

# STATE-OF-THE-ART AND RESEARCH PRIORITIES IN HYDROGEN SAFETY

Kotchourko, A.<sup>1</sup>, Baraldi, D.<sup>2</sup>, Bénard, P.<sup>3</sup>, Jordan, T.<sup>1</sup>, Kessler, A.<sup>4</sup>,  
LaChance, J.<sup>5</sup>, Steen, M.<sup>2</sup>, Tchouvelev, A.<sup>6</sup>, Wen, J.<sup>7</sup>

<sup>1</sup>IKET, Karlsruhe Institute of Technology, P.O. Box 3640, 76021 Karlsruhe, Germany,  
[alexei.kotchourko@kit.edu](mailto:alexei.kotchourko@kit.edu), [thomas.jordan@kit.edu](mailto:thomas.jordan@kit.edu)

<sup>2</sup>IET, European Commission DG-JRC, P.O. Box 2, 1755 ZG Petten, The Netherlands,  
[daniele.baraldi@jrc.nl](mailto:daniele.baraldi@jrc.nl), [marc.steen@ec.europa.eu](mailto:marc.steen@ec.europa.eu)

<sup>3</sup>Hydrogen Research Institute, Université du Québec à Trois-Rivières, 3351 Boulevard des  
Forges, P.O. Box 500, Trois-Rivières, Québec, Canada, G9A 5H7, Canada,  
[pierre.benard@uqtr.ca](mailto:pierre.benard@uqtr.ca)

<sup>4</sup>Fraunhofer ICT, J-v-Fraunhoferstr. 7 Pfinztal, 76327, Germany,  
[armin.kessler@ict.fraunhofer.de](mailto:armin.kessler@ict.fraunhofer.de)

<sup>5</sup>Risk & Reliability Analysis, Sandia National Laboratories, P. O. Box 5800 MS 0748,  
Albuquerque, New Mexico 87185, USA, [jllacha@sandia.gov](mailto:jllacha@sandia.gov)

<sup>6</sup>A.V. Tchouvelev & Associates Inc., 6591 Spinnaker Circle, Mississauga, Ontario, L5W 1R2,  
Canada, [atchouvelev@tchouvelev.org](mailto:atchouvelev@tchouvelev.org)

<sup>7</sup>School of Engineering, University of Warwick, Coventry CV4 7AL, United Kingdom,  
[jennifer.wen@warwick.ac.uk](mailto:jennifer.wen@warwick.ac.uk)

## ABSTRACT

On October 16-17, 2012, the International Association for Hydrogen Safety (HySafe) in cooperation with the Institute for Energy and Transport of the Joint Research Centre of the European Commission (JRC IET Petten) held a two-day workshop dedicated to Hydrogen Safety Research Priorities. The workshop was hosted by Federal Institute for Materials Research and Testing (BAM) in Berlin, Germany. The main idea of the Workshop was to bring together stakeholders who can address the existing knowledge gaps in the area of the hydrogen safety, including identification and prioritization of such gaps from the standpoint of scientific knowledge, both experimental and theoretical including numerical. The experience highlighting these gaps which was obtained during both practical applications (industry) and risk assessment should serve as reference point for further analysis. The program included two sections: knowledge gaps as they are addressed by industry and knowledge gaps and state-of-the-art by research. In the current work the main results of the workshop are summarized and analysed.

## INTRODUCTION

The International Association for Hydrogen Safety (HySafe) is globally the focal point for all hydrogen safety related issues. It is an international non-profit organization that currently has more than 30 members from industry, research organizations and universities representing 14 countries worldwide. The Association facilitates the networking for the further development and dissemination of knowledge and for the coordination of research activities in the field of hydrogen safety. It determines the state-of-the-art in hydrogen safety approaches and assessments and continuously develops further the associated strategic research agenda. The Institute for Energy and Transport (IET) of the Joint Research Centre of the European Commission provides scientific and technical support on energy issues to policy makers of the European Union (EU). Special emphasis is given to the security of energy supply and to more sustainable, safer and cleaner energy production and use for the future.

Although fuel cells and hydrogen technologies are still some distance away from full commercialization, industry has identified early markets that exploit one or more advantages of the technology (high efficiency and reduced energy consumption, low noise, low heat signature, absence of exhaust fumes, reduction of space requirements and weight, longer runtime, etc.) and that can

already be implemented using current technology. For example, in North America within the last 5 years there has been a few thousands of hydrogen fuel cells systems deployed for materials handling (predominantly), back-up power and CHP applications. However, while fuel cells and hydrogen technologies are already penetrating the market in a number of applications, sustained R&D, private and public, is still needed for effectively addressing the remaining high-risk technological barriers in a pre-competitive environment. One of the key R&D areas, preferably to be carried out through international cooperation, is pre-normative research for the establishment of fit-for-purpose Regulations, Codes and Standards (RCS) to ensure safety of fuel cells and hydrogen technologies.

As communicated by the European Commission in COM(2011)808, international cooperation with third countries is necessary to address effectively many specific objectives defined in *Horizon 2020* – the European Union Framework Programme for Research and Innovation (2014-2020). In particular, development of worldwide standards and guidelines was identified as a way to increase competitiveness of industry.

International cooperation is essential for frontier and basic research in order to capture the benefits from emerging science and technology opportunities (COM(2011)808).

Wide spread deployment and use of hydrogen and fuel cell technologies can occur only if hydrogen safety issues have been addressed in order to ensure that hydrogen fuel presents the same or lower level of hazards and associated risk compared to the conventional fuel technologies. To achieve this goal, the hydrogen safety research should be directed to address the remaining knowledge gaps using risk-informed approaches to help develop engineering solutions and RCS requirements that meet individual and societal risk acceptance criteria, yet are cost-effective and market-competitive.

Prioritization of research is best conducted in consultation with a pool of experts representing industry and research organizations. Building on the success of the previous workshop organized by JRC IET in October 2009 to address knowledge gaps in CFD modelling [1], HySafe and JRC IET partnered to organize a Research Priorities Workshop in Berlin on October 16-17, 2012 hosted by BAM (on behalf of HySafe). The participating experts were carefully selected according to their experience/expertise, number of scientific publications and participations to International Conferences, seminars, workshops as well international and European funded projects. By performing a consultation with industry and a broader research community as well as a state of the art review on hydrogen safety issues (including of CFD modelling), a consensus was reached among the experts as to the remaining gaps in the field and on the priority of the research needs.

Identifying the remaining knowledge gaps is a logical necessary step for making decisions on the next steps to carry out the full and safe utilization of hydrogen. The document aims to become a reference document for researchers/scientists and technical (including industry) experts working in the area worldwide. It is also a welcomed contribution for the Fuel Cell and Hydrogen Joint Undertaking and for other funding bodies/organizations that must make decisions on research programmes and during the selection/choice of projects to be financially supported pursuing the safe use of hydrogen within Horizon 2020 framework. The performed analysis and recommendations are expected to work as a catalyst to accelerate both the improvements of existing research programmes and the developments of new engineering guidelines and industrial practices, as well as supporting formulation of and compliance with RCS requirements.

## **INDUSTRIAL PERSPECTIVES**

Consultations with industry stakeholders constitute a critical component in setting research priorities. From this perspective, breadth of industry representation is important. For this reason, the companies operating within different market segments of the hydrogen and fuel cell sector were invited to share their experiences and needs as well their perspectives on research priorities in the field of hydrogen safety and standardization. The industry perspective was presented from four different market segments as follows:

- Hydrogen fuel cells developer and provider – Ballard Power Systems, Canada
- Global industrial gas company – Air Liquide, France
- International fuel and refuelling provider – Total, Germany
- Multi-industrial corporation – Kawasaki Heavy Industries, Japan

A common theme in the industry message that was particularly clearly articulated by Air Liquide was to improve knowledge quality for practical applications. This can be achieved by

- Prioritizing knowledge gaps
- Focusing research on industry needs
- Aiming towards international standardization and shared guidelines.

In reviewing industry views presented by Ballard (Jake DeVaal) and Air Liquide (Sidonie Ruban) it is remarkable to note that Air Liquide's first markets (e.g., telecom backup, mobile generators, forklifts, and automotive) all appear highly similar to Ballard's chosen markets. This suggests that the adoption of fuel cell technologies in the market place treads along the growth of their value propositions in these various applications one-by-one. In terms of the new safety challenges that Air Liquide identifies, (e.g., leak tightness, material compatibility, containment of high pressure, 'indoor' use, and the need to understand failure mechanisms for mobile composite storage), these are common issues that Ballard shares an interest in, especially in its system products; but, due to fuel containment issues typically being the responsibility of the gas supplier or tank manufacturer, the fuel cell industry has typically avoided these, but has addressed some aspects of indoor operation towards understanding leak outcomes behaviour [2].

