Group: Hydrogen

Fundamental Behaviour of Hydrogen to Applied Accident Consequence Analysis for Hydrogen as a Safe Energy Carrier

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Introduction

The Hydrogen Group continued to develop models and in-house specialized reactive CFD codes (GASFLOW and COM3D) and conducted a huge experimental program to deepen the understanding of the mixing behavior and the transient combustion phenomena, flame acceleration and deflagration-detonation-transition, of accidentally released hydrogen. These activities were focussed on phenomena related to cryogenic hydrogen in 2020.

Members of the group are actively transferring their insights and expertise into the respective IEA Hydrogen Task 37, the standards committees of ISO (TC 197), CEN/CENELEC (TC 6) and in the European Hydrogen Safety Panel. Among other third party funded projects the Hydrogen Group has been conducting ambitious experimental programs for the European Fuel Cell and Hydrogen Joint Undertaking (FCH JU) funded projects PRESLHY - Pre-normative research for the safe use of liquid hydrogen - and HyTunnel-CS - Pre-normative research for safety of hydrogen driven vehicles and transport through tunnels and similar confined spaces.

This report present some highlights from the experimental campaigns with liquid, cryogenic respectively, hydrogen conducted in the frame of the European PRESLHY project. The experiments will be presented with increasing confinement of the reactive mixtures and start with a description of pool experiments.

Liqud Hydrogen Pool Experiments

The experimental setup for the Pro Science (PS)/KIT POOL experiments is shown in Figure 1 below. The facility comprises mainly of an insulated square stainless-steel box (50 cm x 50 cm, height: 20 cm) on a scale. Three identical boxes were made and filled up to half the height (10 cm) with different substrates (concrete, sand, water and gravel) prior to the experiments. The concrete pool was prepared already more than one month prior to the experiments, while the experiments with water and gravel were performed with the same box, which was freshly filled with the respective substrate for the corresponding experiment.

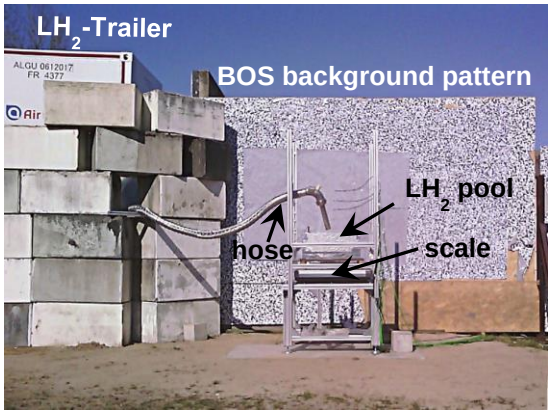

Figure 1: POOL-facility set-up for the unignited and ignited LH2 pool experiments of the PRESLHY project.

Inside and above the substrate, as well as above the facility in total 24 thermocouples were distributed. Above the facility also three measurement positions for remote H2-concentration measurements were placed, see Fig. 2.

Further instrumentation included optical observation and ambient wind measurements. In four of the ten experiments also a large fan was installed to investigate the influence of side wind on the formation and evaporation of LH2-pools.

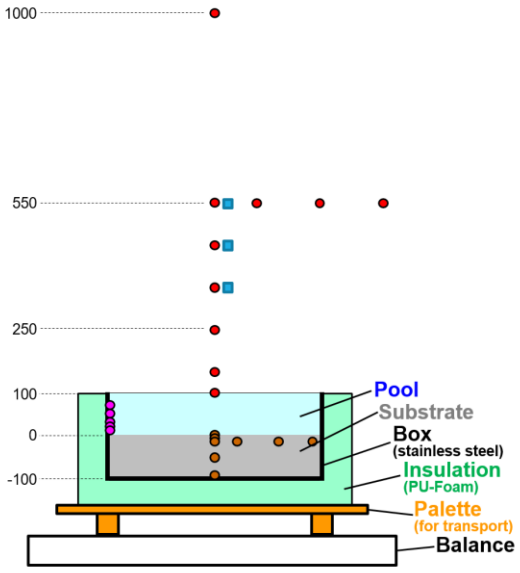

Figure 2: Sketch of POOL-facility set-up

In most of the experiments the pool was filled three times with LH2 until it started to overflow to investigate the influence of different initial temperatures of the substrate. After the fillings the pool was left to evaporate. The filling level of the pool was determined using the weight of the facility and the thermocouples inside the pool in different heights, which clearly indicated their coverage with LH2 by a constant value of approx. 20 K, while they started to show higher transient values as soon as they were exposed to the atmosphere, see Fig. 3.

