

CFD SIMULATION OF HYDROGEN RELEASE, DISPERSION AND AUTO-IGNITION IN ENCLOSURES

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Abstract

CFD simulations of auto-ignition of hot hydrogen flowing into a cylindrical enclosure where ambient air prevails were conducted. One-step global reaction was incorporated into ANSYS® software to simulate the chemical reaction between hydrogen and oxygen.

Auto ignition was obtained for different inflow temperatures, agreeing with classical results of h_2 - O_2 explosion limits, referred to as the "peninsula".

Temperature range of the reaction zone lies below the value of the adiabatic flame temperature of a constant pressure process. This is due to the fact that although the enclosure is of constant volume, ignition takes place in a negligible fraction of the total volume; therefore the absolute pressure remains unchanged.

Ignition delay decreases with the increase of the hydrogen inflow temperature. Moreover, their values are similar theoretical predictions.

Introduction

The breach of a flammable jet into an enclosure is one of several accidental scenarios that are examined associated with mobilization and storage of hydrogen. Hydrogen, which is studied extensively as fuel for various size vehicles, from micro-combustors to "regular" engines, is a highly reactive gas, therefore poses major fire hazards. Also, long-term exposure of containment materials to hydrogen causes a phenomenon known as hydrogen embrittlement, which may result in cracking and hydrogen leakage.

As an energy carrier, hydrogen has some unique properties compared to other combustible gases. Because of its low density, storage of hydrogen at pressures as high as 100MPa is being considered for transportation applications. The low viscosity and small molecular size of hydrogen give it a greater propensity to leak than other common gaseous fuels. Hydrogen is also more diffusive and more buoyant which mean that hydrogen can disperse more easily and rapidly than other fuels after an accidental release. However this also produces a detrimental effect of enabling deflagration and diffusive combustion of hydrogen to proceed rapidly in ultra-lean mixtures. Hydrogen has a much wider range of flammability in air (4% to 75% by volume) and the minimum ignition energy, 0.017mJ [1], is about an order of magnitude lower than that of hydrocarbon-air mixtures. These two properties make hydrogen easier to be ignited.

Many studies research mixing of hydrogen jet, dispersing into enclosures filled with air, however fewer focused on it's the transient behavior at short times. Fewer studied self ignition, and mainly focused on high pressure hydrogen [2][3] numerically, and experimentally [5][4].

The present study focuses on the fundamental features of transient hydrogen dispersion, and examines the flammability limits of non-premixed hydrogen-air mixtures with no external ignition energy, in different cases.

Different thermodynamical cases will be examined in simple geometry.

Physical and Numerical Model of the Flow Field

Spontaneous ignition is studied in a simple geometry of a vertical cylinder diameter, through which hydrogen bursts from the bottom.

Figure 1 presents a schematic description of the enclosure with its relevant physical features, dimensions and grid. The hydrogen leaks through a 10mm aperture and flows upwards due to both its initial momentum and is buoyant nature. This specific geometry was chosen in order to be able to compare the dispersion results with literature. Hydrogen flows through the 10mm aperture by 0.5m/s (see **Figure 1**), thus for atmospheric conditions, Reynolds number is close to 50 (equation (1)), namely laminar flow regime.

$$Re = \frac{\rho \bar{V} d}{\mu} = 50 \quad (1)$$

$$\rho_{H_2} = \frac{p}{R T_0} = 0.0812 \frac{kg}{m^3} \quad (2)$$

Where ρ , R and μ are the density, gas constant and dynamic viscosity of the flowing gas, namely the hydrogen. \bar{V} is the hydrogen velocity at the aperture, d is the aperture's diameter and p and T are its pressure and temperature.

