*Transactions*, SMiRT-23
Manchester, United Kingdom - August 10-14, 2015
Division III, Paper ID 615

# VERIFICATION & VALIDATION OF FLUID DYNAMICS METHODS IN SUPPORT OF HYDROGEN SAFETY MANAGEMENT FOR LEGACY FUEL CLADDING WASTE PROCESSING

**Kevin Malone[1], Darrell Egarr[1], and David Burt[2]**

[1] Senior Consultant, MMI Engineering Ltd, UK
[2] Principal Consultant, MMI Engineering Ltd, UK

## ABSTRACT

This paper presents aspects of fluid dynamic analysis in support of the process and ventilation design for a new Intermediate Level Waste processing plant which will convert legacy fuel cladding sludge wastes into a form suitable for long term storage.

In addition to the radiation hazard which means that the process must be remotely operated from outside the containment structure, the untreated waste evolves hydrogen gas. The waste is largely made up of sludges from long-term corrosion of fuel cladding material. The hydrogen is produced during the cladding material corrosion reaction and so there is a need to manage the potential for hazardous accumulations of flammable hydrogen within the containment structure.

The dispersion of hydrogen gas within the complex process equipment and building facilities has been investigated with Computational Fluid Dynamics (CFD) in great detail by the authors as part of a six-year programme of development for the plant engineering design and safety case. In addition, consequence assessments of the overpressures produced from hydrogen explosion fault cases have been undertaken using the specialist fluid dynamic code FLACS.

The work presented here describes examples of the fluid dynamics analysis which has been undertaken, with particular emphasis on validation and verification studies which have been investigated for the hydrogen dispersion and hydrogen explosion modelling methodologies that have been deployed in support of the development programme.

## INTRODUCTION

Amongst the legacy storage facilities on the Sellafield site is one which contains swarf waste from Magnox fuel decanning operations, and which has been identified by the UK Nuclear Decommissioning Authority as being the highest priority for hazard and risk reduction (NDA, 2014). The original facility was constructed in the 1960s and has subsequently been extended with additional compartments. The swarf material held in wet storage within the facility is an alloy of magnesium, which has corroded over time to produce a waste sludge. The waste also contains traces of uranium carried over from fuel decanning operations. Other silo contents include miscellaneous intermediate level waste items.

It is required to empty the legacy storage facility compartments of radioactive inventory in order to reduce the legacy hazard and enable decommissioning of the facility. The waste will be mechanically retrieved from each compartment and exported in skips to a new encapsulation facility for immobilisation into a wasteform that is suitable for long term storage. The encapsulation process for the legacy sludge wastes will consist of tipping the contents of the waste skips into a mixing vessel, to which dry cement grout powder will then be added. The waste and grout will be tumble mixed and then poured into a steel liner.

Filled liners will be stacked in the processing area until the wasteform is cured, then placed into a $3m^3$ box and exported.

Although on first impression the encapsulation process would appear to be relatively simple, there are significant engineering challenges to be overcome. Due to the radioactivity of the waste, the process must be operated and maintained remotely from behind shielded walls. Additionally, the characteristics of the waste are not well known, as sampling or in-situ monitoring of the current state of the material is not practicable and only historical records are available. Finally, the waste contains some pyrophoric material and is known to evolve hydrogen gas, which means the plant design must manage the potential for flammable gas hazards.

***The Hydrogen Challenge***

The generation of hydrogen gas is a common issue for processes and facilities in which nuclear materials are present. In wet systems hydrogen may be generated by radiolysis, in which ionising radiation causes the water molecule to dissociate into radicals which may then recombine to produce diatomic hydrogen gas. Hydrogen may also be produced as a by-product of corrosion of reactive metals such as sodium, magnesium, and zirconium, see for example Equation (1):

$$Mg + 2H_2O \rightarrow Mg(OH)_2 + H_2 \qquad (1)$$

Indeed, the explosions in 2011 at Fukushima Daiichi are presumed to be due to flammable hydrogen produced from corrosion of the zirconium fuel cladding in the presence of steam (the steam was present due to a loss of cooling water circulation caused by a power failure following the earthquake and tsunami) (ONR, 2011). The effect of these hydrogen explosions on the reactor buildings was devastating (see Figure 1). Equally, hydrogen produced by the corrosion of reactive sodium is believed to have been the cause of an explosion in the Dounreay waste shaft in the UK in 1977 (DSRL, 2015).