A similar situation exists for the industry perspectives on hydrogen refuelling stations (from René Kirchner, Total Germany) and on large-scale liquid hydrogen (LH<sub>2</sub>) infrastructure (from Suguru Oyama and Shoji Kamiya, KHI), where the determination of safety distances, regulator/certification involvement in permitting these installations, and, most importantly, public acceptance of these are the main common themes shared across the diverse industry perspectives. Thus, while each industry tends to come with its own set of barriers and specific challenges, the common thread is the need to understand how hydrogen in production, storage, delivery, and use systems behaves under leak or emission conditions, and measures needed so that failures in these systems, although unlikely, will not pose undue risks to users or the public.

### **Ballard Power Systems, Canada**

Since the early 2000's Ballard is a dominant player in automotive fuel cell development, with Ballard stacks used in Daimler and Ford automotive demonstration programs, and also in the very successful European CUTE bus demonstration project. Based on the progress made in developing several generations of automotive fuel cells, it was recognized that the investment required to develop a commercially-viable automotive fuel cell stack would exceed the company's resources, and the decision was taken in January 2008 to divest the automotive stack development assets to Daimler AG and Ford Motor Company and a newly created private company, AFCC, Auto Fuel Cell Cooperation Corp.

Continuously working in the field of commercialization of bus and distributed generation products, Ballard has identified the following specific areas for near-term hydrogen safety research:

- Improved fuel flow monitoring for hydrogen leak detection in Bus and DG products,
- Tools and approaches for addressing the H<sub>2</sub>/N<sub>2</sub> start-up discharge-emission hazards,
- Improved understanding of fuel cell recombination effectiveness, where recycling leaked H<sub>2</sub> through stacks is highly effective at recombining fuel but can also cause crossover leaks,
- Improved understanding of cathode air filtration effectiveness and H<sub>2</sub> fuel quality issues (e.g., biogas quality), and
- Qualify/use risk analysis tools and develop more meaningful standards.

## **Air Liquide, France**

Air Liquide is a world leader in gases for industry, health and environment. It has a long experience in the sector: more than 40 years in the hydrogen field and 10 years in fuel cell development and deployment. Air Liquide has acquired vast experience in the “hydrogen chain” that includes production, storage, distribution as well as dispensing via hydrogen stations. Through the years Air Liquide built globally an infrastructure that includes more than 200 hydrogen production plants and a broad range of hydrogen distribution assets such as pipelines (more than 1,850 km), trucks and cylinders.

In the materials handling market segment the focus is on hydrogen supply and refuelling of fuel cell forklifts. Hydrogen is either produced on site or delivered via tube trailers as a compressed gas or in liquid form by LH<sub>2</sub> tankers. In order to develop a sustainable mobility with hydrogen, it is necessary to develop a dedicated refuelling infrastructure. This task, in turn, is faced with a number of challenges, in addition to the safety-related issues:

- High pressure and specific mechanical loading
  - Leak tightness
  - Material compatibility (incl. hydrogen embrittlement)
  - Intelligent depressurizing tap
- ‘Indoor’ use (enclosed environment):
  - Natural ventilation
  - Structural strength of enclosures
- High capacity of hydrogen mobile composite storage
  - Failure mechanisms
  - Fire resistance.

In the past few years a number of knowledge gaps have been addressed such as:

- considering pipeline steel microstructure and fatigue enhanced embrittlement in addition to material composition [3];
- sizing release flow rate for fire protection of hydrogen composite cylinder taking into account pressure peaking effect, flame effects and storage leak and no burst phenomena [4]; and
- sizing openings for an efficient natural ventilation of enclosures: parameters such as opening size and position which have a strong influence on dispersion regimes.

## **Total Germany, Hydrogen/E-Mobility**

Total Germany started hydrogen activities in 2002 with a first station delivering gaseous hydrogen to the BVG buses (Berlin public transport authority). Since that time Total Germany acquired experience in hydrogen refuelling via 7 more hydrogen refuelling stations (HRS) projects including its first public hydrogen dispensing combined with a conventional station in Berlin in 2006. This was followed by another HRS in Berlin in 2011 where hydrogen refuelling was integrated into a new design with other fuels like CNG, gasoline, diesel and LPG.

Specific Total’s learning experience:

- local authorities need to be informed and involved as much and as early as possible;
- need for an overall guideline for HRS permitting (DIN, EN or ISO) to feel comfortable with and accelerate the process;
- knowledge dissemination within local authorities in Germany is inevitable and needed;
- exchange of experience within industry (CEP is a good example);
- key open issues as barriers to HRS commercialization;
- hydrogen metering;
- hydrogen quality sampling; and
- refuelling protocol, particularly for 700 bar dispensing.

## **Kawasaki Heavy Industries, Japan**

Founded in 1878, Kawasaki Heavy Industries, Ltd. (KHI), is a leading global diversified manufacturer of transportation equipment and industrial goods. With a broad technological base that encompasses land, sea, and air applications, the KHI Group manufactures ships, rolling stock, aircraft and jet engines, gas turbine power generators, environmental and industrial plants, and a wide range of manufacturing equipment and systems. KHI also produces world-famous consumer products such as motorcycles and personal watercraft.

The biggest knowledge gap here relates to appropriate/optimized safety distances as well as certain elements of safety design like a burn pond or a dike. Existing methods for calculation of safety distances for liquid hydrogen are based on data obtained in early 1960-1970's. Those correlations did not anticipate such significant amounts and as such cannot be used for reliable calculations. New correlations are needed.

A similar situation is with the shipment of large quantities of LH<sub>2</sub>. Large bulk transportation is not covered by any code or standard.

In summary, more knowledge is required related to

- spillage of large quantity of LH<sub>2</sub> on ground or seawater;
  - cloud dispersion of cold hydrogen from vent and its ignition;
  - performance of various thermal insulation options;
  - safety distance as function of LH<sub>2</sub> quantity and re-assessment of the scientific basis for existing correlations;
- evaluation of related hazards and their consequences; and risk assessment of typical accidents.

## **RISK-INFORMED SAFETY SCIENCE**

The safe use of any technology and associated facilities requires that the hazards and associated risk be understood and minimized. This can be accomplished by performing a quantitative risk assessment (QRA) where hazards are identified, possible accident scenarios are delineated, and the resulting consequences evaluated. There are several benefits to performing a risk assessment, either qualitative or quantitative, for a hydrogen facility such as a fuelling station. The most important benefit is that it provides a systematic framework for identifying what can go wrong at a facility and what can be done to prevent or mitigate possible accident scenarios. Thus, the results from a QRA can be used in a science-based, risk-informed process to establish requirements in regulations, codes, and standards (RCS) for the design and operation of hydrogen facilities. A science-based, risk-informed process utilizes science, engineering, and risk insights obtained from QRAs combined with other considerations to establish requirements.

### **QRA state of the art and identified gaps**

A hazard identification is the initial step in risk assessment. The purpose of the hazard identification is to identify all events that can affect facility operation leading to a hazard to individuals or other facilities. It involves not only the identification of accident initiators but also considers the potential scenarios that lead to harm to individuals or other facilities. The hazard analysis can also include a rough order of magnitude assessment of the frequency and consequences of the identified scenarios which is generally used to rate the criticality or importance of individual scenarios. Fundamental methods such as Hazard Identification (HAZID), Hazard and Operability (HAZOP) studies, failure modes and effects analysis (FMEA), and WHAT-IF analysis [5] are tools which can be used to identify the hazards and assess the criticality of possible outcomes. These methods also have the advantage of being sufficiently general for use on hydrogen facilities without specific adaptation. A desired feature of all these methods is that they should be performed by a multidisciplinary team that should include design and operations personnel with technical experience and expert knowledge of the facility design. The qualitative insights from obtained from these hazard assessment methods identify

how to reduce the potential for accidents and the resulting consequences regardless of the risk significance of the accident but provide limited insights on which improvements will provide the greatest reduction in risk. Those types of insights are available from the quantitative results of a QRA which makes it a more powerful tool than qualitative methods listed above for ensuring the safe design and operation of a facility.

In a QRA the total risk is calculated by taking the sum of the risk associated with each identified accident scenarios. In most QRAs of hydrogen facilities performed to date, the different potential accident scenarios have been delineated in event trees. Event tree analysis systematically explores the potential accident scenarios that can occur following an accident initiating event which is influenced by accident phenomena, mitigation system response, and operator actions. The event tree displays the sequences of events involving success and/or failure of the system components, and results in the identification of accident scenarios including their consequences and frequencies. The failure modes of mitigation systems can be depicted in fault trees. An alternative, well known approach to accident sequence evaluation using Bayesian Belief Networks [6] has been utilized in hydrogen applications [7,8]. Both event trees and Bayesian Belief Networks provide the necessary framework for quantifying risk.