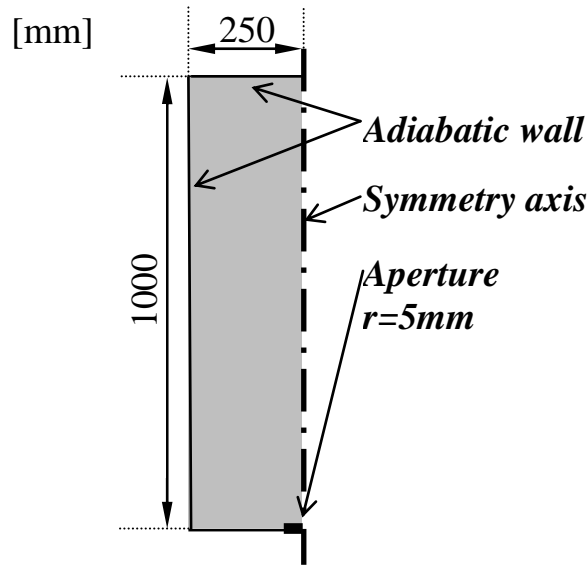


Figure 1. Schematic description of the enclosure.

The numerical approach is based on the solution of transient two-dimensional conservation equations. ANSYS-FLUENT®, version 12.1 is used as the platform.

The numerical model is axisymmetric. Adiabatic, no-penetration and no-slip conditions were assumed for the walls. The discharge orifice, denoted as "Aperture" in **Figure 1**, was modelled as "velocity inlet".

Different grid structures were built; each consisted of cells aligned with the direction of flow, as recommended by ANSYS-FLUENT®. Also recommended is at least 5 cells for each zone, thus this division was set as minimal at the aperture, as indicated in Table 1. Next, radial and axial distributions were set so that the aspect ratio of the cells will remain low near the walls. These values were adopted after a careful grid refinement procedure.

Table 1 – Grid details

Total no. of cells	Axial distribution		Radial distribution**	
	No.	distribution	Aperture	Radius
11625	75	Bi-exponential with $x=1^*$	5	150
21000	100		10	200
31500	150		10	200

* See GAMBIT® User's Guide for details

** Uniform radial distribution

Grid independency was checked, and it was found that a further increase in the grid density had a negligible effect on both the velocity and species fields (less than 0.1%), as demonstrated for two radial positions, axis and aperture's edge, $r=5\text{mm}$ (see **Figure 2**), for the hydrogen mole fraction. Similar results were received for velocity magnitude. In order to facilitate good simulation of the chemical reaction between the hydrogen and the oxygen within the air, the intermediate grid of 21000 cells was chosen.

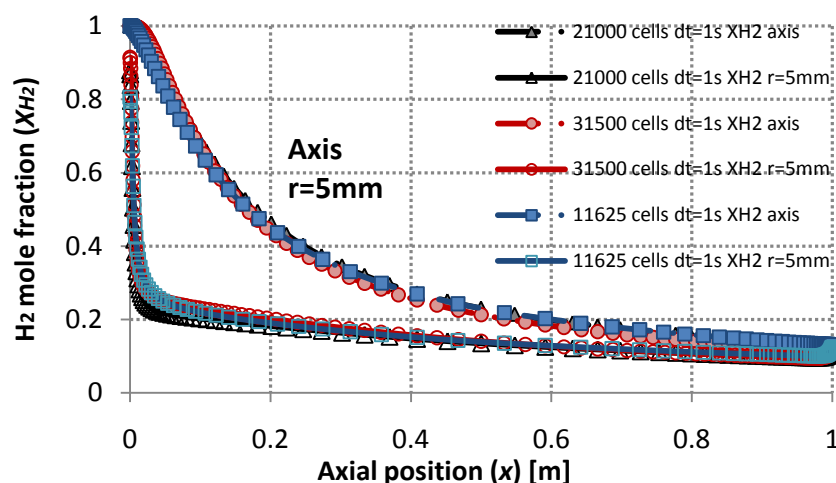


Figure 2. Grid sensitivity analysis – no chemical reactions are modelled.

The model sensitivity to the time step was checked, as well. It was found that the difference in results between time steps of 0.1s and 1s was impossible to notice ($<0.01\%$). Therefore, for dispersion simulations, a time step of 1s was used for validation purpose, as seen in the next section.