Figure 1. Damage due to hydrogen explosions: Left: at Fukushima Daiichi following the March 2011 tsunami (Image: ONR); Right: at Dounreay following the 1977 waste shaft explosion (Image: DSRL).

Mitigation of flammable hazards posed by hydrogen is a significant engineering challenge, as the concentration range between the Lower Flammable Limit (LFL – 4% v/v ) and Upper Flammable Limit (UFL – 75% v/v) in air is large in comparison with other flammable gases. As such strategies for hydrogen hazard management using inert gases such as nitrogen can be costly and difficult to implement/maintain due to the relatively narrow band of oxygen concentrations for which the mixture is not flammable (Figure 2). For this reason, the hydrogen management approach adopted for the encapsulation process is based on ensuring adequate ventilation flow to dilute hydrogen releases to below LFL.

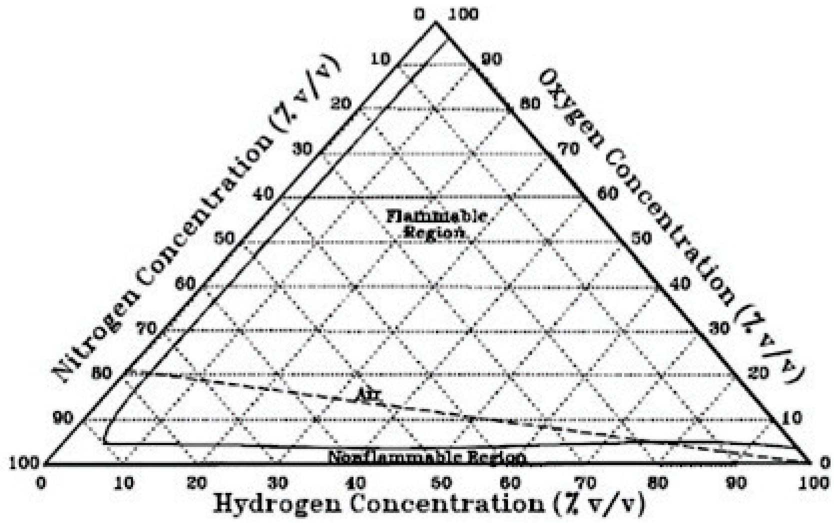


Figure 2. Flammability limits for Hydrogen in a Nitrogen and Oxygen mixture at 298K and 101.3 kPa (NASA, 1997)

***Fluid Dynamics Studies in support of Encapsulation Process Hydrogen Management***

An extensive programme of fluid dynamics assessments has been undertaken in support of the encapsulation facility engineering design and safety case. The focus of these assessments was to ensure that the hydrogen hazard will be adequately managed. Where appropriate, simple hand calculations or one-dimensional approaches were applied; however, due to the geometric complexity of the systems in question, the bulk of the assessments have required 3D Computational Fluid Dynamics (CFD) calculations. Essentially, two types of assessment methodology and software tools have been applied:

1. Dispersion assessments to calculate the potential for a flammable atmosphere to be produced due to a hydrogen release. For these the general purpose CFD code ANSYS-CFX was used.
2. Explosion assessments to determine overpressures that would be produced in the event of ignition of a flammable hydrogen volume. For these the specialist CFD software FLACS was used.

For hydrogen dispersion assessments, CFD calculations were undertaken to determine the hydrogen concentration distribution throughout the entire process area, taking into account credible releases from each process station. The purpose of these calculations has been to underpin the design for the extract ventilation system and ensure that it will be able to prevent large-scale accumulations of flammable hydrogen. In addition, detailed assessments were performed for each of the process stations, in order to confirm whether there is potential for local regions of flammable hydrogen to occur which could pose a hazard to the process equipment.

