VALIDATION STRATEGY FOR CFD MODELS DESCRIBING SAFETY-RELEVANT SCENARIOS INCLUDING LH2/GH2 RELEASE AND THE USE OF PASSIVE AUTOCATALYTIC RECOMBINERS

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ABSTRACT
An increase in use of hydrogen for energy storage and clean energy supply in a future energy and mobility market will strengthen the focus on safety and the safe handling of hydrogen facilities. The ability to simulate the whole chain of physical phenomena that may occur during an accident is mandatory for future safety studies on an industrial or urban scale.

Together with the RWTH Aachen University, Forschungszentrum Jülich (JÜLICH) develops numerical methods to predict safety incidents connected with the release of either LH2 or GH2, using the commercial CFD code ANSYS CFX. The full sequence from the release, distribution or accumulation of accidentally released hydrogen, till the mitigation of accident consequences by safety devices is considered. For specific phenomena like spreading and vaporization of LH2 pools or the operational behavior of passive auto-catalytic recombiners (PAR), in-house sub-models are developed and implemented. The paper describes the current development status, gives examples of the validation and concludes with future work to provide the full range of hydrogen release and recombination simulation.

1.0 INTRODUCTION

Broad experience with the safe handling of hydrogen at a large industrial scale has been achieved in the past decades. Still due to its nature, hydrogen may accumulate with air under certain conditions and form an explosive mixture, therefore representing a potential danger to human beings and environment. This is a major concern for a growing hydrogen market. Transportation and storage of large amounts of liquefied hydrogen (LH2) has been demonstrated is economically feasible and may expand in future. Safety parameters, e.g. safety distances, for the industrial usage of hydrogen are, in principal, not directly transferable to the handling of hydrogen in an urban environment. In case of accidental release of hydrogen, an immense damage of the surrounding due to a possible explosion could be expected. Besides the burnable hydrogen-air mixture resulting from vaporization of liquid hydrogen (GH2) into the atmosphere, LH2 pool spreading also carries a significant hazardous potential to human beings and materials due its cryogenic character. Several phenomena during the spreading of the LH2 and its distribution have to be understood, modeled and validated for reliable and trustworthy safety analyses.

LH2 at atmospheric pressure has a temperature of 20.3 K. Releasing LH2, either accidentally or on purpose during an experiment, the liquefied gas startsinstantaneously to vaporize. The maximum pool distribution is mainly driven by it’s the vaporization rate. The main influence factor of the vaporization rate is the boiling regime which occurs between the liquefied gas and the wall [1], where it is spread. A further important factor of the vaporization rate for the energy conservation is the bulk vaporization. It includes both the condensation of GH2 in the subcooled LH2 pool, and the heat
transfer from the “hot” environment into the “cold” pool and the phase change of the further contributing fluids, e.g. water, oxygen and nitrogen.

The boiling process is dominated by the temperature difference between wall and pool temperature. The wall temperature will decrease, because the heat flux used for the boiling process cools down the wall and the amount of transferred heat is directly proportional to the wall temperature. At the edge of the pool, film boiling is expected, which will change towards transition boiling and then to nucleation boiling depending on the time, since when the wall is in contact with the LH2 pool. Afterwards the vaporized hydrogen will be distributed into the atmosphere, driven by external momentum such as wind, its buoyancy and diffusion. This distribution of gaseous hydrogen consequently forms a cloud where an explosive mixture of air and hydrogen might develop. Due to its relatively low ignition energy this volume might be ignited sooner or later.

Ignition of an explosive mixture might result in major damages. The ability to predict the distribution of the explosive gas and simulate all influencing factors correctly, is essential for the risk assessment of worst case scenarios. The correct distribution helps to identify technical measures to avoid any risk of ignition, such as defined air volume exchange, the usage of explosion proof systems and components or the usage of passive auto-catalytic recombiners (PAR) in confined spaces and/or safety distances for releases into the environment.

