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Explosibility Of Hydrogen-Air Mixtures In Large Spherical Vessel

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***Abstract:** Explosion of hydrogen-air mixtures has been investigated in a 3.66-m diameter spherical vessel over a range of hydrogen concentration (5-40 % by volume) in air at 25^{0}C and atmospheric pressure with two positions of ignition source -central and at 0.3 m from the bottom of sphere. The effects of fan- induced turbulence were investigated qualitatively. Explosion-pressures were measured. Bottom ignition resulted in faster and more complete combustion than central ignition. Turbulence affected combustion significantly. At stoichiometric hydrogen concentration, combustion was complete in 0.1 s without turbulence and in about 0.07 s with turbulence. Data reported here adds to the information on hydrogen combustion at large scale and may be used in devising safety measures for hydrogen handling installations.*

***Keywords:** explosion, hydrogen, explosion pressure, large vessel*

I. INTRODUCTION

Hydrogen is produced in various industries e.g. ammonia manufacturing and refineries. This gas is used in fuel cells, food, chemical processing, pharmaceuticals, aerospace, electronics, petroleum recovery and refinery, power generation and metal production. Hydrogen is the most promising fuel for global use in future as it is an energy-efficient, low polluting and renewable fuel. Introduction of hydrogen as an energy source makes great demands on all aspects of safety of hydrogen installations. On release in atmosphere hydrogen disperses upward rapidly due to buoyancy. It does not spread horizontally very far before the concentration decreases below the lower limit. Hydrogen hazard for gas leak in open is therefore lower. However, the leakage of hydrogen in confined spaces/ enclosures has very high risk of explosion in presence of ignition source as this gas has a wide flammability limit (4-75 %) and low ignition energy (Frank,1996). High burning velocity of hydrogen-air mixture generates violent deflagration (pressure~7-8 bara in confined space) and a transition to detonation (pressure upto 20 bara) under favourable circumstances.

Also interest in hydrogen combustion/explosion research is due to its relevance to nuclear reactor containments. During an anticipated coolant-loss accident in a light-water nuclear reactor, hydrogen is produced in the reactor core by the reaction between zirconium metal cladding and steam in degraded core. The hydrogen and considerable amounts of steam may be released into various containment compartments where they can mix with pre-existing air in quantities sufficient to sustain a combustion wave propagating through the mixture, should ignition occur. The possibility of initiation of explosion and detonation wave in this combustible mixture could pose a threat to the integrity of the containment building. Study of combustion/explosion of hydrogen-air mixtures is important for assessing the behaviour of reactor containment systems during such accidents. Although most postulated accidents scenarios suggest that the mean hydrogen concentration in the containment building will be low, it is possible that regions of high concentrations may occur; therefore, an understanding of combustion at low as well as at high hydrogen concentrations is required for analysis of containment behaviour. The concentration of hydrogen may be such that a deflagration wave, once initiated, will propagate through the mixture.

Evaluation of explosion risk associated with reactor due to hydrogen leak requires information on rate and extent of combustion/explosion as a function of composition, ignition source location and turbulence. Combustion of hydrogen-air systems has been studied extensively to provide burning velocity measurements (Liu and MacFarlane, 1983). There are number of studies on hydrogen-air combustion/explosion reported in different small scale sizes of vessels (Barknecht, 1981- 5 L sphere; Cashdollar et al., 2000- 120 L Sphere; Holtappels, 2002-6 L Semi sphere ,14 L Sphere,40 L Sphere; Jo & Crowl, 2010-20L Sphere; Ma et al., 2014-20 L sphere; NFPA 68, 2007-5L Sphere; Salzano et al. 2012-5 L Sphere; Senecal and Beaulieu, 1998-22 L Cylinder; Tang et al. 2009-5.3 L cylinder). Liu et al. (1980) carried out experiments on the combustion of hydrogen-steam-air mixtures in 2-litre vessel for steam concentrations upto 15 % and hydrogen upto 10 % by volume. To predict hydrogen combustion behaviour in large volumes with confidence, it is necessary to perform experiments at large scale. There exist a few studies for large scale hydrogen explosion. Furno et al. (1971) investigated hydrogen-air explosion in a 3.66 m diameter sphere under quiescent conditions using central ignition. The experiments were concerned with limit flames of hydrogen in air. They observed that extent of combustion and ultimate pressure-rise was very low with upward flame propagation for concentrations of hydrogen between 4 to 8 % air. At 8 % hydrogen in air the combustion level and pressure rise were reasonable. Hertzberg (1981) and Hertzberg and Cashdollar (1983) obtained similar results in 8-L vessel and investigated the effect of initial turbulence. Buoyancy effects, were dominated at 4-8 % hydrogen under quiescent conditions and limited extent of propagation and pressure rises. These were suppressed under turbulent conditions. Turbulent propagation in that composition range generated much higher pressure-rises than laminar propagation. The small scale experiments indicate the general effects of turbulence on explosion. However, the characteristic scales in such studies are different from those expected in nuclear reactor containment. Hydrogen – air - steam mixtures combustion / explosion experiments at large scale have also been done using 2.3-m diameter vessel to study the above effects in detail (Kumar, Tamm and Harrison, 1983,1983b) for concentrations 4-42 % hydrogen and 0-30 steam by volume, effect of fan-induced turbulence, presence of obstacles and ignition source location on hydrogen-air combustion. These two appear to be only large-scale hydrogen-steam-air combustion work reported so far.