For hydrogen explosion assessments, CFD calculations were only undertaken where dispersion assessments indicated that there was potential for flammable hydrogen to arise at a process station; these were generally for fault case and outlier scenarios since the design intent was for there to be no significant volume of hydrogen above the LFL during normal operations. Figure 3 presents some selected examples of the dispersion and explosion assessments that were undertaken.

The outputs from the CFD models were used to underpin the encapsulation facility engineering design and safety case. While some pilot plant trials have been undertaken to prove aspects of the process design, physical trials to confirm the effectiveness of the hydrogen management arrangements were not practicable. For this reason validation studies were undertaken to provide increased confidence in the CFD methodologies, by undertaking test cases which could be compared against literature and experimental data.

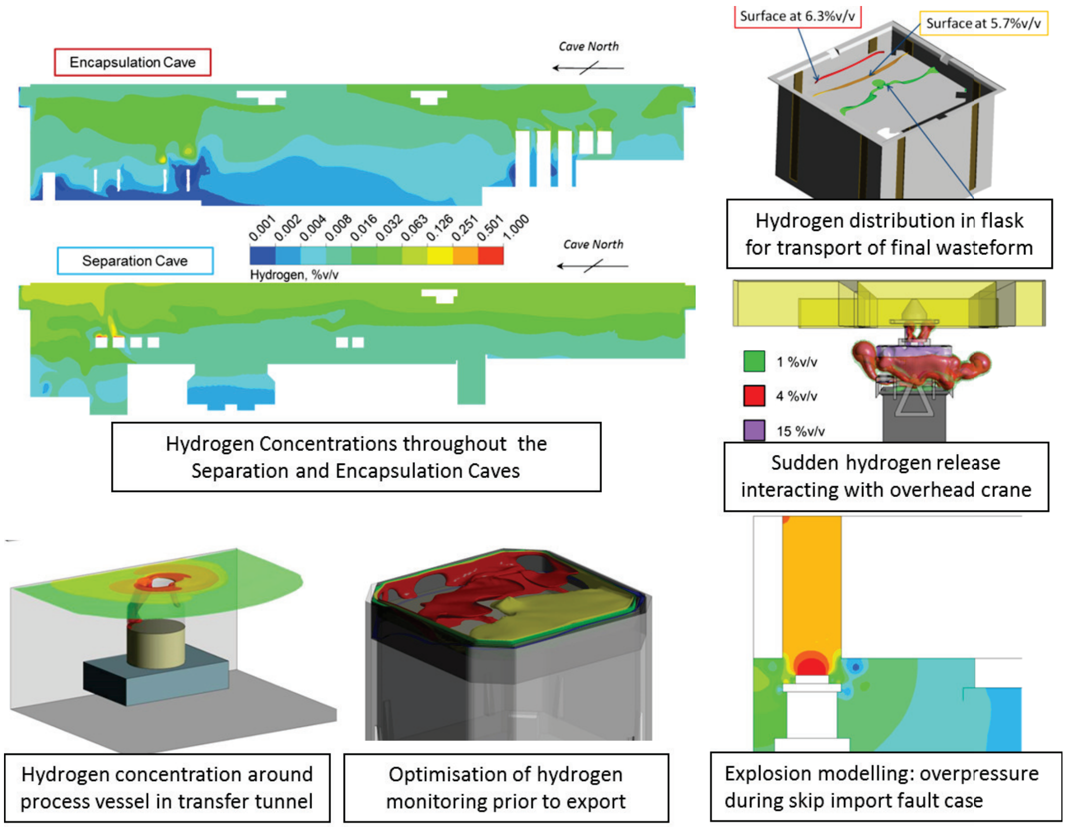


Figure 3. Selected examples of Fluid Dynamics Assessments undertaken in support of the Encapsulation Facility Engineering Design & Safety Case.

