed, there will

be portions of the cloud that do not contribute to a detonation.^[13]

Originally, the study of detonations took place in obstacle-laden tubes, typically in the form of orifice plates, and parameters such as Chapman–Jouguet (CJ) velocity, critical tube diameter (d_c), and cell size (λ) were categorized.^[13,14] For an unconfined explosion, the most important parameter is however the distance from ignition to the DDT point. That distance is known as the run-up length L_d , which is of course related to the minimum diameter of the cloud, hence the critical mass.

Finally, it is worth mentioning that CFD codes have a difficult time predicting detonation, although they do indirectly allow for that. Indeed, there is no universal and/or satisfactory model for the run-up distances of gases and empirical data are only adopted.

3 | THE PREDICTION OF PRESSURE WAVE INTENSITY PRODUCED BY VCE

CFD is the best option for the VCE phenomenon, yet it has large uncertainties. In particular, it is strongly accurate in determining near-field overpressures and determining the pressure loads of targets within the flammable cloud. However, in the far field, the predictions can be difficult due to computational effort in describing very large domains, as for VCE. Nevertheless, with the advent of more powerful computers, CFD studies are becoming more prominent and several commercial or public codes are available such as FLACS by Gexcon, the ANSYS package, and the OpenFoam code, among others.

Within the industry, CFD is often too expensive if large physical domains and far-field effects are needed and—for instance, for risk assessment—it is necessary to run several cases. Hence, semi-empirical approaches are commonly adopted. The most important methodologies are the multi-energy method (MEM), which is largely adopted in Europe, and the Baker–Strehlow–Tang (BST) method, which is used mainly in the United States and other countries worldwide.^[15,16] The MEM was developed from field tests using rigs of various sizes filled with a flammable mixture and then ignited; pressure gauges were placed in various positions of the test field. The data were then tabulated against various fuels and flammable volumes and congestion (obstacle) configuration. The other cited prediction method is known as BST method. Essentially, the BST uses a flame speed table from which the calculations are carried out to determine a flame Mach number M_f . The table is based on a long history of experiments conducted at the Baker

Engineering premises and the original numerical simulations performed by Baker and Strehlow.^[17] This value of M_f is defined through the reactivity of the fuel and the obstacle congestion (Table 1).

Finally, the maximum value P_{\max} for the pressure at the flame front is defined through the following equation:

$$\frac{P_{\max} - P_o}{P_o} = \frac{2.4 \cdot M_f^2}{1 + M_f} \quad (9)$$

Similarly, the MEM identifies the maximum pressure P_{\max} by using a ‘strength factor’ F , numbered with values from 1 to 10 with increasing severity. The F value is associated with the maximum pressure at the cloud barycentre and is only dependent on the obstacle congestion. Different, separated sub-clouds can be then defined based on the level of congestion, each with separate ignition at the centre (no interactions between sub-cloud explosions are considered). Several attempts have been considered for the analytical definition of the strength factor. The F factor is however mainly based on the skill of the operator.

The BST and MEM allow the evaluation of the maximum pressure of the at the flame front with reliable details. Besides, the definition of pressure decay at any distance from the explosion source needs the solution of complex physics, which includes the fundamental equations of mechanical engineering and considers near-field effects and non-acoustic behaviour of the VCE. These effects have been conceptually translated from military science by adopting the density of energy E , which has the dimension of pressure. Indeed, at any distance r from the VCE, it is:

$$\frac{E_{f,\text{tot}}}{V} = \frac{m_f \cdot MW \cdot \Delta H_c^0}{r^3} \quad (10)$$

where m_f is the total mass of fuel participating in the gas deflagration, MW is the molecular weight of the fuel, and

TABLE 1 Flame speeds in flame Mach number M_f as for the Baker–Strehlow–Tang method for a free explosion of gas or vapour in air.

Materials	Congestion		
	Low	Medium	High
High (hydrogen)	0.36	DDT	DDT
Medium (propane)	0.11	0.44	0.50
Low (methane)	0.026	0.23	0.34

Abbreviation: DDT, deflagration to detonation transition.

ΔH_c^0 is the enthalpy of combustion at the standard state per unit mole. Eventually, in the explosion community, the dependency of pressure with distance is typically plotted over the so-called Sachs’ scaled-distance plot, which has been developed for explosives and lately introduced for VCE by BST and MEM. More specifically, Sach’s plot is a log–log diagram of pressure with the energy-scaled distance Z defined as:

$$Z = \left(\frac{E_{f,\text{tot}}}{r^3} \frac{1}{P_o} \right)^{-3} \equiv \frac{r}{\sqrt[3]{m_f \cdot \Delta H_c^0 / P_o}} \quad (11)$$

where the flammable mass m_f is typically considered at the stoichiometric concentration with air. Both BST and MEM models use the Sachs’ plot, however using multiple curves defined based on the strength factor (for MEM) or the flame Mach number (for the BST) (or in other words, the maximum pressure defined above). Similarly, the BST adopts several curves which are identified based on the flame Mach number defined above.