The CSIR-CBRI has a 3.66- m diameter spherical vessel for this type of research**.** Experiments were undertaken in this vessel to improve the understanding of combustion of hydrogen-air mixtures in the concentration range 5-40 % of hydrogen in air. Research will provide data on explosion pressure based on actual measurements in this large vessel to serve as a more reliable and scientific input to the design of explosion safety measures. The study covers comprehensive information on maximum explosion pressure of hydrogen-air mixture and to examine the effect of hydrogen concentration, turbulence and location of ignition source on this parameter. A wider knowledge of the explosion severity characteristics of hydrogen will be an important contribution to the development of codes, standards and regulations related to hydrogen safety.

II. EXPERIMENTAL FACILITY

Fig. 1 shows the experimental facility -3.66 m diameter Spherical Steel Vessel used in this research. There are seven access ports for installation of various systems such as vacuum creation, gas or vapour introduction, circulation, ignition, pressure measurement and a door. Three of these access ports are 6´´ diameter nozzles located 90^o apart from each other on the equator of the sphere; the door opening is centered at this level also. In addition, a 6´´ nozzle is located at the bottom and a 12´´ nozzle at the top. The remaining two ports are 1-inch diameter pipes situated in the lower portion of the vessel. To facilitate operations within the sphere, the chamber is equipped with a removable floor of steel grating. The vessel is fabricated of firebox quality steel and is designed for a static pressure of 20 kg/cm^2. This was stress-relieved and subjected to a hydrostatic test (30 kg/cm^2) and a vacuum test (450 micron of mercury) for 30 minutes. Under transient conditions of explosions the vessel may be expected to withstand pressures even greater than 30 kg/cm^2 depending upon duration of the pressure pulse. However, as a general safe practice, the design pressure should not be exceeded.

Two fans driven by variable-speed air motors were mounted in the sphere diametrically opposite each other and used for producing turbulence. Transient pressures during explosion were measured by two piezoelectric-type transducers mounted flush with the inner surface of the vessel flanges. The gases in the sphere were analyzed before and after combustion using gas chromatograph.

The ignition system installed in the Vessel has two brass electrodes (3 mm diameter) connected to a circuit which produces an electrical spark. The spark generating system has three units: variable capacitor bank having capacitors with different values; variable voltage supply/ continuous spark unit; and step-up transformer. The spark is triggered by high voltage transformer using two-electrode system connected to the secondary winding of the transformer and fitted in the Vessel. The self-inductance of the secondary coil of the trigger transformer is 1 mH. The spacing between the electrodes is 6 mm. The spark may be continuous or of known energy. With variable combination of capacitance and voltage, it is possible to obtain sparks with ignition energies in the range 0.5 mJ – 3.2 J using this circuit. There is precise electronic synchronization between gas circulation and spark onset. Energy discharged from the capacitor is calculated from the following formula, assuming no energy losses in the transformer,

$$W = \frac{1}{2}C(V_i^2 - V_f^2) \quad (1)$$

where,

W - Discharge energy, J

C - Total capacitance of discharge circuit, F

V_i - Initial voltage of charged capacitor, V

V_f - Final voltage of charged capacitor, V

A spark energy measurement system has been integrated in the spark generation circuit and net spark energies generated for various combinations of capacitance and voltages are determined in the conventional way by measuring current and voltage across spark gap as a function of time and integrating the power-verses-time curve. Measured spark

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energies were typically 90-95 % of the theoretical energies computed using equation 1.