Aim of this work, besides the extensive modeling of physical phenomena, is to prove that such an integral simulation is possible with commercial CFD codes, like ANSYS CFX. Therefore missing models for the identified contributing phenomena need to be developed. The other main part of the work is focused on the validation of the code against experiments. This validation is not easy to perform, because not all phenomena are completely understood and separately measured. Therefore only integral experimental data is available.

Either generic or realistic accident scenarios in confined spaces such as underground storage facilities at fueling stations, garages or confined areas in the direct vicinity to large storage facilities with a direct link to the storage fluid are focused in the research work at JÜLICH. The rupture of piping, valves, pumps, etc. in the confined spaces is expected to be a realistic initialization of an accident sequence and hydrogen is released, until safety devices stop the release. Another main objective for the research work is the mitigation of such accidents consequences. Therefore, a special focus is on effects using a passive auto-catalytic recombiners (PAR) as a passive safety device. This work forms the basis for a consequent analysis of possible combustion loads which are however not objective of this paper.

2.0 MODEL DEVELOPMENT

Fig. 1 shows a generic accident operational chart. Either a gaseous, liquid or a multiphase source term is possible. For a multiphase release source term it is most likely that a spontaneous vaporization, i.e. due to isenthalpic expansion, at the point of release occurs. The remaining liquid is spread via a pool onto the surrounding area and will consequently start to vaporize. The spontaneously formed and vaporized gaseous part of hydrogen will distributed mainly by buoyancy driven. An accident of a high pressurized storage facility will lead to a momentum driven distribution of the gaseous hydrogen. Based on this hydrogen transport and mixing phenomena burnable hydrogen-air mixtures may form which could either ignite or be mitigated due to inertization, venting or recombination.

The ability to simulate all parts of this generic accident scenario is mandatory for the safe handling of hydrogen infrastructure and is part of the research work at JUELICH. The modeling and simulation work clearly does not include ignition or detonation simulation, because the prevention and mitigation of possible fatalities has to be the aim for the hydrogen infrastructure.
2.1 Pool distribution and wall vaporization rate of liquid hydrogen

Chitose et al. [2] and Verfondern et al. [3] have shown in their work that especially for the cryogenic pool distribution the wall temperature is a main factor. Consequently there is the possibility to determine the pool distribution by using the boundary wall temperature.

The analytical basis has been developed by Brentari et al. [4] in 1965, who has shown that there is a correlation between Kutdaladze’s nucleation boiling equation, the Breen & Westwater film boiling equation and experimental data for the boiling of hydrogen. Based on this information, Brentari was capable to draw a Nukiyama diagram for hydrogen, refer to Fig. 2.

The vaporization rate is mainly driven by the transferred heat due to boiling and is defined as
\[ \dot{m}_{ev} = \frac{\dot{q}^*}{\Delta h_{fg,e}} \]  

with \( \dot{q}^* \) the transferred heat according to the boiling regime and \( \Delta h_{fg,e} \) the effective latent heat of hydrogen. According to Fig. 2 the difference between fluid and wall temperature defines the amount of transferred heat.

Based on these principals, a wall vaporization model was developed at JÜLICH [5], which predicts, in principle, the dimension of a cryogenic pool using the HSL experiments.

In the course of the model development a main factor influencing the wall temperature and thus the vaporization rate has been identified: the wall heat conduction coefficient. High heat conduction coefficients of the solid material lead to a higher heat flux from the areas farther off the interface between the liquid cryogenic pool and the wall. Consequently, the boundary wall temperature is higher as well as the vaporization rate. For lower heat conduction coefficients, the gradient of temperature in the wall closer to the interface area is higher, the temperature therefore lower as well as the vaporization rate.

2.3 Phase change of hydrogen

Dienhardt [1] has shown in his work that the main contributor for LH2 pool distribution is the transferred heat from the wall (~97%). Other factors are the heat transferred by convection from the air (~2%) and by solar radiation (~1%). Therefore the pool distribution the heat transferred in the fluid domain is not that important. On the other hand there needs to be a link between the phases. Gianassi et al. [8] showed that the gaseous distribution is mainly influenced by phase changes of the containing fluids, such as water (solidification temperature: 273.15 K at atmospheric pressure) in humid air and in wall materials and the components of air, oxygen (boiling temperature: 90.2 K, solidification temperature: 54.8 K at atmospheric pressure) and nitrogen (boiling temperature: 77.35 K, solidification temperature 63.05 K at atmospheric pressure). The released latent energy of these fluids increases the heating of the gaseous hydrogen and affects directly its buoyancy.