4 | THE CRITICAL MASS FOR THE OCCURRENCE OF HYDROGEN VCE

Personal injuries or relevant structural damages (domino effects) have a threshold value of 7 kPa, independent of the duration of the pressure wave, as demonstrated by Cozzani and Salzano.^[18–20] This value considers the escalation of the primary accident (the VCE) due to the propagation of the pressure wave in the far field (domino effects) and injuries, and hence does not take into account the business interruption or the post-accident costs needed for the return-to-service. To add more details, structural damage as the buckling of equipment shells or structures cannot be considered the actual explosion damage unless secondary accidents (fire, explosion, environmental pollution) are produced by the VCE. Eventually, whatever the total mass or the energy involved in the vapour cloud, at any distance, the deflagration of a vapour cloud with maximum pressure lower than 7 kPa can be simply defined as a flash fire. Besides, the threshold value for the pressure wave cannot be merely correlated to the mass of fuel because it may depend on the flame acceleration only, which in turn depends on the fluid-dynamic conditions, as demonstrated above.

To define a critical mass for the VCE, the Sach’s energy-scaled plot, as reported in Figure 1, can be adopted (the methodology can be similarly referred to the MEM also, even if no references are given to the flame speed in this case).

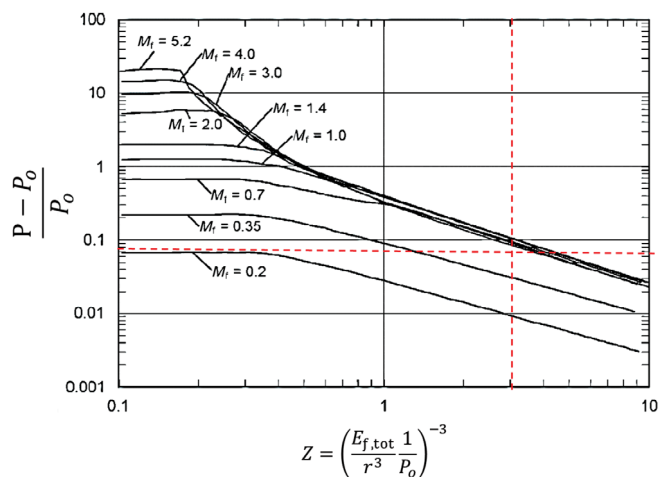


FIGURE 1 The Baker–Strehlow–Tang (BST) Sach’s scaled-energy plot for a dimensional overpressure P generated by a vapour cloud explosion (VCE) at any distance r from the explosion source with combustion energy $E_{f,\text{tot}}$. The values of M_f refer to the flame Mach number as described in the text. See Pierorazio et al. for further details on the plot.^[16]

The plot shows that the difference between a flash fire and a VCE can always be identified for a value for the flame Mach number $M_f \approx 0.2$, which produces a maximum overpressure lower than 7 kPa. Besides, for the same reason, a critical value for the scaled distance $Z_c = 3.0$ for the VCE occurrence can be recognized whatever the total burning mass (i.e., the total energy), the concentration (the volume), the distance from the explosion source, and, more important, independently of the flame acceleration (slow deflagration, fast deflagration, detonation).

Based on these considerations, Figure 2 shows the amount of pure hydrogen which is needed to produce a vapour cloud at stoichiometric concentration generating a minimum pressure wave of 7 kPa at any distance from the release point, calculated by considering a Sach’s scaled distance of $Z_c = 3.0$, obtained by simple calculation through Equation 11 ($\Delta H_c = 241.2$ kJ/mol). On the same figure, the corresponding volume of compressed hydrogen gas (CH₂) at 300 bar and 700 bar (typical values of commercial CH₂ storage) is shown, as calculated through the ideal gas law. The plot is also valid for liquid hydrogen (LH₂) if considering an equivalent mass evaporated from the liquid pool.