In order to quantify the accident sequence models, data for the modelled events must be obtained. The required data includes the frequency of accident initiating events (e.g., hydrogen leaks), component failure probabilities, human error events, and the probability of certain accident phenomena. The initiating event frequencies and component failure, conditional event, and human error probabilities required in a QRA can be obtained directly from historical records. Unfortunately, there are little hydrogen-specific data available, no requirements for collecting data (the exception being the U.S. Department of Energy's (DOE) technology validation program [9], and current data collection efforts such as in the HySafe *Hydrogen Incident and Accident Database* [10] and the U.S. DOE's *Hydrogen Incident Reporting and Lessons Learned* database [11] are not sufficient for utilization in a QRA. Thus, data from other industries, primarily the oil and gas industry has been utilized in hydrogen facility QRAs. A different approach using a Bayesian process to combine limited hydrogen data with data from other industries was used in LaChance, et al. [12]. Without hydrogen-specific data, the fidelity of hydrogen QRAs is less than desirable contributing to the uncertainty in the resulting risk estimates. Thus it is important to begin planning and testing an industry-wide framework for QRA data collection activities and to begin serious efforts for collecting failure data.

In any technological application, humans can make errors resulting in dangerous situations that initiate or contribute to accidents. In fact human errors are often significant contributors to accidents. In general, human errors occur as a result of the lack of familiarity or experience to perform a required task or because they make a slip or error in performing required actions. Human errors resulting in accident initiating events can be identified as part of the hazard identification process discussed previously. Human errors that can exacerbate an accident sequence are identified as part of the accident sequence development process. To date, human errors have not been explicitly included in hydrogen facility QRAs (one recent exception is in Groth, et al. [13]). This lack of explicit treatment especially in light of the lack of hydrogen-specific accident data may result in a significant under prediction of the risk of operating a hydrogen facility. Thus in future QRAs, hydrogen accident models must be expanded to include plausible human failures. There are many HRA methods that can be utilized to quantify the identified human errors and some are more appropriate than others for specific applications. A description of many of these methods is provided in Forester et al. [14] along with an evaluation of their capability to satisfy accepted good practices (considering the current state-of-the-art) for performing a human reliability analysis.

A key parameter in the evaluation of hydrogen accident scenarios is the probability of ignition. There has been much work on the potential for self-ignition and delayed ignition of hydrogen jets but to date it has not been translated into a probabilistic model that is needed in QRA. However, efforts have been made to develop a non-mechanistic ignition probability models based on literature searches, empirical data, and some experimental data [15]. Ignition probabilities from many sources including [16] have

been utilized in hydrogen facility QRAs. More work in establishing a science-based probabilistic ignition model based on hydrogen-specific experimental and analytical information is required to obtain high fidelity risk results.

Accident sequence analysis results in the development of the different accident scenarios including the end states of those scenarios which generally involve hydrogen fires or explosions. Consequence calculations are generally performed for each of the identified end states and combined with the scenario frequency to determine the risk associated with the scenario. The consequence calculations should predict the physical circumstances resulting specifically from the accident (e.g., explosion overpressure and thermal radiation levels). The consequences are usually quantified using mathematical models; these may be either simple engineering models or Computational Fluid Dynamics (CFD) models. Because CFD simulations can take long periods of time, they are not practical for QRAs where the consequences from a large number of scenarios may have to be evaluated. Thus, there is a need to develop more simple engineering models for evaluating the consequences of hydrogen releases.

The results of consequence evaluations must be translated into a probability of causing damage to an individual, component, or structure for use in a QRA. This can be done using Probit functions which provide a statistical correlation between the magnitude of the consequence (e.g., thermal heat flux) and the resulting potential for damage. A good summary of Probit functions was provided in LaChance et al. [17] and included Probits proposed by TNO [18] and utilized in some hydrogen facility QRAs. Available Probit functions for predicting damage from a thermal heat flux are not hydrogen specific and thus introduce uncertainty in the risk results which can be evaluated using sensitivity studies.

In order to utilize the results of a QRA to make a risk-informed decision, it is desirable to establish risk acceptance guidelines or criteria. There are different types of risk criteria including those for individuals (separate criteria may be specified for workers and users of the facility and for people located near the facility) and for the population surrounding the facility. A good summary of potential risk measures is provided in Jonkman et al. [19]. Although each country has its own risk criteria, Tchouvelev et al. [20] has suggested uniform risk acceptance criteria for use in the hydrogen industry.

Finally, when utilizing the results of a risk assessment to make risk-informed decisions it is necessary to address the robustness of the results taking into account the uncertainties in the analysis. Uncertainty in hydrogen facility risk assessments results from many sources including: sparse data on hydrogen accidents, lack of understanding on phenomena (e.g., ignition), modelling assumptions (e.g., leaks modelled as circular orifices), modelling limitations (e.g., inability to model surface effects), and incompleteness (lack of analysis of external hazards such as earthquakes and high winds). Understanding the sources of uncertainty will indicate the parts of the QRA model that could be affected, and ultimately the results from the QRA model that may be impacted. Guidance on making decisions in light of uncertainties is needed and can potentially be adapted from other industries (e.g., see [21]).

## **SIMPLIFIED TOOLS**

One of the key tasks of scientific work is to translate fundamental scientific findings into practical formulas, which are easily applied in daily work. In many cases financial or computational resources and - even more often - time is limited, such that a physically resolved numerical solution of the complex phenomena relevant for hydrogen safety issues is practically impossible. Although CFD solvers and computer performance are developed further continuously, validated simplified correlations often suffice for first estimates or sensitivity studies with a large number of varying parameter settings.

So, the simplified toolkit consists of all kind of empirical correlations, criteria, statistics and models based on first principles, which are needed to assess the risk implied with certain hydrogen inventories or fluxes in user defined scenarios. Such a toolkit shall be based on robust, published, state-of-the-art

correlations. The design shall be highly modular and fast response times of the system are necessary to make it different from the more complex tools like CFD.

At the same time, a probabilistic component can be added up that would include an agreed set of failure frequency data, ignition probabilities and a set of appropriate probit functions. In combination and in its final version, the toolkit will allow a user to perform a QRA within those parameters, which should be a valuable tool for various stakeholders involved in the hydrogen FC field: design and process engineers from industry, standard development experts, risk analysts and QRA professionals, and technical authorities.

The tools shall be maintained by the hydrogen safety research community itself. As safety is a public concern and a big part of relevant scientific work is funded by public agencies anyhow, the toolset should be an open and free software system, which is well documented and quality assured in a cooperative manner. IEA HIA appears to be most suitable vehicle to foster and host global collaboration on toolkit development, while HySafe could serve as a natural custodian of the toolkit ensuring its global relevance as well as its timely maintenance.

Each tool of the toolkit shall consist of a set of input parameters and a set of output or result parameters. The model calculates the output with the actual input. All input and output parameters are elements of the respective scenario. Each tool shall be described in detail, the valid range of input parameters has to be controlled appropriately, literature references should be given and model tests have to be provided.

A typical use case will consist of a user defining explicitly a new scenario by giving the inventory or hydrogen flow, geometrical settings like confinement and/or congestion, mitigation measures, up to a leak size. For a statistical analysis any of these scenario properties might be defined by a probability distribution instead. Then the user might choose a tool to act on the scenario. The input, which was not yet defined but required by the selected tool, shall be input by the associated tool interface. If any of the input parameter lies outside the models validity range an appropriate reaction shall be taken. Appropriate measures are warnings or even an exception. Any output defined before the execution of the model, shall be overridden by the model. A warning will be issued.

Users with appropriate rights may edit tools or define new tools. New tools might use existing ones by calling them in a specific sequence or even recursion should be possible. These kinds of models shall be called "super-models". An example for such a super-model could be the tool for calculating flame radiation which would rely typically on the determination of a Froude number, suitable models for flame length and width, residence time, radiant fraction and so on.

With all requirements defined so far a WEB2.0 kind of implementation is envisaged. A system which allows for immediate testing and on-the-fly editing of the tools is the Smalltalk dialect Squeak based dynamic web development framework Seaside.

## **SOURCE, RELEASE AND DISPERSION FOR GASEOUS H<sub>2</sub>**

The evaluation of the safety of hydrogen systems requires methods to characterize the release of hydrogen and the determination of the extents of the flammable clouds for the establishment of the safety distances and hazardous zones. Jet and plume releases constitute a most common mechanism of release and dispersion. The scaling behaviour of the concentration field in the expanded region of jet releases have been characterized and validated experimentally and numerically. CFD simulations have been extensively used bringing the issue of the validity of the various approximations and models used to perform those simulations to the forefront of hydrogen safety research. In this context, benchmarking and validation exercises, originally initiated by the HySafe Centre of excellence, are still being pursued in the context of HySafe organisation to explore the usefulness and limitations of the modelling strategies and numerical approximations commonly used in CFD simulations. These



exercises should lead to the instigation of a standard validation matrix for CFD simulations that could be used as benchmarks for dispersion simulations.