At the start of experiment, the gas manifold and the sphere is evacuated to 0.02 bar using a vacuum pump. Next, hydrogen and air are added at partial pressures required to give the desired mixture composition. Gas-air mixture was then circulated through a 6´´ diameter sidearm gas circulation system, until gas samples from the top and bottom gave identical fuel concentration within 0.1%, to ensure homogeneous mixtures. Samples of test mixtures were taken prior to ignition and were analyzed by a gas chromatograph. For the present experiments, ignition was done with a 0.6 cm gap continuous spark from the discharge of a 10 kV transformer. Spark duration was held constant, approximately at 0.2 second. Dynamic pressure during explosion was measured using piezoelectric and strain gauge pressure transducers provided at two ports. Pressure-time curve was recorded by storage oscilloscope or high speed chart recorder. The electric pulse generated at the moment of spark firing is used to trigger the digital storage oscilloscope that monitors and stores both the pressure verses time signal and triggering signal. Experiments for explosion violence measurement were conducted over a wide range of gas concentration. Each experiment was repeated thrice. The volume concentration of hydrogen was ranging from 5 to 40 %.

Fig.1. Large- 3.66-m diameter Spherical Vessel for explosion research studies

Figure 1

III. RESULTS AND DISCUSSIONS

All experiments were conducted at 25°C and 1 atm. The mixture was ignited by a single-capacitance spark. Two igniter positions were available at bottom and centre of the sphere, but only one igniter was used in any experiment. Fig. 2 shows explosion pressure-time curves for hydrogen-air mixtures near lower explosible limit of hydrogen (5,7,8,8.5 % hydrogen in air) after reproduction of measured data, with ignition at 0.30 m from the bottom of sphere. The pressure peak for 8.5 % hydrogen is much higher than with 5 % hydrogen, as expected. During explosion of hydrogen at low concentrations (~ 5 vol. %) burning velocities are low and combustion is incomplete and dominated by buoyancy effects. It is assumed that fireball starts near the ignition point at the bottom of sphere, moves upward at a speed greater than the burning velocity of the mixture, and downward propagation does not occur (also observed by Furno et al. (1971)). The fireball sweeps a conical volume, and the burnt amount is a function of initial hydrogen concentration. A small fraction is burned at lower hydrogen concentration. Higher hydrogen concentrations show larger fractions of hydrogen burned, and higher peak pressure. As the fireball reaches the sphere top, it is quenched by heat transfer to the walls, and the pressure in the system decays. At hydrogen concentration ≤ 8% the pressure rises were very small since little of the combustible mixture was consumed by upward flame propagation. With the onset of downward propagation at 8.5 % hydrogen combustion was almost complete.

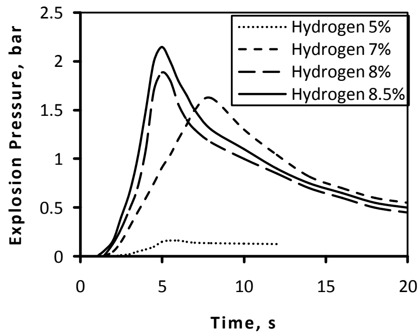

Figure 2: Explosion pressure curves for hydrogen-air-mixtures (ignition at 0.30 m from the bottom of sphere)

Fig. 3 shows explosion pressure-time histories for 9.5,10 and 12 % hydrogen after reproduction of data. Upward followed by downward flame propagation persists at 9.5 % hydrogen. Complete combustion of hydrogen was observed for mixtures containing 10 % hydrogen. At 12 % hydrogen, the pressure transients approximate that of spherical propagation. The results presented in Figs.2& 3 are for single spark ignition. Some experiments were conducted using continuous sparking (duration - 3 s) for 7 % hydrogen concentration with ignition at the bottom. The pressure-rises were higher (1.9 bar) than that with a single spark (i.e. 1.625 bar). The analysis of residual mixture showed that the hydrogen was reduced to 0.2 % to effect more complete burning within the column of flame lets in upward propagation in comparison to 40 % using single spark.