## VALIDATION STUDIES FOR HYDROGEN DISPERSION MODELLING METHODS

In terms of defining requirements for validation datasets for the hydrogen dispersion modelling methodology, four distinct types of release have been modelled in support of the encapsulation facility design:

1. A sudden gas release or 'thermal'. An example is shown in Figure 4a where a given quantity of hydrogen was released within the mixing machine.
2. A plume from a continuous point source.
3. Hydrogen release in to a stratified environment. An example is shown in Figure 4b where due to the operation of concept Passive Autocatalytic Recombiners (PARs), the gases within the process cave have been heated. Hydrogen released from the top of a stack of waste liners is less buoyant than the gas into which it is released resulting in a stratified layer of hydrogen.
4. A uniformly distributed hydrogen release such as from the surface of a curing waste product liner.

A selection of the various validation cases undertaken is discussed below.

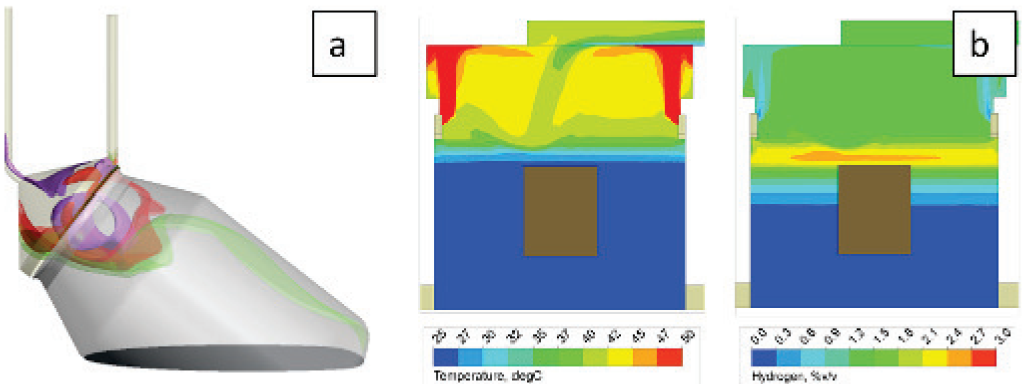


Figure 4: (a). Hydrogen distribution at 1%v/v (green), 4%v/v (red) and 15%v/v (purple) following a sudden gas release within the mixing vessel. (b) Hydrogen released in to a warm environment resulting in a stratified layer of hydrogen

***Hydrogen plume from a continuous point source in a quiescent and linearly stratified environment***

Hydrogen releases into a stratified and stable environment occur where there is accumulation of hydrogen or heat above the hydrogen source. An example is where there are hot gases following operation of PARs as illustrated in Figure 4b above. For a plume released in to a stable environment, the plume will have finite height as the density of the fluid in the plume will eventually equal the ambient density and the absence of a positive buoyancy force prevents the gases from rising. The elevation at which there is equal density is the equilibrium height. Initially however, the momentum of the gas in the plume causes it to overshoot the equilibrium height, after which the gases in the plume drop to the equilibrium height and spread radially outwards in order to obey continuity. This is illustrated in Figure 5.

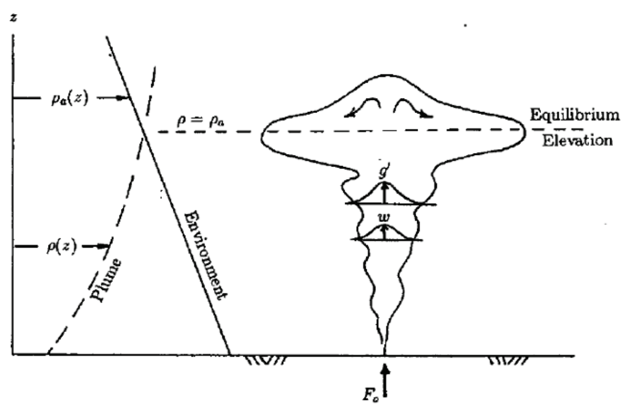


Figure 5: The spreading of a plume in a stratified environment (Lee and Chu, 2003)

The height of the plume in a stratified environment is a function of the buoyancy flux, $F$, and the stratification, $S$, which are defined as shown in Equations (2) and (3).