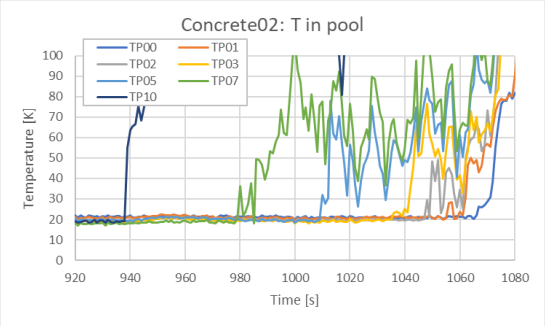

Figure 3: Temperature recordings for experiment Concrete02.

With these measurements the evaporation rates for the different substrates have been determined, see Figure 4. Also the influence of side wind on this phenomenon was assessed. In all cases much faster evaporation rates were determined for side wind conditions (vWind ≈ 5 m/s). A further important outcome of the experiments was information on the concentration distribution above the pools that was used to define the ignition position and point in time for a second experimental series in which the gas cloud above the pools is ignited.

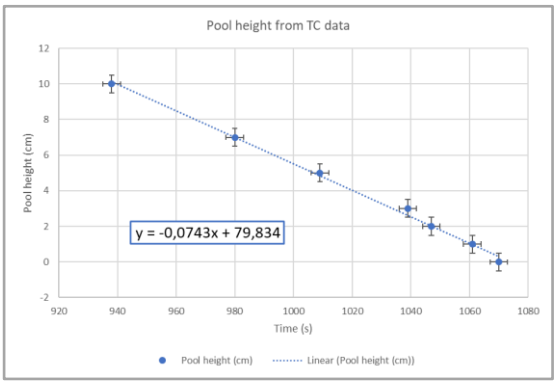

Figure 4: Example for determination of LH2-evaporation rate; here experiment Concrete02.

With the same set-up and experimental procedure also experiments on the ignition of LH2-spills were performed. For these experiments the instrumentation above the pool was removed (compare sketch in Figure 2) and replaced by 6 fast pressure sensors that were

positioned in heavy adapters on the ground in the close vicinity of the facility. The gas cloud above the pool was ignited using two electrodes in between which a high frequency spark (60 kV, 200 Hz) was generated. In total 14 ignited POOL-experiments with the four substrates concrete, sand, water and gravel were performed. In the tests the same pools as in the unignited experiments were used and mainly ignition point in time and height were varied. The ignition settings were chosen on the basis of the results of the LH2 filling level measurements and the H2-concentration measurements of the unignited tests performed before.

The ignition was initiated during the evaporation phase after the second filling of the pool, since after the first filling vigorous boiling behaviour above the still rather warm substrate occurs. After the second filling the boiling behaviour above the precooled substrate is calmer and thus the filling level and concentration measurements give more precise values.

In the ignited experiments different degrees of damage were observed for the different substrate materials. For the substrates with a rather low porosity almost no or only minor damage was observed (see left part of Figure 5), while the experiment Gravel04 with highly porous substrate of gravel showed a complete destruction of the facility (see right image of Figure 5).

The reason for this exceptional behaviour, shown with a sequence of frames from the high speed video in Figure 6, is potentially condensed or frozen oxygen located in the free space between the stones of the substrate layer. This mixed with the refilled LH2 and finally reacted in the observed explosion.