So far, a crucial point is the definition itself of the VCE, which includes the definition of ‘far-field’. We can assume that a VCE is defined only if the pressure wave is larger than 7 kPa at a distance from the cloud border which is equivalent to the distance travelled by the sound speed (pressure wave velocity) in the same time range of the explosion duration. This concept could be addressed in the case of vapour clouds formed from hydrocarbon pools or cryogenic fuel release, in particular liquid

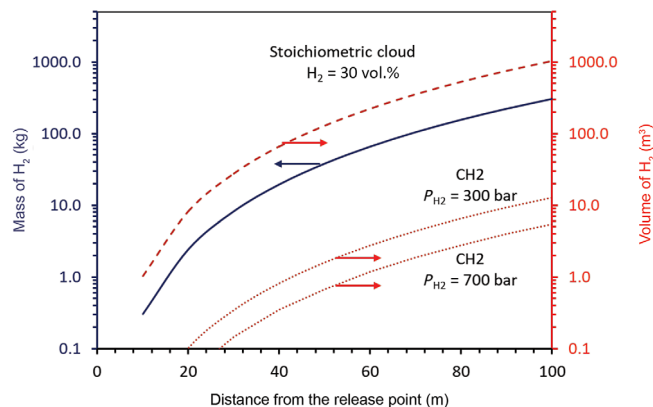


FIGURE 2 The amount of pure hydrogen which is needed to produce a VCE with a pressure of 7 kPa at any distance from the explosion source, provided a Sach’s scaled distance $Z = 3.0$. The dotted line is the cloud volume (spherical) at the stoichiometric concentration (30 vol.%). The volume of compressed hydrogen gas at 300 bar and 700 bar needed for the cloud formation is also included.

hydrogen. Another option refers to the maximum jet length produced by the gas release through any failure or cracks on the containment system (pipeline, tank) in the case of pressurized gases. In this case, the distance depends on the internal pressure or the flow diameter. This length is however affected by buoyance effects, which is typically prevailing on momentum effects at a distance of ~ 20 m even for highly pressurized gases such as compressed hydrogen at 300 or 700 bar.^[21,22] A similar threshold distance can be also considered for the blast wave from CH₂ tank rupture, which is lower than 20 m according to the literature.^[23] Eventually, the value of 20 m can be considered as the threshold limiting value for the VCE occurrence and the critical mass is defined from the plot reported in the previous figure or from the definition of scaled distance:

$$m_f^{\text{crit}} = \frac{r^3 \cdot P_0}{Z^3 \cdot MW \cdot \Delta H_c^0} \quad (12)$$

Eventually, for hydrogen deflagrations or detonations, the critical mass for the VCE of hydrogen can be calculated by Equation (12) approximately as $m_f^{\text{crit}} = 4.0$ kg. This value corresponds to a homogeneous stoichiometric cloud (30% v of hydrogen in air) with a volume larger than 10 m³ (after dispersion), or to the entire content of a 200 lt of a bottle of compressed hydrogen. Quite clearly, these last two assumptions are conservative if considering the very low density of hydrogen at ambient temperature and allow to consider, for example, the VCE from hydrogen car storage as unlikely. Indeed, the high buoyancy of hydrogen affects its dispersion more than its high

diffusivity in a closed, partially confined, or open environment.^[24,25] On the contrary, it is quite consistent in the case of liquid hydrogen, when the ultra-low temperature changes the dispersion phenomenon as the vapour can be heavier than air at a small distance from the liquid pool.

5 | CONCLUSION

The hazard of a VCE of hydrogen released into the open atmosphere and in partially confined geometries is dramatically reduced by buoyancy and by the high probability of early ignition. Nevertheless, due to high reactivity, even a small cloud can generate high pressure and destructive pressure waves in the far field.

In this work, two critical values for the critical Sach's energy-scaled distance and the total mass participating in the VCE have been defined, respectively. For the first parameter, a general value of $Z_c = 3.0$ can be considered as the reference point for the distinction between flash fire and VCE. Besides, a critical mass m_f^{crit} of about 4.0 kg for the hydrogen cloud to behave as a VCE has been defined, based on the fundamental theory of combustion and assuming a minimum distance of 20 m for the far-field definition (lower values are within the range of jet fire). Any non-stoichiometric hydrogen concentration in air or lower amount of hydrogen would decrease either the flame Mach number M_f (lower the critical value of 0.2), or the total energy, thus resulting in an energy-scaled distance which is lower than Z_c of 3.0. In this sense, the effects of buoyancy, diffusivity, and weather conditions on the dispersion of hydrogen should be taken into account.

AUTHOR CONTRIBUTIONS

Ernesto Salzano: Conceptualization; data curation; formal analysis; funding acquisition; investigation; methodology; project administration; resources; software; supervision; validation; visualization; writing – original draft; writing – review and editing.

PEER REVIEW

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DATA AVAILABILITY STATEMENT

Data sharing not applicable to this article as no datasets were generated or analyzed during the current study.

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