$$S = \frac{-g}{\rho_{ref}} \frac{d\rho_a}{dz} \tag{2}$$

$$F = \frac{F_0}{\pi} \tag{3}$$

Where g is acceleration due to gravity, $\frac{d\rho_a}{dz}$ is the density gradient along the plume, $\rho_{ref}$ is the average density of the environment, and $F_0$ is the specific buoyancy flux which depends on the release rate. Carrazo et al (2008) provide data for a range of plume equilibrium heights in a linearly stratified environment, and Equation (4) presents a correlation for equilibrium height by Briggs (1969).

$$z_{eq} = 5F^{1/4}S^{-3/8} \tag{4}$$

A CFD model was generated based on the INERIS 'hydrogen garage' scenario (Venetsanosa et al, 2009) which had been considered as a separate validation case. The 'garage' had dimensions of 7.2 x 3.78 x 2.88 m (length, width and height respectively), and featured a hydrogen injector of 120mm diameter and 265mm height at floor level. A structured hexahedral mesh was generated comprising 307,000 elements. The initial condition was a quiescent hydrogen-air mixture such that the hydrogen concentration at floor level was 0%v/v and increasing linearly to 2%v/v at the top boundary.

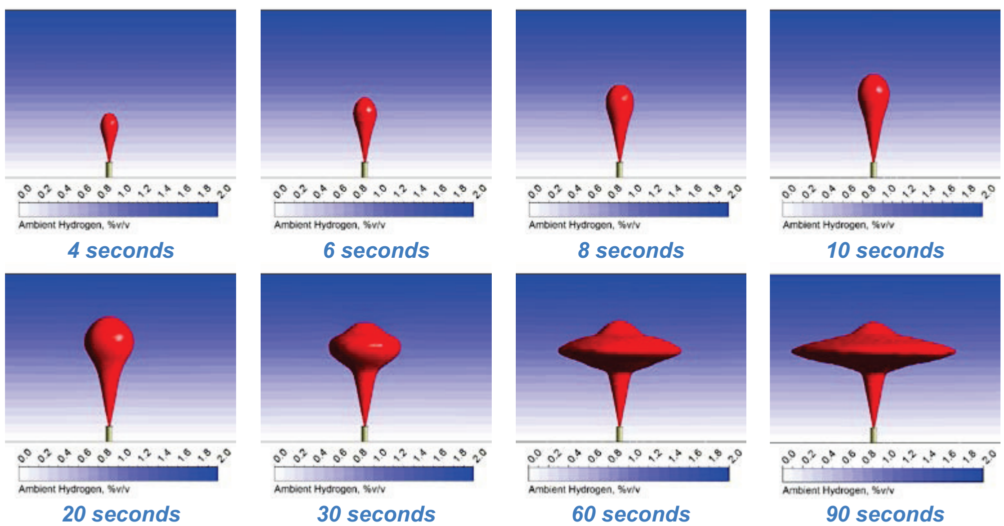


Figure 6: Development of a hydrogen plume in a linearly stratified and quiescent environment.

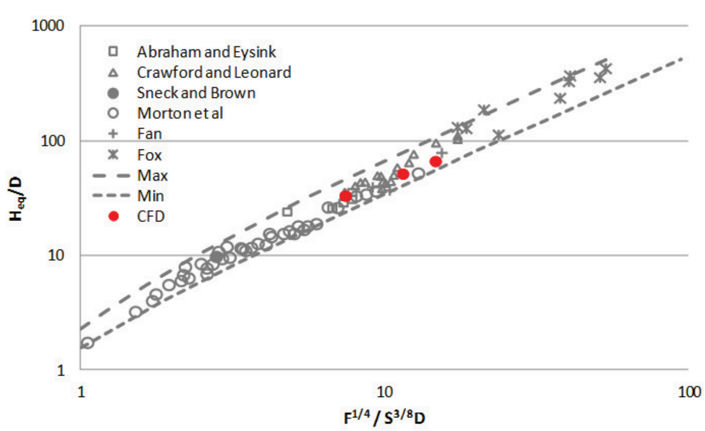


Figure 7: Comparison of empirical data for the equilibrium height of a plume from a continuous point source in a quiescent and linearly stratified environment (Carrazo et al 2008) with CFD results.