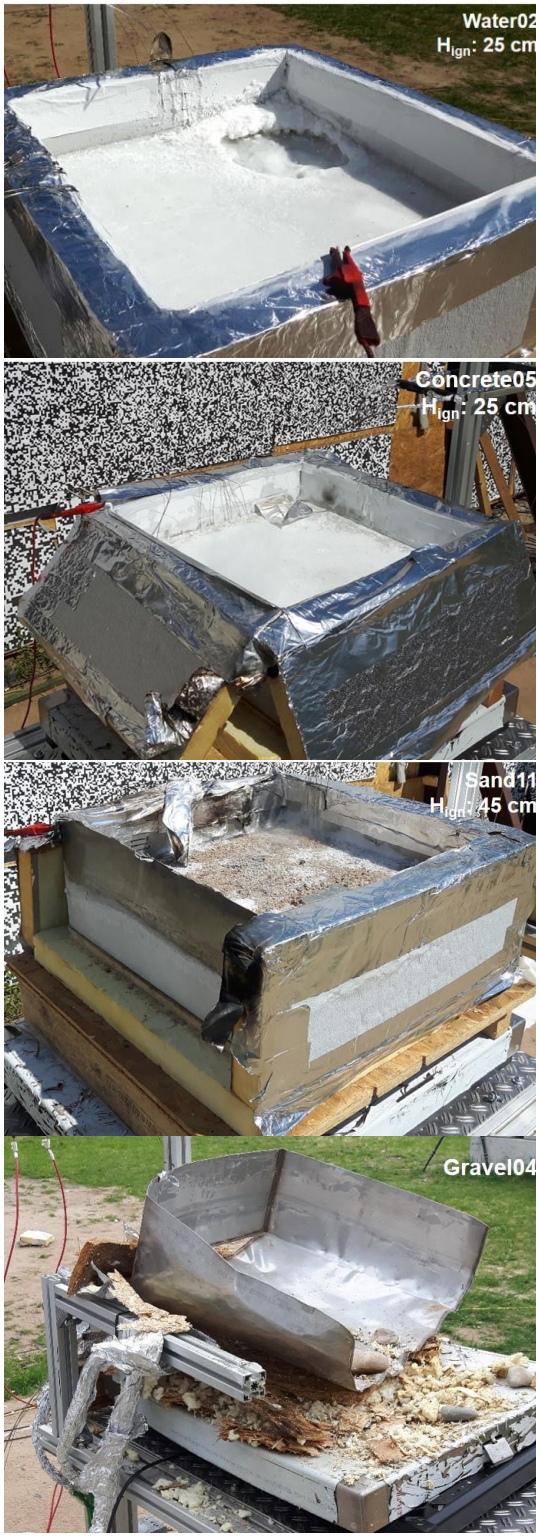

Figure 5: Different degrees of damage to the facility observed in the ignited pool experiments for the different substrates

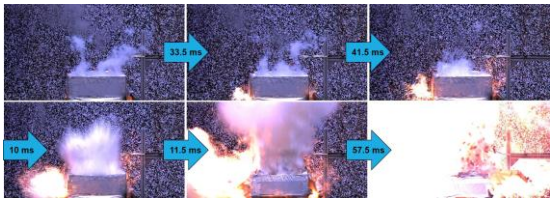

Figure 6: High speed video sequence of the experiment with ignition Gravel04 (PHOTRON High-Speed Camera, 2000 fps)

In summary, the observed evaporation rates provided valuable validation data for existing correlations and the observed massive explosion of the gravel substrate will initiate some modified recommendations for suitable ground materials for LH2 installations.

Transient cryogenic jet ignition

Transient cryogenic hydrogen jet fire behaviour, including scaling and radiation properties has been investigated with the small scale discharge facility DISCHA. The discharged hydrogen inventory varied with the initial pressure and temperature from 4.4 to 138 g. Similar as for the unignited discharge experiments done in 2019 [1] the release nozzle inner diameter was varied from 1-4 mm. Measurements consisted of background imaging system (BOS) combined with a high speed camera, fast pressure sensors and thermocouples. Additionally, a fast thermos-vision FLIR camera allowed monitoring the transient temperature fields. With mass flow rate and hydrogen distribution profiles known from the precursor unignited experiments, transient ignition phenomena and further flame development have been investigated with respect to maximum combustion pressure, temperature and heat flux radiation for model validation and hazard distance evaluation.

Unburned cold hydrogen jet was ignited with different delay time after jet initiation at different distances from the nozzle. Then, either a steady flame or a strong explosion with formation of a spherical shock wave might occur just after ignition (Figure 7). The over-pressure from 0.04 to 0.115 MPa corresponds to a visible shock wave velocity from 390 to 480 m/s measured by high speed BOS imaging.