For a coupled modeling of the liquid and the gaseous distribution, a link is needed between these two phases of hydrogen. In several papers, e.g. Venetsanos et. al [6] and Middha et. al. [7] it has been shown that a thermodynamic equilibrium model for a volume element is an accurate enough approach to predict the bulk vaporization. Based on their work it is planned to implement an enthalpy-based equilibrium approach for a volume element into ANSYS CFX. It is expected that this approach has the advantage of being expandable also for other contributors, such as water, oxygen and nitrogen, at least for the thermal energy in a fluid domain. The bulk vaporization rate, the corresponding latent heats are calculated using such a model for one discrete volume element in the fluid domain. Source and sink terms are applied to fulfill the demand of conservation of energy and at least the conservation of mass for the hydrogen part of the fluid domain.

To obey the conservation of mass, liquefied and solid fractions of oxygen, nitrogen and water have to be considered. At this stage this consideration is expected to be complex. The contribution of the latent heat of water for the buoyancy of the gaseous hydrogen cloud has been shown by Gianassi et al [8] and remaining solids have been found after conducting the HSL experiment by Hooker et al. [9]. The influence of the solids on the distribution of a pool and on the cloud cannot be quantified. Due to its complexity at this point, it is not possible to predict either liquid or solid motion of the mentioned substances.

Beside the implementation of the thermodynamic equilibrium model the correct modeling of the properties of the fluids, which take part in the simulation is also mandatory. At this point the property models based on the Helmholtz energy are considered to be the most accurate. In the past years, state equations using the Helmholtz energy description have been found pretty accurate and are widely
distributed and freely available in form of tables and the corresponding equations by the National Institute of Standard, USA [10] and for hydrogen described by Leachman et al. [11].

2.2 Gas dispersion of hydrogen

Gas distribution of hydrogen accidently or experimentally released is mainly driven by buoyancy. Especially cryogenic conditions and therefore real gas properties have an influence on the distribution. Also the correct property predictions are essential for a reliable simulation, which has been seen during the work for HySafe Standard Benchmark Exercise SBEP-V21, summarized by Venetsanos et al. [12]. The buoyancy or momentum gas mixing is predicted by applying models available in ANSYS CFX [13]. In general, a U-RANS approach, closed by the Shear Stress Transport (SST) turbulence model is used. The production and dissipation of turbulence due to buoyancy is considered by means of additional terms in the k and \( \omega \) equation. Dependent on the scenario different heat transfer mechanism, such as conjugate heat transfer, thermal radiation can be included. The approach is been widely validated in the frame of different projects or benchmark activities, e.g. Reinecke et. al. [14] which are mainly related to nuclear safety issues. The modeling and validation is performed under consideration of well-known best-practice guideline, e.g. ERCOFTAC (Casey & Wintergerste, [15]) or ECORA (Menter, [16]).

2.3 Recombiner modeling

Passive auto-catalytic recombiners (PARs) are widely used inside the containments of nuclear power plants (NPP) for the removal of hydrogen that may be generated during specific reactor accident scenarios (IAEA, 2001 [17]). Due to their ability to convert hydrogen and oxygen into water already at low (ambient) temperature, PARs provide a hydrogen sink even in situations where dilution and venting is limited or impossible. As a measure avoiding hydrogen accumulation and explosion, PARs are passive safety devices without the need of external power supply. PARs may be used in the future as safety devices inside confined areas for the removal of accidentally released hydrogen. In Reinecke et al., 2012, [14] it was demonstrated that PAR designed for hydrogen removal inside a NPP containment would principally work also inside a typical surrounding of hydrogen or fuel cell applications. In the study, a hydrogen release scenario inside a garage has been simulated with and without PAR installation.