Figure 6 presents the development of the hydrogen plume, and Figure 7 presents a comparison of the equilibrium height of the plume from empirical data (Carrazo et al, 2008) with CFD results. Figure 6 clearly shows that at the centreline the height of the plume overshoots the equilibrium height due to its momentum, while Figure 7 demonstrates that the model results are within the range of the empirical data.

***Hydrogen thermal in a quiescent and uniform environment***

A thermal describes the release of a finite volume of buoyant fluid. The potential for a hydrogen 'thermal' exists due to agitation of the waste e.g. tipping of waste skips into mixing vessels. Scorer (1957) studied thermals of heavy salt solutions in a water tank and proposed that the distance of the height of the cap of the thermal relative to the virtual origin (z) is proportional to the spread (r) multiplied by a spreading factor (n) as shown in Equation (5).

$$z = nr \tag{5}$$

The virtual origin is the apex of the cone swept by the largest horizontal section of the thermal. Thus, the rate at which the thermal grows is a constant. The values for n from Scorer's experiments were in the range 2.9 to 5 and the average was 4. Equation (6) presents a correlation by Bailey (2003) which defines the height to which the flammable volume rises in a turbulent thermal, based on the released volume V.

$$z_{LFL,\max} = 12V^{1/3} \tag{6}$$

Based on the assumption of a spherical volume of gas at time zero, the equivalent spreading rate for equation (6) has been calculated in order to compare against CFD. The spreading rate was calculated to be approximately 8.5. Bailey (2003) suggests that a release greater than 0.5 litres of pure hydrogen would create a turbulent thermal. A CFD calculation was therefore undertaken in which a 5 litre volume of hydrogen is allowed to rise and disperse.

The model geometry was defined with length and width of 9.5D, and height of 19.5D, where D is the diameter of the sphere of hydrogen at zero seconds. The initial hydrogen sphere was located approximately 1.75D from the base of the domain. A single symmetry plane was used to reduce the extent of the domain. A structured hexahedral mesh was generated comprising 394,000 elements. The initial condition was a quiescent atmosphere i.e. fluid velocities at 0m/s.

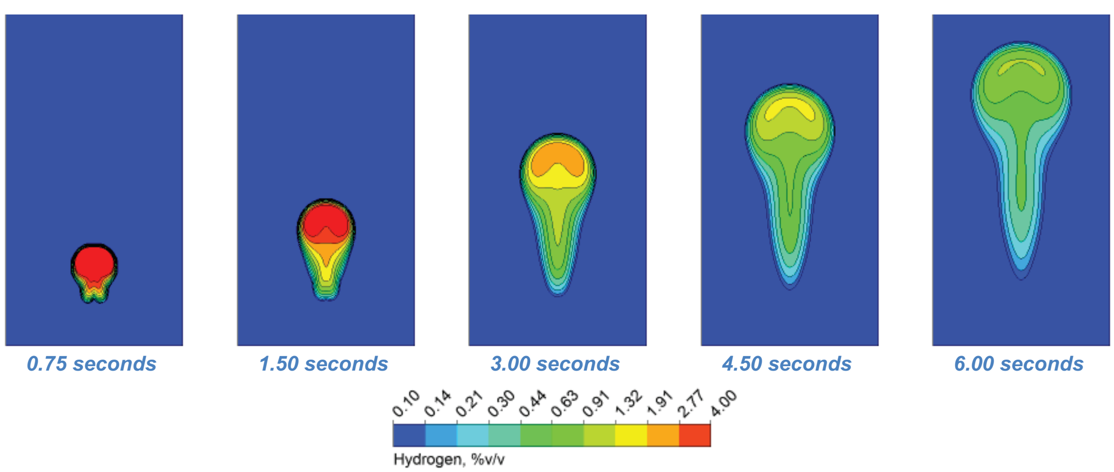


Figure 8: Development of a hydrogen thermal in a uniform and quiescent environment.