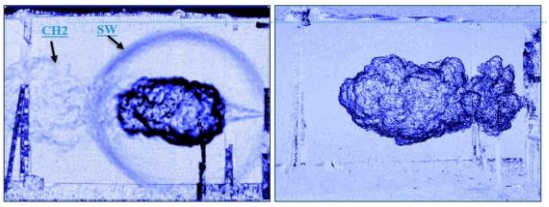

Figure 7: A shock wave formation (left) and a stationary jet fire (right) established under ignition of 4-mm nozzle and 20 MPa pressure hydrogen release: SW –shock wave; CH2 –unignited hydrogen

A sequence of frames with temperature distribution is obtained by thermo-vision FLIR-Camera (15 fps) (Figure 8). For ambient conditions 285K the local maximum combustion temperature changes from 1100 to 540K corresponding to maximum heat flux of 85 kW/m2. At the same time, the average integral heat flux of whole surface is about 6.5 kW/m2. At cryogenic temperature of 80K, the maximum temperature changes from 1330 to 710K corresponding to maximum heat flux of 177 kW/m2 in the center of jet fire. The average integral heat flux of whole surface is about 11 kW/m2. The reason of such difference is four times larger hydrogen inventory and also 2.5 times higher mass flow rate at cryogenic temperature leading to 1.3 times higher temperature, 2 times higher heat flux of flame radiation, 1.5 times larger flame length and 1.4 times longer release time.

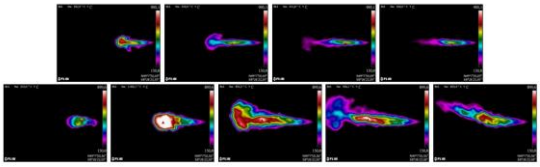

Figure 8: Temperature distribution within jet fire structure. Jet fire radiation for 2-mm nozzle and 10 MPa of initial pressure at T=285 K (upper sequence of frames) and T=80 K (lower sequence of frames).

Using the pressure records and the results of extensive optical observation with up to five cameras the ignition and combustion behaviour of the jets could be analysed. After ignition the flame either burns back to nozzle or not. This difference is important since in case the flame burns back to the nozzle it will continue burning, while in case the flame does not burn back to the nozzle it will quench after the ignition source is turned off. Using sensor records and optical observation on flame behaviour tests can be divided in two groups, where the flame burns back to the nozzle or not. When this data is combined with the results of the H2-concentration measurements of the unignited tests a clear trend can be observed. For very low H2-concentrations in the ignition position no ignition occurs, while for H2-concentrations lower than approx. 10 Vol% the jet is ignited but does not burn back to the nozzle. For H2-concentrations higher than approx. 10 Vol% in the ignition position the flame burns back to the nozzle.

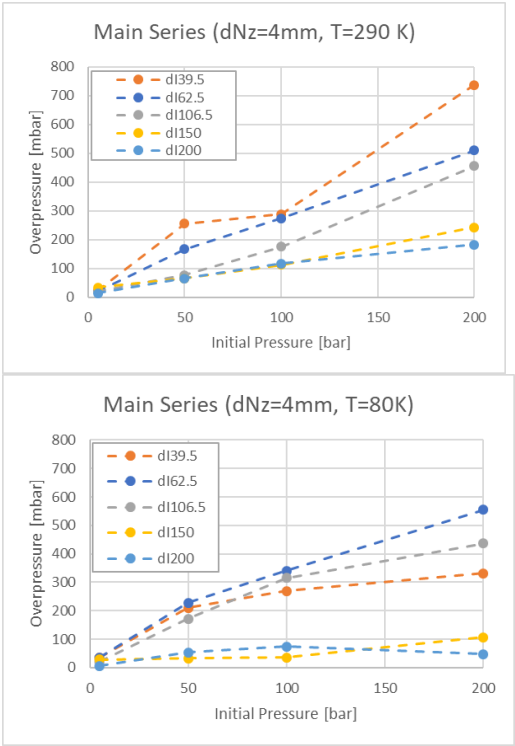

Figure 9. Maximum measured overpressures in experiments at ambient (top) and cryogenic temperature (bottom) of the discharged gases.

Concerning the combustion overpressures the following observations are made. In general, the maximum measured overpressures increase with increasing nozzle diameter, while the gas temperature in the reservoir seems to have no significant influence on the maximum pressure load. At ambient gas temperature the measured overpressures increase with decreasing ignition distance (Figure 9 top), while in the cryogenic tests the highest overpressures are measured for an ignition distance of 62.5 cm (Figure 9 bottom).