### **Properties of jet releases**

The concentration field of turbulent hydrogen jets in air decays inversely proportional to the distance for round jets and to its square root for planar jets. Jets from non-circular orifices with aspect ratios far from unity are expected to behave like planar jet close to the source with an eventual crossover to the behaviour of round jets in the far field. Near-field studies of jets originating from elliptic orifices of various aspect ratios have been performed in the context of jet acoustics [22], and recently by Paraschivoiu et al [23] in the context of self-ignition of hydrogen jets. Detailed studies on the effect of the orifice shape on the  $H_2$  concentration in the expanded region seem, however, to be scarcer, despite the fact that realistic sources are likely to be linear and even annular.

In 1984, Birch et al proposed a methodology to evaluate the decay of the mean concentration along the centreline of a supercritical free jet [24]. They used a notional approximation, in which the concentration field is calculated as if it were produced by an effective orifice larger than actual nozzle diameter. Such effective source methodology has been extensively used to eliminate the need of carefully considering the shockwave structure of sonic jets close to the nozzle (Mach disk) in order to reduce computational requirements for numerical simulations. Subsequently, Birch [25] reformulated their effective diameter definition based on the conservation of both mass and momentum and later, Houf et al. [26] further developed their approach.

Typical modelling strategies are based on a three stage lumped parameter approximation (inside the storage unit, at the actual orifice, at the effective nozzle). Other approaches rely on the properties of the Mach disk, such as the models of Winters et al (2007) and GexCon (2008). In addition to the conservation equations, both use the normal shock relations to specify the flow properties at the various stages. Both approaches yield similar predictions [27, 28]. A detailed evaluation of recent notional nozzle models for free-shear under-expanded hydrogen jets has recently been performed by Papanikolaou et al. [29]. Simulations were performed using three turbulence models (the standard  $k-\epsilon$  model, the baseline  $k-\omega$  model and the shear-stress transport approach) and compared with experimental data for jet releases from three orifices with diameters of 0.25, 0.5 and 1 mm performed at the Karlsruhe Institute of Technology. Given a turbulence modelling strategy, the best accuracy was obtained when using the notional nozzle approximation of Birch [25] and Schefer [31], followed by Birch [24] and Ewan [30], and finally Hastard [32]. At high pressure where real gas effects become relevant, it was demonstrated that Schefer's model is the most accurate among the ones that were considered in that investigation. Despite the successes of the notional nozzle approximation, it remains unclear to what degree such approaches can be applied to the description of the attached or confined jets. A detailed analysis of the effect of this approximation on the transverse velocity distribution of under-expanded turbulent jets as a function of position along the axis of the jet should also be performed. The applicability of notional approximations to different shapes of the orifice of the release has not yet been looked in details.

For realistic jet releases, the development of the boundary layer of jet releases is initially driven by the momentum of the injected gas. However because of the low density of hydrogen, the driving force for the development of the boundary layer eventually becomes the buoyant forces. Buoyant jet releases occur for small, often subsonic releases, which do not in principle follow the scaling laws. Their behaviour can however be predicted numerically. Low momentum hydrogen jets (forced plumes) have been studied by Houf and Schefer [23]. They measured the jet mass fractions (Froude numbers of 99, 152, and 268) and compared the results with their integral model accounting momentum [34] and buoyancy [35] contributions. A non-Boussinesq engineering model was proposed by Xiao et al. [36] for fully turbulent horizontal jets. Deviations from the scaling behaviour of expanded jets along the centreline of the jet for subsonic low momentum horizontal jets (which could be defined operationally as the curve representing the positions of the local maxima of the concentration profile of the jet) are discussed in reference [37].

Predicting the scaling behaviour of the concentration as a function of distance is useful for hazard analysis as it can be used to contribute to the definition of exclusion zones based on the lower flammability limit of hydrogen. It is interesting to note that Molkov and Saffers recently [38] established a general correlation for the length of hydrogen flames normalized by the nozzle diameter as a function of the product of the ratio of the density of H<sub>2</sub> at the nozzle to the ambient density by the cubic power of the Mach number. This correlation was shown to be applicable to all flow regimes, including both buoyancy and momentum-dominated. In view of the fact that there are indications that the flame length is correlated to the size of an unignited jet [39], it would be interesting if a corresponding correlation could be derived for the flammable size of a hydrogen release.

The proximity of jets to surfaces will modify the scaling behaviour of expanded jet and typically increase their flammable lengths (4% vol. for H<sub>2</sub>). Proximity effects can play an important role for risk assessments (e.g., for consequences of a flash fire event, for which is usually assumed 100% lethal within the confines of the lower flammability limit (LFL). It may induce a Coanda effect that manifests as a creeping behaviour of the hydrogen concentration field close to the surface, a recirculation zone between the nozzle and the surface and generate transient behaviour such as puffing, which may temporarily increase the flammable extent [40]. The properties of attached jets have been studied numerically [41, 42] and to a lesser extent experimentally for unignited [43] and ignited [44] releases. In the simulations performed for H<sub>2</sub> and methane (CH<sub>4</sub>) jets over wide ranges of pressures (100-700 bars), an interesting behaviour was observed for the normalized overextent of the flammable distance induced by the presence of the surface when expressed as a function of a normalized distance of the orifice from the surface. Because of the simplifying modelling assumptions these results require experimental confirmation and cross-checking with simulation results obtained using more complex turbulence modelling.

### **Releases in enclosed areas**

Hydrogen concentration build-ups inside enclosed areas present particular issues with respect to safety because of reduced dispersion and the increased chance that an ignition event might occur. Numerical and experimental studies performed in [45, 46] show that hydrogen accumulation leads either to a stable, stratified distribution of concentration or to the formation of a homogeneous layer if the convective flows at the top of the enclosure are high enough [47-49]. The experiments [51] have shown two qualitatively different gas-dynamic patterns of gas cloud formation termed as «filling box» and «fading up box», depending essentially on the release speed. In a «filling box» case (at a low speed), the explosive cloud initially forms as a thin layer at the ceiling and then expands downward. In a «fading up box» case (at a high speed), the explosive cloud forms nearly uniformly throughout the whole volume. A study investigating the discharge of H<sub>2</sub> from onboard storage tanks through a pressure relief device (PRD) inside a garage like enclosures with low natural ventilation has been performed in [52]. It was demonstrated how for a constant release of 0.39 kg/s H<sub>2</sub> into a 30.4 m<sup>3</sup> garage with a single one-brick vent the overpressure within the enclosure reaches a level of 10-20 kPa, capable of destroying the garage within only 2 s. A phenomenological model has been developed, and compared with CFD simulations to predict the hydrogen release dynamics within an enclosure.

### **Computational fluid dynamics simulations**

CFD simulations of H<sub>2</sub> releases and their subsequent dispersions have the potential to be powerful tools to help assess the physical consequences of hydrogen releases, and to help study physical phenomena efficiently and to a high level of details. Benchmarking and validation exercises (*Standard Benchmark Exercise Problems*), originally initiated by the HySafe Centre of excellence [28], are still being pursued in the context of the international association HySafe to explore those issues and should eventually be the topic of a detailed, specific report. These exercises should lead to the instigation of a *standard validation matrix* for CFD simulations that could be used as benchmarks for dispersion simulations. The validation matrix would be constituted of state-of-the-art, high quality experimental datasets covering regions of parameter space applicable to hydrogen safety problems.

During the filling process of hydrogen tanks, the gas temperatures inside the tank can reach high values, potentially jeopardizing the structural integrity of the storage system and reducing the state of charge of the tank. Several research teams have performed CFD validation studies [53-60]. By comparing the simulation results with the experimental data they demonstrated that the current CFD models are capable of capturing the maximum temperature histories inside the tank with a sufficient level of accuracy. CFD can be instrumental in identifying the best filling protocols for the RCS.

## **SOURCE, RELEASE AND DISPERSION FOR LIQUID H<sub>2</sub>**

Liquid hydrogen is one of the promising alternatives for hydrogen storage and transport due to its larger density (~71 kg/m<sup>3</sup> at 20 K) compared to that of compressed gaseous hydrogen (0.08 kg/m<sup>3</sup> at 300 K). It was shown that when cryogenic pressure vessels for the automotive industry are filled with LH<sub>2</sub> or CCH<sub>2</sub> (cryo-compressed hydrogen), these vessels contain 2–3 times more fuel than conventional ambient temperature compressed H<sub>2</sub> vessels [61, 62]. For the same reason, liquid hydrogen is extensively used in rocket applications and it is considered one of the most promising solutions for the future of aviation [63-65].

Safety analysis of the release and dispersion of liquid hydrogen requires the understanding and quantification of phenomena that are different from those in the release of hydrogen in gaseous form e.g. two-phase release sources, multi-phase jets, gas behaviour at low temperatures, phase changes, pool formation and spreading, heat transfer with the surrounding environment, effect of weather conditions e.g. temperature, humidity, wind and atmospheric stability, effect of ground and roughness/obstacles configuration, and effect of turbulence and buoyancy on all the above phenomena. The quantity and the level of details of the experimental data that are currently available in the scientific literature are limited. The available data do not allow for the complete accurate quantification and modelling of the phenomena and for the validation of the models. The acceptance criteria for model performance evaluation that were developed and applied for other fields (e.g. air quality, liquefied natural gas (LNG) dispersion) should be revised for hydrogen. Regardless of the fact that the analytical models for the whole release and dispersion process and in some cases only for specific stages of the release have been developed, a complete validation of those models is missing.