Model sensitivity was checked also for auto-ignition simulations at the time of auto-ignition. It was found that regardless of the number of cells, auto-ignition was obtained after 1.5ms for the case of hydrogen at a temperature of 850K flowing into the cylindrical enclosure in which ambient air prevails. **Figure 3** presents the centerline (axis) hydrogen mole fraction (X_{H_2}) at auto-ignition times. It seems there is a minor difference in hydrogen mole-fraction profile at the axis for the two denser grids (21000 cells and 31500 cells). However, for the same numerical parameters (under-relaxation factors, number of iterations etc. the more spacious grid was not well converged. The same was found the other parameters as velocity magnitude. Hence, for the auto-ignition simulations the grid with 21000 cells was used.

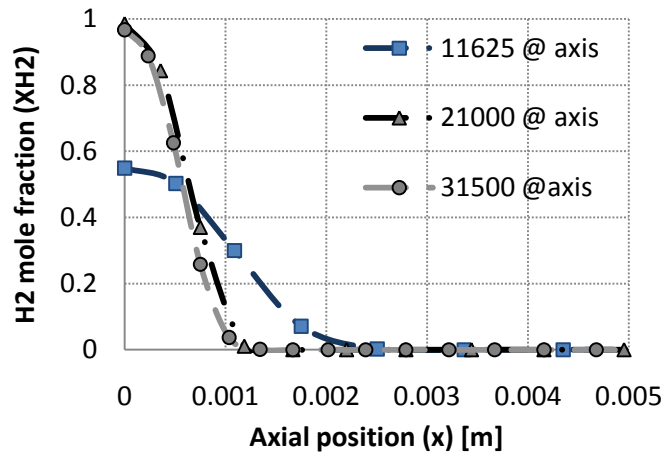


Figure 3. Grid sensitivity analysis – global *Hydrogen-Air* reaction is modelled; comparison of centreline H_2 mole fraction; results for auto-ignition at time step of 0.02ms.

Validation

Validation of the flow field was conducted by comparing dispersion results with recent results of Vudumu and Koylu [6].

Figure 4 presents comparison between published results and the current simulation results. Although some difference may be noted, it is attributed mainly to the quality and reproducibility of the color maps. However, since no XY plots were provided for the case of hydrogen leak at the bottom of the cylinder, and despite this shortcoming of the color maps, they were used for comparison. One may see that hydrogen concentrates at the top of the cylinder due to both buoyancy and initial momentum, and as time advances, so does the hydrogen, towards the bottom.

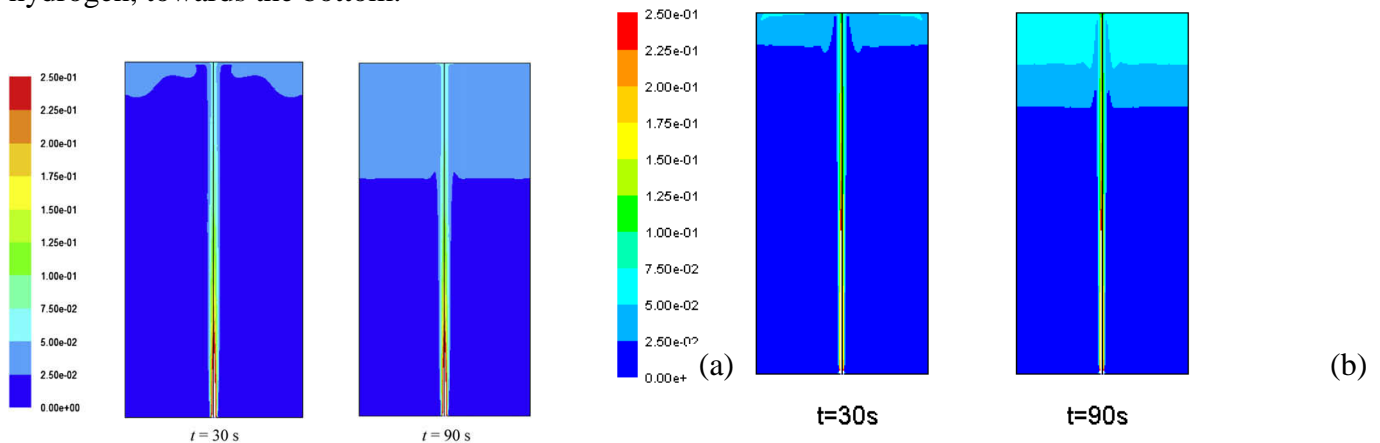


Figure 4. Dispersion map at t=30s, 90s – comparison between (a) published results [6] and (b) current simulation