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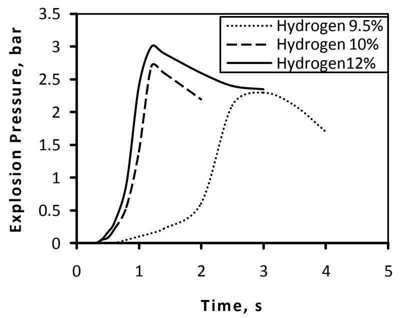

Figure 3: Explosion pressure curves for hydrogen- mixtures (ignition at centre of sphere)

The explosion pressure verses time curve for stoichiometric (29.5%) hydrogen- air mixture is reproduced in Fig.4. It has been reported that burning velocity for hydrogen-air mixture attains its highest value at 42 % hydrogen, nearly 1.5 times as high as the burning velocity at 29.5 % (Liu and MacFarlane, 1983). Experiments were therefore conducted at 42 % hydrogen concentration. Combustion at 42 % hydrogen was complete in about 30 % less time than for 29.5 % hydrogen.

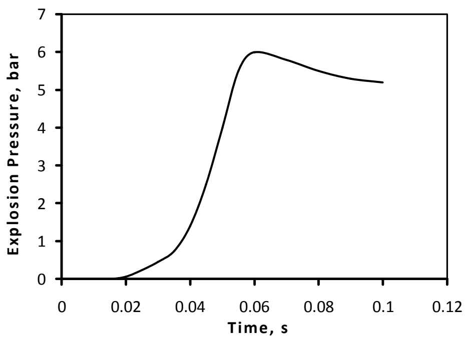

Figure 4: Explosion pressure-time history for stoichiometric hydrogen-air mixture

Fig.5 shows explosion peak pressures verses hydrogen concentration for central ignition at 25^{o}C. The peak pressure increases as the hydrogen concentration is increased, reaching a maximum at stoichiometric concentration. Beyond this, the peak pressure drops. Below stoichiometric composition all the hydrogen is consumed and above stoichiometric composition all the oxygen is consumed. For quiescent combustion, the rise in peak pressure is abrupt for hydrogen concentration above 8%, suggesting that the nature of flame propagation may have changed. The agreement between present data and those of Kumar et al. (1983, 1983b) and Furno et al. (1971) is good.

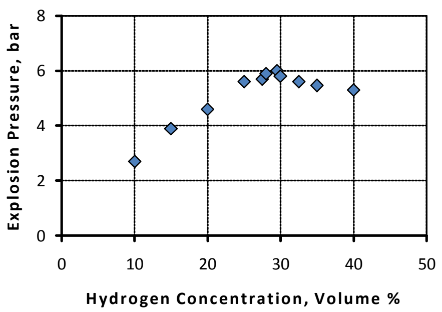

Figure 5: Variation of maximum explosion pressure with hydrogen concentration

The experimental data for fraction of hydrogen burned as a function of initial hydrogen concentration for experiments presented in Figs. 2, 3 & 4 are given in Fig.6. For hydrogen concentrations near lower explosible limits (5,7,8 & 8.5 % hydrogen) for quiescent mixture with ignition at the bottom, the burned fraction increases as the hydrogen concentration increases and reaches 100 % at 8.5 % hydrogen concentration. For higher hydrogen concentrations (9,9.5,10, 12 & 29.5 % hydrogen) experiments were conducted with centrally ignited mixtures. The burned fraction for 9.5 % hydrogen has been found 80 % which is 100% with ignition near the bottom of sphere. The combustion was complete at 10% hydrogen concentration. The burned fraction for centrally ignited mixture increases and stays at 100 % until the stoichiometric composition is reached. As expected, beyond this concentration the burned fraction decreases with hydrogen concentration since all the available oxygen is consumed.

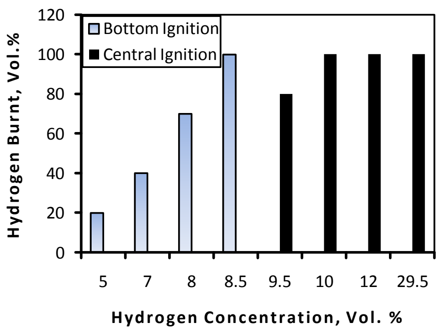

Figure 6: Hydrogen burnt as a function of initial hydrogen concentration in hydrogen-air mixture