### **State of the Art**

According to the Pritchard and Rattigan' position paper [66], “applications involving liquid hydrogen present additional fire and explosion hazards to those arising from use in gaseous form ...” and “the consequences of an accidental spillage or leak of liquid hydrogen are poorly understood, particularly the initial stages of pool spread and vapourisation”. Their conclusions are consistent with the findings of [1] on the gaps' identification of CFD modelling of accidental hydrogen release. In that report, it was highlighted that a limited number of experiments of LH<sub>2</sub> spillages are available in the scientific literature, e.g.: by the time of the report [67-69], then with liquid helium by Proust and co-workers [70], and more recently experiments at HSL/HSE [71, 72]. Their experiments provide a further confirmation that a pool can be formed if a liquid release is made on the ground and the ground surface is sufficiently cooled. Moreover oxygen and nitrogen freeze, forming a solid deposit on the ground. Friedrich [73] performed experiments of release and combustion of cryogenics hydrogen jets, providing an estimate of safety distances and an extrapolation model for other jet conditions. In general in the experiments in the literature, mainly the LH<sub>2</sub> release and dispersion are investigated while experiments with the entire sequence of release and dispersion followed by explosions and/or fires are rare [74].

Several physical phenomena have to be modelled in numerical simulations in order to describe accurately the LH<sub>2</sub> release and dispersion: two-phase release sources, multi-phase jets, gas behaviour at low temperatures, phase changes, pool formation and spreading, heat transfer with the surrounding environment, effect of weather conditions e.g. temperature, humidity, wind and atmospheric stability, effect of ground and roughness/obstacles configuration, effect of turbulence and buoyancy on all the above phenomena. Schmidt [75] performed CFD simulations of the experiments that were carried out

at BAM [76]. They assumed a pure gas release and have found noticeable discrepancies with the experimental results, connected in their opinion with the above assumption. The requirement for extended and systematic experimental campaign is stated in their paper. In [77] the results of CFD simulations of experiments with a release rate of 0.37 kg/s are described. By accounting the heat transfer to the ground the agreement between experiments and simulations was significantly improved and the maximum concentration was in most cases predicted within a factor 2. Later, Venetsanos [78] used the same CFD model (ADREA-HF) to investigate the effect of some parameters in reproducing the NASA experiments [79, 80] with a release rate of 9.5 kg/s. By modelling the source as a two-phase jet (compared to a pool) and including the heat transfer to the ground, the best agreement between experiments and simulations was achieved. The observed discrepancies were attributed to wind meandering that was not modelled and to a low value of the heat flux from the ground. Few years later in [81] the same CFD modelling strategy was applied to the numerical analysis of LH<sub>2</sub> release and dispersion in a mock-up re-fuelling station, investigating the effect of wind direction and the presence of an obstacle on the flammable mass and volume. The NASA experiments [79, 80] were also used for validation of a LES method [82].

Another challenge in simulations of LH<sub>2</sub> is the modelling of the source term. Experimental observations have indicated that the flow is already two-phase at the exit orifice [76, 77]. In their CFD analysis (FLACS) of the HSL/HSE experiments [71, 72] (release rate = 0.071 kg/s), Ichard [83] performed a sensitivity study, increasing the gas volumetric fraction VF of the source term from 0.76 to 1. They achieved the best agreement with volume fraction (VF) equal to 0.99. In [84], the simulated maximum concentrations were within a factor of 2 compared to the experimental data, as well as in CFX code validation [85] against the NASA experiments. Recently in [86] a validation against the HSE/HSE experiments [72] and a sensitivity study on the percentage of liquid/gas H<sub>2</sub> at the source term for 100%, 75%, 50% and 25% of the release mass flow were carried out. They achieved the best agreement with the experiments in term of pool distribution with 75% and 50%. Nevertheless the authors stated that the temperature distribution at the wall and the pool front velocity were not in the range or close by the experimental data and that they need further investigation.

Besides CFD studies analytical models were developed to describe specific stages of the liquid hydrogen release. Kim [87, 88] found that the perturbation method provides nearly identical results to the numerical solution when third order perturbation solutions are considered for the pool volume. The governing parameter is the evaporation rate. Epstein and Fauke developed a top-hat jet/plume model to obtain simple closed-form expressions for the total mass and volume of the flammable cloud for a gas or volatile liquid release [89]. Harstadt and Bellen [90] proposed analytical expression for the minimum pool evaporation time for the H<sub>2</sub> film-boiling rates. Verfondern and Dienhart [68] developed a computer model LAUV to simulate the spreading and vaporization for different grounds (solid or water) showing a satisfactory agreement between the model and the experimental data. A homogeneous non-equilibrium, two-phase, critical flow model, the homogeneous direct evaluation model (HDE), was developed in [91]. The model was validated with NASA data for liquid and supercritical hydrogen, methane, nitrogen, and oxygen in terms of critical mass fluxes for a range of stagnation conditions. Houf and Winters [92] developed a series of models to describe the whole release process of a small and slow leak (at very low Mach number) from a LH<sub>2</sub> storage system based on NIST REFPROP subroutines [93]. Later they [94] developed a similar multi-zones model for high-pressure liquid release, adding a model for the zone of under-expanded flow. The only validation is for the model for gaseous hydrogen leaks, demonstrating a favourable agreement between the model and the experiment for H<sub>2</sub> concentration along the centreline.

Li [95] used the PHAST software to calculate the harm-effect distances of LH<sub>2</sub> releases and of cryo-compressed hydrogen releases [96]. Their analysis and results have a high level of uncertainties because PHAST is based on simplified models and correlations that do not take into account all the relevant parameters, are not valid for all range of possible conditions and hazardous materials and have very limited examples of results validation for hydrogen.

The CFD capabilities of predicting hydrogen concentrations seem to be accurate within a factor 2 in some analyses [77, 84, 85]. Although a factor 2 is considered as acceptable in other fields, for the specific H<sub>2</sub> applications it could not be acceptable. Overestimating or underestimating concentrations by a factor 2 can cause a much larger discrepancy in the calculations of the overpressures that can be generated by potential explosions. For LNG there already exists a Model Evaluation Protocol for the assessment of models for dispersion: “the protocol comprises scientific evaluation of the numerical and physical basis of models for the dispersion of LNG vapour, model verification, and validation; ... A supporting suite of validation data, and guidance on the use of this data, has also been produced” (citation from [97]). The Fuel Cells and Hydrogen Joint Undertaking (FCH JU) co-funded SUSANA project is starting in 2013 with the target of developing a Model Evaluation Protocol for hydrogen, including a validation matrix for CFD simulations.

## **ACCIDENTAL IGNITION**

It is now widely accepted that accidental releases of pressurised hydrogen could lead to spontaneous ignition. Astbury and Hawksworth [98] postulated four potential ignition mechanisms: the reverse Joule–Thomson effect, the electro-static ignition, diffusion ignition (ignition behind a shock wave) and hot surface ignition and the possibility that two or more of these mechanisms could be present together. Subsequent research since then has ruled out the reverse Joule–Thomson effect as a potential trigger for spontaneous ignition, but found evidence of all the others. The majority of the published body of research has focused on diffusion ignition while there are few papers which addressed alternative mechanisms.

### **Ignition by electrostatic discharge**

Hooker et al. [72] investigated experimentally electrostatic ignition by corona discharge. They found that H<sub>2</sub>-air mixtures can be ignited by the discharges of a potential of several tens of kV above the surrounding atmosphere. Such situations could be expected at the top of tall vent stacks, tens of metres above ground, in the presence of large atmospheric electric fields (e.g., during snow fall). The incendive corona discharges which would ignite the released hydrogen were thought to be unlikely in horizontal releases of H<sub>2</sub> close to ground level. They also found that dispersion of dusts up to 160 g with H<sub>2</sub> released from 200 barg did not appear to generate hazardous electric fields. Merilo et al. [99] found that even a small quantity of entrained particulates can be a source of spontaneous ignition. They believed that both electrostatic discharge and corona discharge could be responsible for some of the ignitions occurred in their tests. They found that significant quantities of iron oxide particles were entrained, and a sharp-pointed ungrounded conductor was charged to a high potential with no ignition occurring. It led to the comment that an incendive brush discharge might be unlikely to form due to the quantity of particulates required to generate high electrostatic potential and it is unlikely that a typical hydrogen application will have this quantity of particulate present internally. This is consistent with the findings of Hooker et al. [100].