Results and Discussion

Detailed *Hydrogen-Air* reaction mechanism consists of about 20 elementary reactions [7], however simplified system of reactions, consisting either 4 or 5 reactions involving hydrogen and oxygen may be used. While detailed reaction mechanisms provide insight into flame structure and reactivity of gas mixtures, there is a great need for one-step global kinetics for use in complex fluid mechanics codes. In the current simulation a single step reaction (equation (3)) with a global reaction rate (equation (4)) was used [7]:



(4)

Where, \bar{R} is in J/kg·K, T is in K and k is in cc/mol·s

Auto Ignition

Explosion limits are the pressure-temperature-mole-fraction boundaries for a specific mixture that separate the regions of slow and fast reaction [8]. **Figure 5** shows the explosion limits of a stoichiometric mixture of hydrogen and oxygen.

Other well known explosion limits are given as the Low / Upper Flammability Limit (LFL / UFL) diagrams at different temperature and pressures. Some are brought here in Table 2.

Table 2 – Hydrogen flammability limits at various temperatures and pressures; DIN51649-1[9]

T [°C]	20			100	200	300	400
p [atm]	1	10	100	1	1	1	1
LFL*	4.1	4.9	5.8	3.4	2.9	2.0	1.4
UPL*	75.6	72.4	74.1	77.6	81.3	83.9	87.6

* By volume fraction

Theoretically, the definition of an ignition event is somewhat arbitrary. In the present work, we have adopted Kumar's approach [10]. Thus, we have defined an auto-ignition event as the instant at which the local temperature of the mixture exceeds the temperature of the hot hydrogen that enters the enclosure through the aperture, and continues to rise.

Auto-ignition was examined for a scenario of a hot hydrogen flow enters a cylindrical enclosure in which ambient air prevails. For a stoichiometric mixture, auto-ignition should occur at temperature of 850K and above, as seen on **Figure 5**.

In the present work, five cases were simulated at atmospheric pressure, and two of them are indicated as circles on **Figure 5**. Solid circle represent hydrogen inflow temperature of 850K in which ignition occurred, while the outlined circle represent hydrogen inflow of 800K with no indication of ignition. It may be seen that the simulation results agree with the literature.

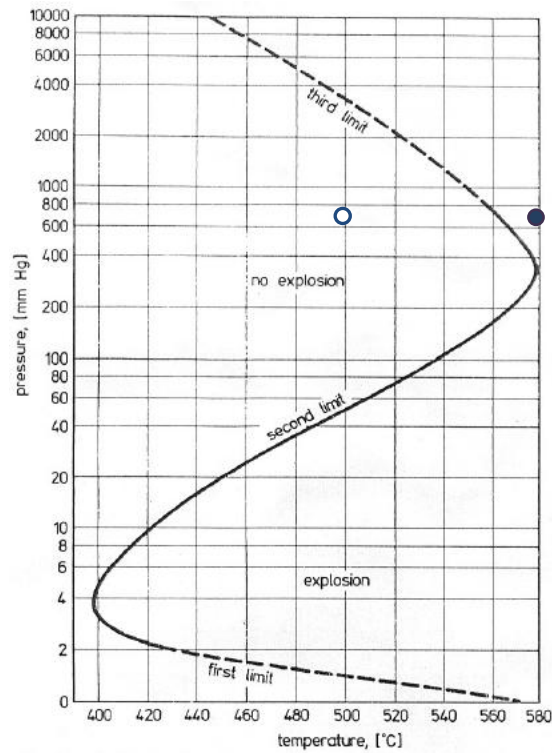


Figure 5. Explosion limits of a stoichiometric *HCl* Coated vessel of 7.4cm diameter (after Lewis & von Elbe [1]).