Turbulence enhances the rate and extent of combustion (Abdel-Gayed and Bradley, 1976). Figs. 7 & 8 summarize the results of several experiments with and without turbulence. Fig.8 shows effect of initial turbulence on combustion with bottom ignition for 6 % hydrogen. The dashed curve is for an originally quiescent mixture, where the extent of combustion is only 30%, because of buoyancy effects. When initial turbulence is present, the rate of combustion is greatly increased and nearly 80 % of the hydrogen is consumed. The effect of turbulence on maximum explosion pressure for various concentrations of hydrogen is shown in Fig. 8. The measured r.m.s. intensities varied from about 2 m/s to less than 1m/s at large distances from the fan. At higher hydrogen concentrations, fan generated turbulence accelerated the combustion slightly. At stoichiometric hydrogen concentration, without turbulence, combustion was complete in 0.1 s, and with turbulence in about 0.07 s. At high hydrogen concentrations, burning velocities are already high and turbulent intensities lower than the burning velocity itself may not accelerate the combustion much. Similar observations were made in earlier investigations (Kumar et al., 1983b) who observed that for 27% hydrogen in air at 100^{0}C, in 2.3-m diameter vessel, combustion was complete in 0.09 s, and with turbulence in about 0.065 s. With turbulence, buoyancy effects become less important. Since turbulent burning velocities are much higher than laminar burning velocities, combustion is over before buoyancy-induced velocities become appreciable. Further, it is difficult to speak of a single fireball when turbulence is present. The combustion becomes more distributed, and large burning eddies become fragmented into several small ones moving in different directions. These set up their own flame centres similar to the effect of multiple ignition sources, as discussed by Hertzberg (1981b).

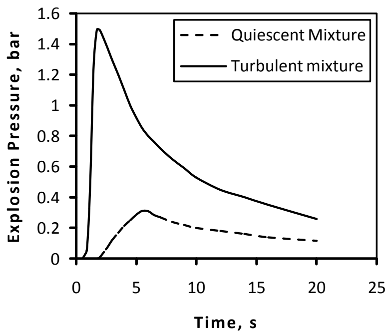

Figure 7: Effect of turbulence on explosion of hydrogen-air mixture (6% hydrogen in air, ignition at 0.30 m from bottom of sphere)

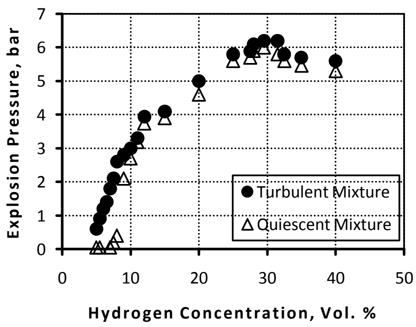

Figure 8: Effect of turbulence on maximum explosion pressure for hydrogen-air mixture (ignition at centre of sphere)

Ignition source location affects the rate and extent of combustion. Fig.9 shows effect of location of ignition source on maximum explosion pressure for hydrogen-air mixtures for 7 % hydrogen. Bottom ignition leads to faster combustion. A greater fraction of the hydrogen is burned during upward flame propagation with bottom ignition than with central ignition. The subsequent downward rate of propagation depends on fireball size reaching sphere top.

IV. CONCLUSIONS

The explosion severity of hydrogen-air mixture has been measured as maximum explosion pressure using 3.66 m diameter spherical vessel. The pressure-time data between concentrations 4 to 40 % of hydrogen in air reported in this paper are important for designing explosion safety measures to protect hydrogen handling installations. The experimental results showed that bottom ignition results in larger extent of hydrogen combustion and is more effective than central ignition in establishing a flame, even at very low concentrations and combustion time is shortest for stoichiometric hydrogen-air mixtures. Turbulence increases the rate and extent of combustion in almost all cases. The effect of fan-induced turbulence is considerably less at high hydrogen concentrations (10-42 %) than for near-limit lean mixtures.

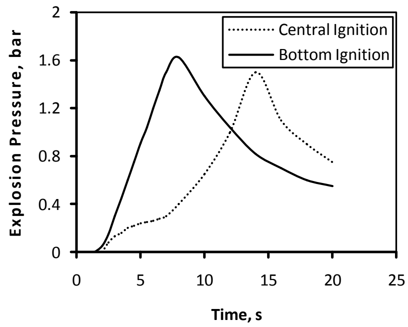

Figure 9: Effect of location of ignition source on maximum explosion pressure for hydrogen-air mixture (7% hydrogen in air)

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