Imamura et al. [101] investigated the effect of the outlet shape on the possibility of the ignition by electrostatic discharge at a ventilation duct outlet. Four types of outlets were designed: outlet A (6.35 mm pipe), outlet B (12.7 mm pipe), outlet C (25.4 mm pipe) and a tapered porous outlet (called TP outlet in this paper). Iron (III) oxide particles were used as the model dust. It was clarified that if the ventilation duct outlet is grounded, most of the electrostatic charges are not generated, but not all of them can be removed by grounding only. However, the voltage and energy of the mixture at a certain downstream position can be reduced by using the TP outlet.

### **Ignition by mechanically generated sources**

Welzel et al. [102] investigated the ignition by mechanically generated sources for two different hydrogen/air mixtures. Their work pointed out limiting power densities for the ignition of 10% and 30% hydrogen/air mixtures in friction processes with mild and stainless steel. Limiting values in 10%

hydrogen/air mixtures are lower than those in 30% hydrogen. Limiting power densities for stainless steel are lower than those for mild steel.

### **Diffusion ignition**

Recent experimental studies have been conducted to demonstrate diffusion ignition of pressurized hydrogen release through a tube [103-112] for free jets. The releases all passed through a section of tubes and bursting disks were used to initially separate the pressurized hydrogen and air. Both Golub et al. [105] and Mogi et al. [108] found that the minimum release pressure required for spontaneous ignition depends on the tube length. In general, the experimental results suggest that the propensity to spontaneous ignition increases with the increase in reservoir pressure, tube diameter and length. As the tube length increases, the minimum release pressure required to trigger a spontaneous ignition was found to decrease. Results of Golub et al. [105, 106] suggested that the cross section shape of the tube is of importance because it affects the flow boundary layer. They showed experimentally that at the same cross section area the spontaneous ignition in narrow rectangular tube occurred at lower pressure than that in round tube. At the initial pressure in high-pressure reservoir lower on 1.5–2 MPa (shock wave pressure lower on 0.2–0.5 MPa) spontaneous ignitions occurred with the rectangular tube. Using photodiode signals and flame images, Lee et al. [112] observed the propagation of a flame inside the tube is confirmed and the flame is detected near the rupture disk as the bursting pressure increases. When the tube length is not long enough to produce self-ignition, a hydrogen flame was observed only in the boundary layer at the end of tube and it quenched after the flame exited the tube. It was postulated that the formation of a complete flame across the tube is important to initiate self-ignition, which sustains a diffusion flame after jetting out of the tube into the air. Kitabayashi et al. [111] found that the storage pressure sufficient for spontaneous ignition was a function of the tube length with characteristic minimum at about 3.8 MPa for tubes with 10 mm internal diameter. Below this critical diaphragm bursting pressure and with longer or shorter tubes than the length of about 1100 mm, no ignition occurred.

Golovastov and Bocharnikov [113] studied the influence of rupturing process at a pulse discharge into an open channel. It was shown that the possibility of self-ignition is defined by H<sub>2</sub> initial pressure and diaphragm rupture rate. In the range of initial pressures 5.0–14.0 MPa the rupture rate was varied from 5 to 20 μs. More rapid opening of the diaphragm accelerated the formation of the shock wave and led to rapid heating of the air behind the shock front. The given dependence was found practically did not depend on initial pressure in the range indicated. Ignition delay can be decreased up to 23 μs behind the shock wave. These findings were found to be in line with the numerical predictions of Xu et al. [114] for release into air without channel. In the recent flow visualization study of Kim et al. [115], it was found that the flame ignited at the boundary layer follows up the mixing front and spreads to the mixing tail of the mixing zone as the shock wave moves downstream. It is presumed that the flame along the boundary layer is turbulent induced flame, based on the glinting of the flame along the boundary layer. The experimental findings have consistently indicated that the bursting disk rupture process has an important influence on the mixing and ignition through multi-dimensional shock formation, reflection and interactions. However none of the experimental groups investigated in detail the influence of different internal geometries, which was postulated to have some influence by Dryer et al. [103] and subsequently the subject of several numerical simulations [116, 120].

Any accidental releases, in practice, would often involve releases through a section of a tube. Large due to this reason, most subsequent numerical studies have focused on this type of release scenarios. The independent studies of several groups [104-107, 116-125] have all identified that the air behind the leading shock is shock-heated and mixes with the released hydrogen in the contact region. Ignition is firstly initiated inside the tube and then a partially premixed flame is developed. Significant amount of shock-heated air and well developed partially premixed flames are two major factors providing potential energy to overcome the strong under-expansion and flow divergence following spouting from the tube. Yamada et al. [111] also identified generation of vortices behind the shock in a long tube; the later led them to postulate the possibility of an auto-ignition induced by vortices. Parametric studies conducted by Wen et al. [116] revealed that the rupture process induces significant turbulent

mixing at the contact region via shock reflections and interactions. Their further work [114] revealed that if the tube length is smaller than a certain value for a given tube diameter, even though ignition could take place inside the tube, the flame is unlikely to be sufficiently strong to overcome under-expansion and flow divergence after spouting from the tube and hence is likely to be quenched. This was later confirmed by the experiments of Golovastov and Bocharnikov [113]. Xu et al. [119] investigated the effect of a thin flat obstacle on the ignition and found that the presence of the obstacle plays an important role in quenching the flame following spontaneous ignition. More recent investigations of Xu and Wen [120] examined the effect of local contraction within the tube. They found that a local contraction can significantly increase the propensity of spontaneous ignition by producing elevated flammable mixture and enhancing turbulent mixing from shock formation, reflection and interaction.

Bragin and Molkov [123, 124] numerically demonstrated the transition from spontaneous ignition inside the tube into a sustained jet flame. They reproduced the experimentally observed phenomenon of flame separation and suggested that the transition to the sustained jet flame is largely dependent on the initial stage, where the developing annular vortex pushes the combusting mixture upstream into the recirculation zone. Once the flame is stabilised near the tube exit, it acts as a pilot flame and ignites a jet fire later on. Bauwens et al. [125] and Maxwell et al. [126] investigated theoretically partially confined jets and found that the ignition limits of H<sub>2</sub> releases into confined environments depends strongly on the strength of the shock that is driven into the oxygen/air ahead of the jet and size of the release orifice.

### **Catalytic ignition**

In order to determine the potential of the ignition in the presence of a catalyst, Brady et al. [127] experimentally investigated lean pre-mixed hydrogen/air mixtures using a stagnation-point flow configuration against a platinum surface. They observed two distinct regimes - catalytic surface reactions and gas-phase ignition and demonstrated dependence catalytic reactions on surface heating. Their findings indicated that ultra-lean hydrogen/air mixtures can be ignited even in the absence of external heat addition, thus confirming a fire safety risk even at room temperature. Further investigations by Brady et al. [128] indicated that catalytic ignition occurred after the leaked H<sub>2</sub> comes in contact with the catalyst having near room temperature. After ignition, the surface temperatures were stabilized in the range of 600-800 K.

## **COMBUSTION**

Depending on the hydrogen mixture characteristics, such as concentrations, temperature, pressure, and flow geometry, the combustion process can undergo strong flame acceleration and /or deflagration-to-detonation (DDT) transition. Dependence of the potential danger of the combustion process appeared to be very sensitive to the geometrical conditions of the processes, mostly to the confinement and congestion level within the enclosures. Other factors are, indeed, quite important, but below the consideration will be concentrated on the geometrical factors and partially on the mixture properties. Currently a unified physical model and corresponding numerical instrument which can be used over the entire range of phenomena is not available. Detailed clarification of the combustion physical nature and creation of the simplified engineering models for the separate phenomena as well as comprehensive numerical models, allowing predictively simulate the whole sequence of combustion events during possible accident is a continuous challenge for the researchers.

### **Influence of confinement**

As it was found already in the early studies, the possibility of the flame to accelerate is very sensitive to the confinement of the volume. The combustion of the fully confined volumes is relatively good understood and its effects can be quite successfully predicted by modern combustion models [129].

The necessity to take into account more practical configurations initiated consideration of the combustion in the volumes with one or more vent areas of varying shapes. Starting with the channel-like experiments [130] it was shown that transverse venting can substantially reduce flame speed and in case of detonation even cause its failure. Further results of the influence of the transverse and longitudinal venting were obtained by several research groups [131-133] and the criteria for the evaluation of the flame acceleration risk were proposed [134]. Further extension of the vent influence studies involved investigation of the semi-confined volume, such as horizontal flat layer of the H<sub>2</sub> distribution limited from top [135-138], where new engineering correlation accounting for the layer thickness, obstacles' interval and blockage ratio was introduced. Similar practically important configuration which however has too little data available is a one-side bounded semi-confined vertical flat layer. Such configuration can be easily realized when, for example, a jet impinges vertical wall and flows along the wall. Clarification of the possibility of the strong flame acceleration and DDT in the fully unconfined space (recent example, e.g. [139]) still waits for the augmented attention.