Hot hydrogen flows into ambient air, where it is mixed. As it flows into the closed cylinder, it displaces the air due to both its initial momentum and its natural buoyancy. This is manifested through Richardson number, which represents the ratio between buoyancy and kinetic energy. In the present case, the value of Richardson number exceeds unity (see equation (5)), therefore buoyancy effects are dominant.

$$Ri = \frac{(\rho_{Air} - \rho_{H_2}) g d}{\rho_{H_2} \bar{V}^2} = 5.3 \quad (5)$$

Ignition is obtained within 1÷1.5ms, thus the characteristic length due to thermal diffusion alone is estimated as 0.5mm (see equation (6)).

$$L \approx \sqrt{\alpha \cdot t} \approx 0.5 \text{ mm} \quad (6)$$

$$\alpha_{H_2} = \frac{k}{\rho C} = 1.4 \cdot 10^{-4} \frac{m^2}{s} \quad (7)$$

Where α is the hydrogen's thermal diffusivity, k , ρ and C are its thermal conductivity, density and specific heat constant.

Figure 6 presents temperature contours in the vicinity of the aperture. It may be seen that the thermal effects reach 1.5mm in the axial direction and 6mm in the radial direction. Air is heated due to three mechanisms listed below, thus the heated region is larger than that predicted solely by thermal diffusion:

- Thermal diffusion.
- Thermal convection and buoyancy effects (see Ri number, equation (5)).
- Heat release due to chemical reactions.

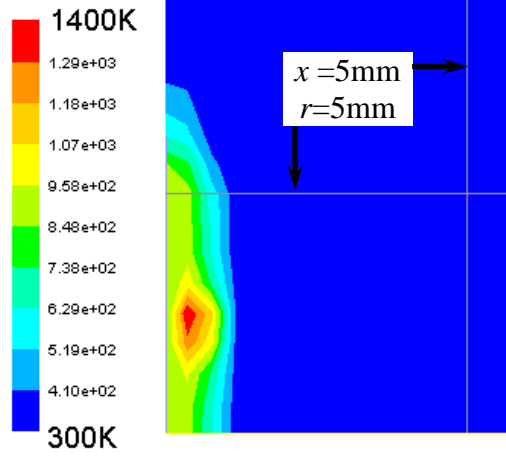


Figure 6. Temperature contours; $T_{H_2}=850K$, $T_{Air}=300K$.

The stoichiometric fuel to oxidizer (FOR_{st} , equation (8)) of the hydrogen-oxygen reaction (equation (3)) is 0.67. The volumetric value of fuel-to-oxidizer ratio (FOR_{vol}) equals its molar one, assuming the mixture is characterised by the same temperature and pressure (T , p , equation (9)).

$$FOR_{st} = \frac{n_{H_2}}{n_{H_2} + n_{O_2}} = 0.67 \quad (8)$$

$$FOR_{vol} = \frac{V_{H_2}}{V_{H_2} + V_{O_2}} = \frac{n_{H_2}}{n_{H_2} + n_{O_2}} \quad (9)$$

$$FOR_{vol} = FOR$$

Where n is number of moles and M is the molar mass.

Figure 7 presents the volumetric fuel-to-oxygen ratio for the same case that is presented on **Figure 6**. The adiabatic flame temperature varies from 2200K to 3400K [11] for a constant pressure and volume processes accordingly. Although the present work concerns with enclosures of constant volume, the ignition process itself may be assumed to be a constant pressure process. In the present case, ignition occurs within a negligible volume from the total enclosure's volume (the volume fraction that is occupied by the products, is about 10^{-7}), and the absolute pressure within the entire enclosure is practically unchanged. Moreover, since it is the onset of the chemical reaction, the combustion process is not yet completed, therefore is not an ideal adiabatic one. Hence, the reaction temperature at the onset of ignition (**Figure 6**) agrees well both in the maximal value ($T_{max}=1400K$) and in its place of occurrence. It may be seen that the temperature decreases as the FOR decreases (**Figure 7**), towards the internal volume. The temperature also decreases towards the aperture. This effect is attributed to the relatively low inflow temperature (850K).