Figure 8 presents contours of hydrogen concentration showing the growth of the hydrogen thermal. A difference between the simplified theoretical model and the CFD results is that the hydrogen plume does not persist as an oblate spheroid as can be seen from the contour plots. This is because a trail of hydrogen is generated as it mixes with the ambient air.

Figure 9 compares the spreading rate of the thermal with Scorer's results where the spreading parameter is between 2.9 and 5. A comparison is also made to the derived spreading rate based on Bailey's correlation for the height of the flammable zone. The spreading of the thermal in the CFD results is based on the 0.01%v/v contour. The spreading rate for the CFD result is estimated to be approximately 5 which is close to the average spreading rate in Scorer's experiments which was 4. It is suspected that there may have been some conservatism built into Bailey's correlation.

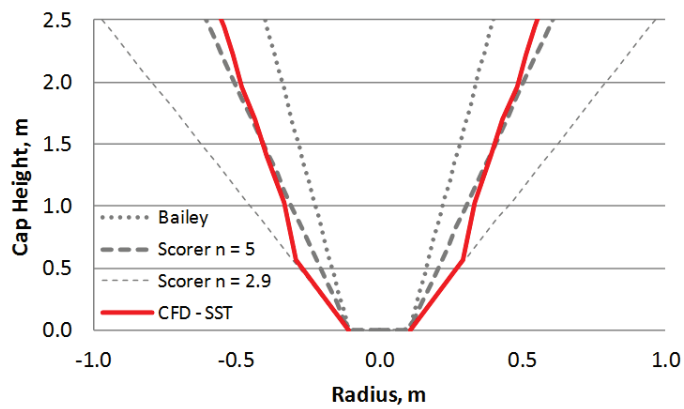


Figure 9: Comparison of the spreading of the hydrogen thermal with semi-empirical models.

The ability of CFD to calculate the spreading rate within the range of experimental data and the same flow characteristics as described in literature provides confidence that the model captures the correct physical flow behaviour.

## VALIDATION STUDIES FOR HYDROGEN EXPLOSION MODELLING METHODS

In terms of defining requirements for validation datasets for the hydrogen explosion modelling methodology, the various cases modelled for the encapsulation facility consisted of similar scenarios:

- Hydrogen inventory of relatively small volume (~3m$^3$ of pure hydrogen or less),
- Initial gas cloud concentration in the range 10-30% v/v hydrogen in air,
- Initial conditions were ambient temperatures of 25-30°C and pressures close to atmospheric,
- The output variable of interest was the overpressure generated by the explosion, particularly that witnessed by key parts of the plant containment structure e.g. viewing windows.

British Nuclear Fuels Limited had previously undertaken a number of hydrogen explosion experiments at the Health and Safety Laboratory in Buxton in support of waste retrieval from the legacy storage facility (Pritchard et al, 2004). This data was available to the encapsulation facility design project and was considered to be a suitable validation test case.

The experimental system consisted of a cuboidal steel test chamber of approximately 60m$^3$ total internal volume. This was part-filled with water to give a 1m deep ullage space of approximately 20m3. Four 0.3m x 0.1m rectangular slots were located in the chamber upper walls, through which the explosion products were vented directly to atmosphere. The initial gas clouds were contained within a plastic bag located in the centre of the ullage space. Tests were carried out with hydrogen-air mixtures at hydrogen concentrations of 10, 20 and 30% v/v in air, and explosion pressures were measured using pressure transducers located in the test chamber walls.

A simplified but representative FLACS model geometry of the experimental system was constructed. The approach taken for geometry simplification was similar to that used for FLACS analyses of process stations, with the structure constructed from combinations of cuboidal objects which were aligned to the structured Cartesian grid required by the FLACS software. Pressure monitor panels were located at the positions where transducers were positioned in the experimental system. The water surface was modelled as a rigid surface.

Figure 10 presents a comparison of the FLACS overpressure time history plots with the results from experiments. There was reasonable agreement between the model and the experiments for the 20% and 30% gas clouds in terms of both the magnitude and timing of the peak overpressure. The FLACS results for the 10% gas cloud did not show as good agreement with the experimental results; the peak overpressure from the model was of greater magnitude and occurred significantly later than in the experimental trial.