Another practically important phenomenon is *vented explosion*. Experimental data of different scales and boundary conditions (e.g., [140-145]) and theoretical models are available in the literature. Proposed by NFPA a standard [146] on vent sizing is basically focused on the natural gas and in [145, 147] it was found that the model is over-conservative and for the stoichiometric hydrogen mixtures is not directly applicable. S. Jallais in [149] has shown that the recommendations from [147] have limited validity, while model of [148] globally provides good accuracy with slight overestimation. Most of the existing models are targeted to estimate only pressure peak disregarding other parameters, such as ignition location, obstacles, etc. The analytical expression presented in [150] allows calculation of the both pressure peaks and takes into account many of influencing factors. In [149] it is reported that the correlation is adequate for the hydrogen vented explosion volumes from 1 m<sup>3</sup> to 120 m<sup>3</sup> and H<sub>2</sub> concentrations from 10% to 30% vol. In the currently on-going EC project HyIndoor [151] a systematic study of the venting methodology is undertaken and a formulation of the improved correlations and CFD numerical tools are expected to be proposed. Accounting of other confining factors, such as covering vent grid, relative localization of the vent, and particularly important vent cover inertia are still not properly addressed. Comprehensive overview of the state-of-the-art on explosions with inertial vent covers is available in [152], which among others names the following most significant knowledge gaps: little data on the inertia effect; the recommendations for open and non-inertial vents require specific validation; no data on the turbulence generation by vent outflow.

### **Influence of congestion**

In numerous studies it was shown that the details of the obstacle configuration can decisively influence on the regime of the combustion. One of the main parameters which are commonly used for the obstruction characterization is blockage ratio. However other geometrical characteristics can and actually affect the combustion process as well. Already in [153, 154] the different geometrical forms were studied (tube bundles, gridiron, plate with rectangular opening): it was shown that flame can accelerate for all blockage ratios. Influence of the variation of blockage ratio, distance between obstacles, imitation of rough walls, etc., were studied in the works of Teodorczyk [155, 156]. Global quenching was observed in the vertical facility with partially obstructed channel [157]. Parametric study on the evaluation of limits for effective flame acceleration in obstructed closed geometries was carried out in [134, 158]. Deeper understanding of flame acceleration was obtained after remarkably detailed CFD simulation in [159]; however it is not yet possible to state that everything is fully clear and therefore further efforts are continuously undertaken to improve the knowledge and understanding of the role of obstacles. Among recent studies, for example, in [160] an onset of detonation behind a single obstacle was studied; and in [161] a possibility to simulate an onset of detonation using different techniques was considered. In the frames of the EC project HyPer [162] a study of leakages and releases followed by combustion inside a generic fuel cell cabinet for a range of leak rates, blockage ratios and vents were investigated. On the basis of the experimental work of [136-138] on the flame acceleration in flat layer, an attempt to generalize utilization of the congestion characteristics using numerical simulations was made in [163]. The correlations proposed in [138] considers



dependence on blockage ratio, distance between obstacles and layer thickness, while in the numerical experiments of [163] most of possibly significant geometrical parameters of the layout were taken into account. It is well known that rough tubes without material obstacles inside them and even smooth tubes are able to promote FA and DDT [164]. It was found that consideration of the boundary layer can give practical results for unobstructed tubes [165].

Note, that the most of the studies are made for artificially created obstacle sets, such as repetitive periodic grids, circular orifices in the tubes at the constant mutual distance, etc., while the real industrial configurations will definitely include the obstacles irregularly placed in the volume with the very different characteristic sizes. Further analyses in this direction would have considerable practical outcome.

### **Mixture properties**

Non-uniformities of the gas distribution can considerably affect regime of combustion, however, only limited amount of the experimental data are available on the behaviour of the H<sub>2</sub> in the presence of the concentration gradients (e.g., [166, 167]). In the recent studies [168] the data on the flame acceleration of lean H<sub>2</sub>-air mixtures with vertical concentration variation and comparison of the obtained flame speeds with those in uniform mixtures were provided. Evaluation of the combustion regimes in the stratified horizontal layer of the hydrogen-air mixtures were performed in [169, 170] (with a hypothesis of the decisive role of the maximum concentration in the layer) and the numerical study for the corresponding regimes is presented in [171]. Large scale vertically stratified mixtures were experimentally studied in [172]. In [173] the results of the benchmarking of the different CFD codes on the basis of the data from [168] are presented, the obtained results demonstrated that most of the available CFD codes still exhibit the lack of the predictive capabilities in case of the complicated initial conditions which includes mixture composition non-uniformity.

Intrinsic nature of the flames and supplementary events can promote numerous flame instabilities, leading to the considerably enhanced burning rates. The diversity of the possible instabilities, despite numerous experimental data available (e.g., [174-176]), and considerable success in the theoretical understanding of the governing mechanisms [177-183], constitutes the origin of the status that only limited successes were achieved in the creation of the unified approach to their modelling. Recent numerical simulations [184-186], which take into account the instabilities effect, demonstrate that essentially successful approaches should be further generalized with the view to provide established methods for the engineering CFD simulations. In [184] the simulation of the vented explosion introduces combustion model considering additional flame wrinkling which is described by a transport equation with the generation and removal of flame surface wrinkling. The relative simplicity and transparent physical basis of the method promise high potential for the use in applied simulations, however currently the method requires calibration which reduces the value from the standpoint of the immediate use of it. In [185], using numerical solution extending the theory of [183], study of the existence of spontaneous transition from the acoustic to the parametric instability and their growth rates were evaluated for a set of mixtures typical for hydrogen based applications. The accounting of acoustic instabilities was successfully utilized in benchmarking simulations [173], although some *backfitting* took place. A SGS combustion model of [180], which uses the flame area growth equation based on fractal theory, provided reasonable agreement with experimental data down to ~12.8% H<sub>2</sub> concentrations, although requiring further development for other mixtures.

### **HYDROGEN FIRES**

Hydrogen release might occur on storage, transport and handling. An ignition is likely for all forms of releases due to the wide range of flammability and low ignition energy. The type of storage or transport as well as the type of opening will guide the resulting fire, which can be a significant danger for the surrounding objects and humans. The investigations of fires use video/camera techniques, species sampling of the flame, temperature measurement and measurement of the emitted radiation. A pure hydrogen fire is nearly invisible (only weak water bands in the red spectral range), radiation is

mainly emitted in the UV by OH-bands. Water bands cover the NIR and MIR spectral region. However, real hydrogen flames often include impurities or entrain them on propagation, resulting in addition of lines, bands and (Grey body) continua.

### **Jet fires from openings in storage containers or broken fittings**

Releases from small leakages from hydride tanks generate normally a low momentum and ignition forms sustained laminar or turbulent diffusion flames. Detailed investigations occurred at laboratories to derive burning rates, species and temperature profiles. The results provided a good basis for hydrogen combustion mechanisms, fire modelling and CFD simulation. The data on flame velocities depending on pressure are still limited to relatively low pressures (<3MPa) [187-190], despite high request for such data exists. Emission spectroscopy has been already applied to small scale turbulent hydrogen diffusion flames [191, 192] and those under pressures up to 3 MPa [188], correlated to flame evolution. Simultaneous water vapour concentration and temperature measurements were performed in transient hydrogen flames [193]. For extremely high pressures up to 200 MPa the flames were studied applying UV-Vis emission spectroscopy with analysis of the OH-bands for temperature estimations with diatomic band evaluation method (see ref. [194, 195]). Still the detection and identification of small hydrogen releases and beginning fires are difficult [196-199].

Momentum driven jet fires can be generated from high pressure storage, liquid tanks, or fuel cells. These jets might impinge constructions or humans up to 15 m, depending on vessel pressure and mass flow rate. During jet head propagation entrainment of air can create conditions for the early ignition which is very likely will develop into a fire [200-202] or a jet-fire [203-208] depending on the impetus. In case of accumulation, delayed explosion can occurs. Under-expanded jets can be extremely dangerous, as they may increase in size and keep higher temperatures, downstream. The typical research studies are measuring the length and width of the fire and the radiative properties depending on the initial pressure in the tank and the opening diameter. Several studies are performed on the effect of barriers on the fires, e.g. [218, 219].

The H<sub>2</sub> jet after ignition might establish a sustained turbulent flame, developing in two highly transient steps: starting from the ignition point the flame propagates simultaneously downstream and upstream followed by the flame pulsating before flame stabilization. In case of delayed ignition the first phase can develop as a gas explosion. For high initial mass flow rates (> 400 g/s) the apparent flame velocities might approach near sonic speeds and generate substantial pressure waves (see visualization by BOS-method [205-208]). The molecular band modelling code BAM from Fraunhofer ICT (details see [209-211]) enabled the time resolved evaluation of the species concentrations and temperatures using robust fast scanning IR-Spectrometer [212, 213] and analysis by a least squares fit procedure recording also fluctuations. Temperatures found are close up to 2000 K with transient zones up to 2400 K [210, 212]. The results of Houf and Schefer [214] are obtained by estimates from the RADCAL code [215]. It is interesting that the strong CO<sub>2</sub> band from entrained air can be modelled to estimate the air content at the point of view (ref. [212]). The exhaust from an opened liquid hydrogen tank [216] led to the initiation of pulsating transient jets subsequently overlapping and lasting some seconds. Generated pressure waves were moderately in the kPa range [210, 216] with temperatures typically 2000 K with some hot spots up to 2400 K [205, 210, 214]. Sound levels and radiation measured in [217] for quasi-stationary jets from liquid reservoirs give some doubts of the adequacy of the reliability of their results.