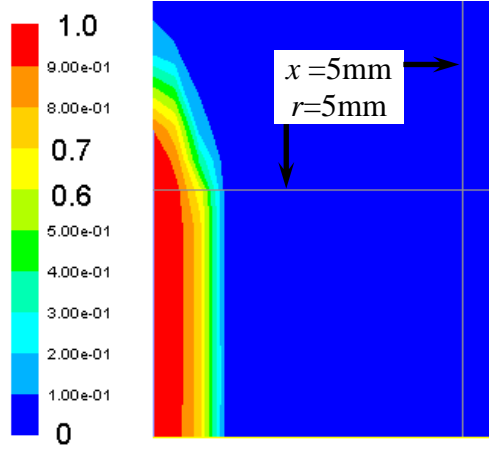


Figure 7. Contours of hydrogen to oxygen ratio (*FOR*); $T_{H_2}=850K$, $T_{Air}=300K$.

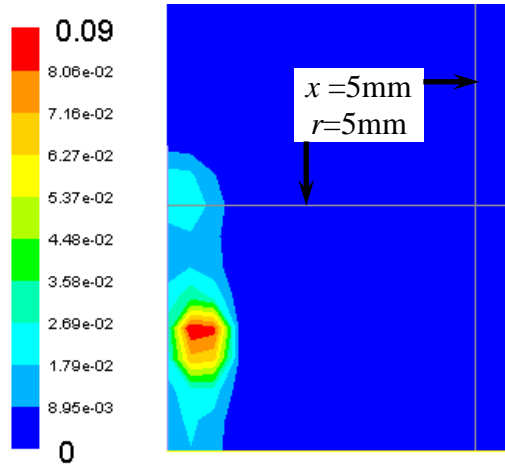


Figure 8. Contours of water; $T_{H_2}=850K$, $T_{Air}=300K$.

Ignition delay

Ignition delay is the time elapsed from the moment the hot hydrogen entered the cylindrical enclosure till the time at which the local temperature of the mixture exceeds the inflow temperature. It may be evaluated by the following expression, assuming negligible reactant consumption during the ignition delay (t_i) [11]:

$$t_i \equiv \rho_{mix} C \frac{R T_0^2}{E} \frac{\exp\left(\frac{E}{R T_0}\right)}{\Delta H_c k_n [F_0]^n} \quad (10)$$

Where ρ is the density of the mixture, C is its specific heat, \bar{R} is the universal gas constant and T_0 is the initial temperature of the mixture. E is the fuel's activation energy, ΔH_c is its heating value, k_n is the pre-exponent factor of the global reaction rate, $[F_0]$ is the fuel's concentration and n is the reaction order. Thus, for the current reaction, at atmospheric pressure these assume the following values:

$$\rho_{mix} = \frac{p}{R T_0} = 0.41 \frac{kg}{m^3} \quad (11)$$

$$[F_0] = FOR \cdot \rho = 0.019 \frac{kg_{H_2}}{m^3} \quad (12)$$

Thus, for stoichiometric mixture at atmospheric pressure and at 850K, ignition delay is estimated by 1.1ms.

Figure 9 presents ignition delay times for a stoichiometric *Air-Hydrogen* mixture at different temperatures and pressures. The dashed line represents a mixture at 2atm taken from [7]. The solid line represents results from equation (14) for atmospheric pressure and the solid circles represent the current simulations results. Higher pressure induce longer ignition times, hence the higher results of dashed line at 2 atm., as compared with the solid one at 1 atm. It seems that ignition delay times obtained from CFD simulations indeed decrease with the increase of temperature. Also, values are at same order of magnitude for temperature range of 850÷1000K.