The code REKO-DIREKT describes all relevant aspects of the operational behavior of PARs, i.e. start-up and hydrogen conversion under different boundary conditions. The code calculates not only the conditions at the PAR outlet (i.e. gas temperature and concentrations, mass flow) but also the local catalyst temperature and local gas concentrations along the catalyst sheets inside the PAR. The only input parameters required for the calculation are temperature and gas composition at the PAR inlet and the absolute pressure. The model is coupled by means of a data and program flow controlled interface routines to CFX and allows to study PAR operation inside compartments.

The principle of a PAR is illustrated in Fig. 3a. Inside the open bottom part of a steel housing, catalyst sheets form a set of parallel vertical flow channels. On the catalyst surface, hydrogen entering the PAR is converted with oxygen to water. Due to the exothermal reaction, a buoyancy-driven flow is induced inside the chimney part which ensures a continuous gaseous flow through the PAR. In order to assess the efficiency of PAR applications, substantial efforts have been spent on developing model strategies (Reinecke et al., 2010a, [18]).

Fundamental principle of PAR modeling is the interaction of the chimney and the catalyst section (Fig. 3b). For this purpose, the 2D code simulates all relevant heat and mass transfer processes inside the catalyst section (Böhm, 2006, [19]). A chimney model describes the mass flow through the PAR box due to density differences.
The reaction model of the heterogeneous catalytic reaction is based on mass transfer correlations. In this model, the conversion rate of hydrogen corresponds to the diffusion of the gaseous hydrogen through the boundary layer to the surface

\[ \dot{r} = \beta \cdot \Delta C_{H_2}, \]  

(2)

with the reaction rate \( \dot{r} \), mass transfer coefficient \( \beta \), and the concentration difference of hydrogen between bulk flow and surface, \( \Delta C_{H_2} \). The mass transfer coefficient \( \beta \) is calculated according to

\[ \beta = Sh \cdot \frac{D_{H_2}}{d}, \]  

(3)

by means of Sherwood laws

\[ Sh \sim Re^n Sc^m, \]  

(4)

where \( Sh \) is the Sherwood number, \( Re \) is the Reynolds number and \( Sc \) is the Schmidt number. The coefficients \( n \) and \( m \) are semi-empirical values which depend on the specific local flow conditions. \( D_{H_2} \) is the diffusion coefficient of hydrogen and represents the characteristic length.

3.0 VALIDATION

3.1 Validation Strategy

Due to the strong interaction of the different phenomena and due to the fact that mostly integral effect tests are available, the validation procedure is based on different experimental programmes and test series (compare Tab. 1).
Table 1. Available experiments and the observed phenomena.

<table>
<thead>
<tr>
<th>Test / Phenomenon</th>
<th>NASA</th>
<th>HSL</th>
<th>BAM</th>
<th>THAI (Becker Technologies)</th>
<th>REKO-3/4 (JÜLICH)</th>
<th>MISTAR (CEA)</th>
<th>PANDA (PSI)</th>
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<tbody>
<tr>
<td>LH2 release</td>
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<td>LH2 pool spreading</td>
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<td>GH2 cloud distribution</td>
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<td>GH2 transport and mixing</td>
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<td>X</td>
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<td>PAR atmosphere interaction</td>
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</table>

The CFD approach for predicting the LH2 spreading, the vaporization and the GH2 distribution is validated separately by means of different release experiments, conducted in the past years, like the NASA experiments [20] and the HSL experiments [9]. During both experiments LH2 is released into the environment. The distribution of the liquid pool for the NASA experiments is documented in several papers, i.e. [1]. For the HSL experiments, conducted in the UK, ground level thermocouple from 0.5 m every 0.1 m have been recorded the temperature. According to Dienhard [1] the liquid reaches the thermocouples, when they show the expected temperature of the liquid. For gaseous distribution several tower are aligned around the release point for both experiments. The concentration is the derived from temperature measurements. Additionally the concrete temperature is measured during the HSL tests, which might give indications about the heat flux from the ground into the pool.