Under laboratory conditions at small scale jet fires are well understood, however at larger scales in jet profiles there might occur deviations. It was shown, that the air entrainment can be obtained by fast IR-spectroscopy analysing the strong CO<sub>2</sub> band (wavelength 4.25 μm) [211, 214]. It would be wise to apply imaging spectroscopy resolving fluctuations to the jet fires to get a full spectral picture in comparison to the video analysis. Realistic integrated radiation can be derived only by this approach and simulated by obtaining temperature and species distributions over the jet fire applying codes like the RADCAL [214, 215] or the BAM [209-212] codes. For visualization the BOS (Background Oriented Schlieren)-technique [205, 220, 231] should be applied which has been adopted by some

research groups [207-209], however certain improvements could be obtained by combining with brightness subtraction [205, 221] or one-dimensional frame compacting the latter provides transient flame profiles [205-209]. The use of barriers and walls for preventing jet impingement has to be carefully planned [218, 219].

Metal hydrides [222] might also meet some US DOE [223] system targets for automotive H<sub>2</sub> storage due to recent materials' development and basic hydride research but might be more appropriate for stationary storage. The hazards in such systems, however, can occur if the unpassivated light metals can get access to air: the nano-structured metal is immediately oxidized [224, 225] strongly heating the storage device. A destruction of the storage containers might be expected generating a highly pyrophoric cloud in hydrogen [226-230].

### **Contours and radiation from hydrogen fires**

Currently, high speed video techniques have mainly been applied to record the shapes of emitting fires, however, shapes of flames have to be recorded by seeding tracing material into the flame. In technical flames, these are sodium or soot as "dye" from impurities for the "invisible" hydrogen fires. *Schlieren* techniques can make the structures of the flames visible [231]. The BOS, background oriented Schlieren technique, can visualize the shapes of H<sub>2</sub> fires and also their detailed structures, including statistical data of turbulence [203-208, 220, 232]. There are indications that such technique can reveal flame structure details similar to *Schlieren* technique, identify air entrainment and blast waves [205-209], especially when using additionally brightness subtraction methods [205, 221]. A comparison with detailed imaging spectroscopy and CFD simulations could outline the correlation to real turbulent effects.

Beneath of direct contact with the flame, radiation is the second important stand-off effect. Radiation intensity depends on species, duration in the hot zones and the temperature distribution. Whereas in laboratory experiment all types of modern spectroscopic methods (UV-Vis-IR-, Raman Spectroscopy, CARS, LIF, PLIV, etc.) were applied, for large scale H<sub>2</sub> fires robust emission spectroscopic methods have to be preferred. Such fires are band emitters with temperatures reaching to 2500 K, predominantly bands of OH in the UV-spectral range (band maximum 309 nm) and of H<sub>2</sub>O in the NIR and IR spectral range (various bands in the wavelength range from 0.64 - 9 μm with varying emissivities at maxima). Since the different bands approach the intensity of a Black Body radiator at different wavelengths of band maximum for large fires [233], commonly used pyrometers are useless. To measure the air entrainment [212], CO<sub>2</sub> with its strong band at 4.25 μm can be used. Impurities contribute single atomic lines (Na, K, Li, Ca, Fe etc.) and continua (mainly soot from organic contaminants) [218]. For measuring turbulent structures or transient phenomena the fast scanning spectrometers should be used. Currently, robust FTIR-spectrometers are available for fast scanning and imaging spectroscopy which provide higher wavelength resolution. To study the OH-band in the UV there have been already OMAs (Optical Multi-Channel Analyser) used for more than 30 year with time resolution of 10 ms and more [188, 195, 195, 209, 211]. The evaluation of the 3-atomic molecules uses the "Handbook Infrared Spectra of Hot Gases" which gave the basis of Hitran, Modtran and Lowtran- code series of the NASA [234, 235]. These codes were made applicable for every-day use by the codes RADCAL code [214, 215] and the BAM code, in addition, is able to perform a least squares fit procedure of spectral data (including also Grey Body soot emission) and has been applied in numerous cases (see e.g. ref. [209-212, 233]).

The radiation emitted from the fire ball which contains also some OH radicals strongly interacts with the radical OH, which is effectively true as it shows strong self-absorption and even more with H<sub>2</sub>O. Further molecules play important roles in the reaction mechanism like H<sub>2</sub>O<sub>2</sub> (see e.g. ref [192, 236]) and HO<sub>2</sub>, the reaction fronts being exposed to the intensive radiation of the H<sub>2</sub>O fireball interior) and might therefore modify the reaction mechanism used in combustion modelling. It might be found that the radiation emitted from the fire ball interacting with flame front species contributes also substantially to the increase of burning velocity depending on the size of hydrogen-air mixtures (jets

and clouds) as compared to self-induced turbulence. Including chemical kinetics into CFD- modelling might contribute to an improved understanding of DDT.

## IDENTIFICATION OF KNOWLEDGE GAPS

On the basis of the survey made by the participants of the Research Priorities Workshop held by HySafe the following issues can be qualified as having highest priorities in availability of experimental data, CFD models, and engineering correlations.

- Effects of surfaces on jets (attached jets, impinging jets), multiple jets, and the shape of a nozzle on the release (slits, elliptic orifices, rectangular orifices, effects of the aspect ratio of asymmetric orifices on the scaling laws) including flapping sources.
- Detailed validation of notional nozzle theory (proximity to a surface, effects of buoyancy, effects on the lateral concentration distribution, nozzles with small diameters).
- Effect of weather conditions on the release including cryogenic, e.g. humidity, temperature, wind speed and direction, atmospheric stability class.
- Universal scaling law for the flammable extent of jets.
- The physical properties of liquid hydrogen and gaseous hydrogen at very low temperature (also of O<sub>2</sub>, N<sub>2</sub>, H<sub>2</sub>O – close to saturation) including differences with the ideal gas law, cryogenic jets, effect of buoyancy and turbulence for jets and spills, critical conditions for FA and DDT in cryogenic hydrogen-air mixtures.
- Phase change for cryogenic jets and spills such as the hydrogen evaporation and the condensation and solidification of nitrogen, oxygen, and water in the air.
- Conductive, convective and radiative heat transfer between the cold hydrogen and the surrounding environment including air and the ground.
- Limited studies have been carried out for accidental ignitions caused by mechanically generated sources.
- The effect of tube internal geometries on the propensity to spontaneous ignition.
- The effect of nozzle shape, especially non-circular nozzle, on the propensity to spontaneous ignition.
- A database on the probability of hydrogen spontaneous ignition for typical release scenarios similar to that available for hydrocarbon fuels.
- Flame acceleration and deflagration-to-detonation transition in a semi-confined to open geometries.
- Vent sizing methodology, including high pressure releases in enclosed areas (with natural or forced ventilation) and effect of vent cover inertia on vented deflagration dynamics.
- Effect of obstruction characteristics on flame dynamics (acceleration/deceleration) and DDT for different confinement, including global and local quenching phenomena in different geometries and scales for premixed and partially premixed cases.
- Effect of hydrogen concentration gradient and stratification on flame dynamics (acceleration/deceleration) and DDT for different confinements.
- Flame instabilities (acoustic, parametric, Rayleigh–Taylor, Kelvin–Helmholtz, Richtmyer–Meshkov, Landau–Darrieus) and their effect on the flame dynamics including scaling conditions.
- Experimental data for the transient pulsating jet fires from liquid hydrogen or tanks at high pressures, further development of high resolution optical method, as e.g., scanning spectroscopy.
- Basic investigations of small scale hydrogen fires with materials, radiation, contact at various distances and large scale hydrogen fires with the view to develop reliable radiation models
- Effective fire ball scales, cooling down and movements, especially for large clouds, where cooling occurs mainly by radiation.

- Investigation of the hydrogen release from various types of currently favoured hydrogen storage materials and the effects of real storage containers, depending on loading status, operational state, ambient temperature etc., including hydride storage facilities.
- Further development of BOS technique: by synchronized 2 cameras, develop 3D BOS video analysis by comparison with CFD simulation, correlate 3D BOS with radiation shapes and CFD modelling including radiation transport.
- Constitution of a validation matrix for CFD simulations (validation against experimental data and inter-comparison), including turbulence modelling, combustion models, accounting of flame instabilities, and mesh sensitivity issues.

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