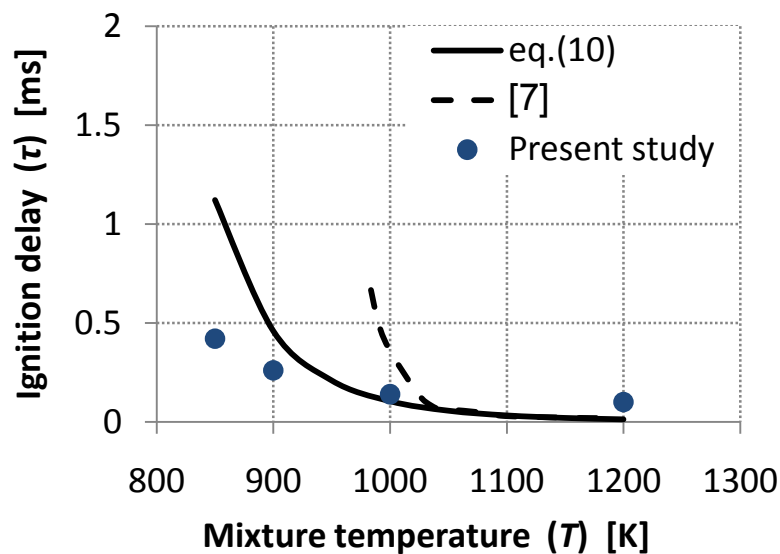


Figure 9. Ignition delay times of an *Air-Hydrogen* mixtures for a stoichiometric mixture at 2atm [7] (dashed line), at 1atm according to equation (10) (solid line) and at 1atm according to simulation results (solid circles)

Conclusions

CFD simulations of release and dispersion of hot hydrogen into a cylindrical enclosure with an ambient air were conducted. The hydrogen flow regime is laminar. Often, these kinds of simulations are conducted in order to obtain dispersion maps, from which flammability zones are detected. These zones are hazardous, since hydrogen is a highly flammable gas.

The present work focuses on the transient behavior of hot hydrogen that is released to an enclosure at short times.

Computational evidence of the time of auto-ignition was observed, which could be used to "translate" dispersion maps into "auto-ignition maps".

Auto-ignition event was defined as the instant at which the local temperature of the mixture exceeds the temperature of the hot hydrogen that enters the enclosure through the aperture, and continues to rise.

Ignition was obtained by incorporating a one-step global reaction between hydrogen and oxygen and the appropriate reaction rate into ANSYS® software. Auto ignition was obtained

for different inflow temperatures, agreeing with classical results of h_2-o_2 explosion limits, referred to as the "peninsula".

Hot hydrogen flows into ambient air, where it mixes. As it flows into the closed cylinder, it both displaces the prevailing air and heats it. Air moves due to both hydrogen's initial momentum and its high tendency to buoyant ($Ri = 5$). It is shown that during ignition delay time, a region at the order of the characteristic diffusion length is being heated. Yet, since air is heated as a result of three mechanisms, the heated region is larger. The heat transfer mechanisms are thermal diffusion and convection, and heat release following the exothermic chemical reaction.

Flowing into the enclosure, an inhomogeneous mixture of hydrogen and oxygen is being formed. Auto-ignition occurs where the hydrogen to oxygen fraction (FOR) and high temperature combines to create the perfect conditions for auto-ignition. Therefore the maximal temperature is obtained in the center of the reaction region. In that region, hydrogen to oxygen fraction is closest to stoichiometric, and the temperature is the highest.

After ignition, temperature is still highest at the "center region", however decreases due to both mixing with the prevailing air and with the relatively cold hydrogen inflow.

Ignition delay times show decrease with the increase of the hydrogen inflow temperature. Moreover, the values of simulated results are similar to the theoretical predictions.

Further simulations are needed to obtain the ability to produce "auto-ignition maps" in a wide range of pressures and temperatures.

Nomenclature

C	specific heat
d	aperture's diameter
E	activation energy
$[Fo]$	fuel's concentration
FOR	fuel-oxygen reaction
g	gravitational acceleration
Hc	heating value
k	thermal conductivity
k_{global}	global reaction rate
k_n	pre-exponent factor
L	characteristic length
M	molar mass
n	number of mole
p	pressure
R	gas constant
\bar{R}	universal gas constant
Re	Reynolds number
Ri	Richardson number
T	temperature
t	time
V	gas volume
\bar{V}	Velocity
α	thermal diffusivity
Δ	difference
μ	dynamic viscosity
ρ	density

Subscripts

H_2	hydrogen
O_2	oxygen
<i>mix</i>	mixture
<i>st</i>	Stoichiometry
<i>vol</i>	volumetric

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