The CFD approach for predicting the GH2 distribution is also validated separately by means of different experiments on various mixing mechanisms in small-scale facilities, like MiniPanda (ETH Zürich, Switzerland) and in technical-scale test facilities like THAI (Becker Technologies, Germany), PANDA (PSI, Switzerland), and MISTRA (CEA, France). In this context, especially PANDA tests performed in the frame of the OECD/NEA SETH2 project are used for validation. Within these so-called ST5 test series, the PAR behaviour is simulated by means of a heat source, and the effect of the hot exhaust plume on the atmospheric mixing and erosion of a light gas cloud is investigated (Mignot et al., 2012 [21], Andreani and Kelm, 2012 [22]). Similar tests will be conducted and used for validation in the frame of the EU-ERCOSAM project in the PANDA and also in the MISTRA facility (Paladino et al., 2012 [23]).

An on-going validation of RD (stand-alone) is performed by means of the separate effect test facilities REKO-3 (forced flow conditions) and REKO-4 (5.5 m³ test vessel) at JÜLICH (Reinecke et al., 2012
[24]; Simon et al., 2012 [25]). In technical scale, post-calculations of five integral PAR performance tests in the THAI facility have been performed with good overall results (Reinecke et al., Int. Mtg. Safety and Technology of Nuclear Hydrogen Production, Control and Management, 2010 [26]). A validation of the coupled RD-CFX approach is performed based on the OECD/NEA THAI-HR tests series (Poss et al., 2010 [27]). The test matrix includes 32 PAR tests investigating the operational behavior of three different commercial PAR units (AREVA, NIS and AECL) under free, unrestricted natural convection. It covers a broad range of relevant boundary conditions, e.g. elevated temperature and pressure levels, and different gas mixture compositions.

3.2 Validation of the liquid hydrogen distribution

In addition to the development of the models for simulating and mitigating possible accident scenarios the validation of the models is required. Available experiments used for validation of the distribution of hydrogen were performed by NASA [20] and HSL [9]. Due to the lack of detailed experimental data which measure the single phenomena, the validation of the each model itself is not possible. Only the global validation of all models is possible using integral experimental data such as from release experiments conducted by Hooker et al. [9] or Witcofski et al. [20]. For each single model itself, only plausibility checks are possible. It has to be noted that for both experimental data sets the inlet conditions are not fully known. A part of the inlet mass flow is vaporized instantaneously at the release point. The fraction of flash-vaporization hydrogen has been guessed by performing a parametric study and it was concluded that 25 % of the inlet mass flow to be immediately vaporized gives a plausible result.

To give an insight into a calculation and the contributors, an exemplary result of a pool distribution calculation is discussed. The results can be seen in Fig. 4a (for further information regarding the simulation, refer to [5]). The experimentally measured length of the pool is between 1.2 and 1.3 m after 30 s experimental time. In each figure the pool distribution can be found in the upper part and the temperature distribution of the wall temperature in normal direction (left) and the boundary temperature (right) can be found in the lower part. Fig. 4a shows the results using a homogenous heat conductivity of the concrete. In Fig.4b, the same calculation was repeated; the only difference is a higher (effective) concrete heat conductivity in wall interface normal direction. It can be seen that the pool length is lower and the reason can be found in the higher wall temperature at the interface between fluid and solid. The maximum pool length is 1.1 m for this case. In comparison with the experimental data the pool distribution is not within expectation, but the wall temperature distribution in normal direction is more comparable with the experimental data. This leads to the conclusion that the wall heat conduction has to be further examined and the inlet mass flow concentration has to be better predicted or calculated for future calculations. This is an example of the difficulty of validation of such models, because the identified contributors, such as wall temperature of inlet gas concentration, have not been measured during the experiments.