CryoTube Experiments

More than 100 experiments were made with the cryogenic combustion tube CRYOTUBE with L=5 m and 54-mm id and three blockage ratios BR = 0, 30 and 60% (Figure 10). About half of the experiments were made with hydrogen-air mixtures at cryogenic temperatures (approx. 80 to 130 K). The temperature was supported by the location of the tube under a layer of LN2 at T=77K (Figure 11). Subsonic, supersonic deflagrations and detonations were monitored for cryogenic hydrogen combustion by light sensors and pressure gauges located along the tube.

Figure 10: CRYOTUBE experiments mounted on a rack for the ambient warm pre-cursor tests.

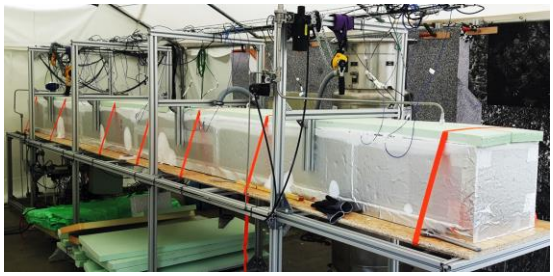

Figure 11: CRYOTUBE immersed in a bath of LN2 with supporting structure.

The critical conditions for flame acceleration to the speed of sound or detonation were evaluated as a function of initial temperature. Particularly at 100 K, it shows a much higher hydrogen concentration of 16% H2 leading to sonic deflagration than that of 9.6%H2 predicted by advanced extrapolation of existing high temperature data before the tests. The correlation for the critical expansion ratio based on current experiments is quite simple and useful in a very wide range of initial temperatures (T = 90 - 650 K):

$$\sigma^* = 2200 \cdot T^{-1.12}$$

The run-up distance to the speed of sound or detonation in a smooth channel (BR=0) at cryogenic temperatures is two times shorter than at ambient temperature. For the first time, a steady-state flame propagation regime in a smooth channel with the speed of sound in combustion products was frequently observed in cases with suppressed detonation.

The detonation cell sizes at cryogenic temperature T = 100K have been evaluated on the basis of existing criteria for detonation onset in smoot and obstructed tubes and can be approximated by a polynomial function of hydrogen concentration [H2]:

$$\lambda[\text{mm}] = 0.0006724[H2]^4 - 0.1039[H2]^3 + 6.0786[H2]^2 - 159.74[H2] + 1603.3$$

It appears that the detonation cell sizes for hydrogen-air mixtures at cryogenic temperature T=100K only insignificantly differ from that at ambient conditions. With the correct detonation cell size, all known DDT criteria can be used to assess the detonability of hydrogen – air mixtures in different geometries and scales at cryogenic temperatures.

The maximum combustion pressure at cryogenic temperatures is 2-3 times higher than that for ambient conditions. Theoretically, the maximum combustion pressure for sonic deflagration, which is roughly equal to adiabatic combustion pressure, is proportional to the temperature ratio T0/T, where T0 =293K and T is the cryogenic temperature. Similarly, the maximum detonation pressure can also be predicted. It demonstrates a very high level of hazard under cryogenic hydrogen combustion. Even a sonic deflagration can be 1.5 times more dangerous than the detonation of the same hydrogen-air mixture at ambient conditions.

A broader overview on all PRESLHY results may be found in [2], [3] and in particular in [4].

Partner

We would thank Andreas Friedrich, Joachim Grune, Anke Veser from Pro-Science for the good work, which they have done.

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[2] Jordan, T., Safe Use of LH2 in Non-industrial Settings, Contributions of the FCH JU project PRESLHY 13th plenary SFEM Working Group HYDROGEN, 28 January 2020, Brussels, Belgium (https://hysafe.info/wp-content/uploads/sites/3/2020/02/20200128_SFEMWorkingGroupHYDROGEN_30min.pdf)

[3] Jordan, T.; Hecht, E.; Odegard, A.; Low T LH2/cryogenic related progress in Safety Research HySafe Research Priorities Workshop, 30 October 2020, online presentation (https://hysafe.info/wp-content/uploads/sites/3/2020/11/ResearchPriorities_LH2_Jordan_Hecht.pdf)

[4] https://preslhy.eu/meetings/dissemination-conference/