Fig. 5 shows a comparison of the experimental data of the ground thermocouples of test 5. The different “columns” on ordinate show the different thermocouples, which were installed on the ground in a distance of 0.1 m starting at 0.5 m. On the abscissa, the time of the experiment is plotted. The temperature data has been colored according to the temperature value. A blue color indicates a temperature close to the LH2 boiling temperature of 20.3 K. Based on these information, the length of the pool has been drawn with a dotted line for the sake of clarity. Furthermore the results of both calculations described before have been marked in the figure. At this point it can be concluded that the model predicts the pool spreading and vaporization within the range of experimental and numerical uncertainty.

3.3 Validation of the gaseous hydrogen distribution

For the gas distribution plausibility checks using the experiments conducted by Witcofsky et al [20] have been performed as well as calculations using the HySafe garage experiments conducted by CEA, France ([12] and [14]).
Figure 4: Calculation of cryogenic liquefied hydrogen pool distribution (a) homogenous heat conductivity of concrete and (b) inhomogeneous heat conductivity of concrete, effective heat conductivity increased in normal direction. [5]

Figure 5: Temperature plot of the experimental data from the ground thermocouples of the HSL test 05
Figure 6: Comparison of the calculation results and the calculation of the HySafe Garage experiment, for further information [14]

The gaseous distribution of a jet has been validated using the HySafe Garage experiments. Fig. 6 shows the distribution and accumulation in different layer of the height of the garage. The shown values are area averaged values. Due to safety issues helium has been used instead of hydrogen, but a similar behaviour of the gas distribution is expected.

The above mentioned differences show the necessity of a plausibility check of the results for the several models. The validation of the results in the end is only completely possible using all models, calculate and compare the data with the available experimental data and verify against already validated codes.

3.4 Validation of Recombiner modeling

The stand-alone application of the REKO-DIREKT code has been successfully validated against experimental data (Reinecke et al., 2006 [28], Reinecke et al., 2010b [17]) and could also predict the behavior of the PAR under oxygen depleting conditions (Reinecke et al., 2007 [29]). The coupled approach is validated against the comprehensive database of PAR performance experiments (Poss et al., 2010 [27]) performed within the OECD/NEA THAI project. Fig. 7 gives a principle overview of such a test by means of the test HR2. A defined amount of hydrogen is injected at low elevation into a dry or humid air atmosphere during a first period (here t~1200 s). After a short delay, the PAR starts operation and the hydrogen is recombined (t<5000 s). The atmosphere is well mixed due to the hot exhaust gas plume.
In order to validate the hydrogen mixing the H₂ in- and outlet concentration are compared to the experimental results (compare Fig. 8a). The values at the y-axis of the figures are removed as the experimental database is restricted to the THAI project partners. Qualitatively and quantitatively the GH2 concentration histories are predicted well, except a visibly stronger slope during the early depletion phase (t = 1200 s - 2000 s). The PAR model performance is rated by means of the H₂ recombination rate and efficiency (compare Fig. 8b). Considering a very good agreement between predicted and measured PAR conversion efficiency (i.e. hydrogen recombined vs. hydrogen entering the PAR), the reaction model is also in very good agreement with the experiment. The remaining deviations related to the prediction of the reaction rate can be traced back to slight differences in the experimental and predicted PAR throughput.
A comprehensive summary of the test HR2 can be found at (Kelm et. al, 2012 [30])

4.0 CONCLUSION AND FUTURE WORK

The modelling strategy of simulation of hydrogen accidents from the release and distribution of liquid hydrogen, the vaporization of hydrogen and the gaseous hydrogen has been described and was completed by consideration of PARs as a possible mitigation measure in confined volumes. At the present state the single models were developed, implemented and a first successful validation was varied out. Due to the fact that there are uncertainties related to the experimental data and most of the experiments are integral effect test more different test cases need to be analysed in order to eliminate these uncertainties from the validation process.

The main objective is to develop an integrated approach for the full sequence of phenomena occurring during an accident. It will be applied in order to analyse generic or realistic accident scenarios and to obtain a better understanding of the interaction of the physical phenomena. A focus on mitigation of such accidents will clearly limit the consequence of accidents related to hydrogen and will lead to a safer handling of hydrogen.

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