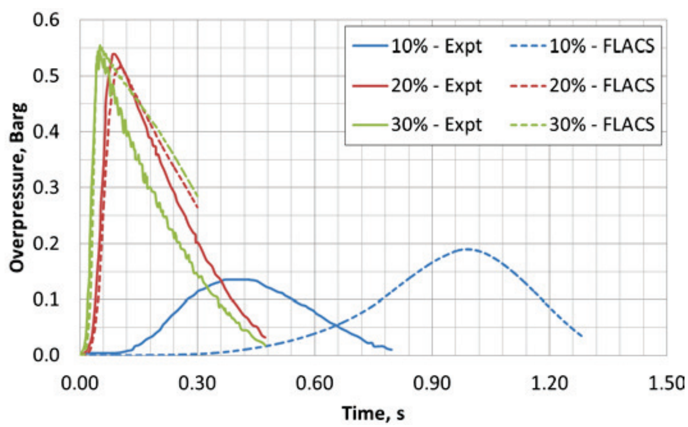


Figure 10. Comparison of overpressure results from experiments (Pritchard et al, 2004) with overpressure time history from FLACS cases.

Table 1 Comparison of FLACS Results with Experimental Data, Initial 10, 20, and 30% Gas Clouds.

| Gas Cloud | FLACS Max Panel Pressure | Peak Experimental Overpressures | | | |
|---|---|---|---|---|---|
| | | Mean | Std. Dev. | Minimum | Maximum |
| | *barg* | *barg* | | | |
| 10% | 0.19 | 0.09 | 0.04 | 0.05 | 0.14 |
| 20% | 0.52 | 0.56 | 0.01 | 0.54 | 0.58 |
| 30% | 0.56 | 0.55 | 0.04 | 0.48 | 0.59 |

Table 1 presents summary overpressure data from the experimental tests (Pritchard et al, 2004). The FLACS results for the 20% and 30% clouds agreed well with the mean values from the experiments. The difference for the 10% gas cloud was more significant, with the FLACS overpressure being 0.1 bar higher (+110%) than the mean value from the experiments. However, as highlighted by the standard deviation, minimum and maximum values presented in the Table, the experimental results for the 10% cloud showed a greater variation in measured overpressures across all of the tests undertaken.

## CONCLUSIONS

For the dispersion validation cases, the ability of CFD to capture flow characteristics which have been described in the literature e.g. overshoot of plume height, and to calculate parameters such as the equilibrium height of the plume and the spreading rate of the thermal are all in good agreement with literature data which provides confidence that the model captures the correct physical flow behaviour.

For the explosion validation study, the FLACS model provided good agreement with the results of hydrogen explosion experiments at higher gas cloud concentrations, but over predicted overpressures for the 10% gas cloud case. As the FLACS results for the lower cloud concentration are conservative, it is considered that the explosion methodology is appropriate for the purposes of assessments in support of the safety case.

## ACKNOWLEDGEMENTS

The authors are grateful to Sellafield Ltd for their guidance and support throughout this project.

## REFERENCES

Bailey, G.H., (2003). “Buoyant Dispersion of Sudden Hydrogen Releases”. *BNFL Report RAT (03) 3764*

Breitung, W., Chan, C., Dorofeev, S. , Eder, A., Gelfand, B., Heitsch, M., Klein, R., Malliakos, A., Shepherd, E., Studer, E., Thibault, P. (2000). “State-of-the-Art Report on Flame Acceleration and Deflagration-to-Detonation Transition in Nuclear Safety”. *NEA/CSNI/R(2000)7*, OECD Nuclear Energy Agency.

Briggs, G. A. (1969), Optimum formulas for buoyant plume rise, *Philos. Trans. R. Soc. Lond.*, 265, 197–203.

Carazzo, G., Kaminski, E., Tait, S. (2008). “On the rise of turbulent plumes: Quantitative effects of variable entrainment for submarine hydrothermal vents, terrestrial and extra terrestrial explosive volcanism”. *Journal of Geophysical Research*, Vol. 113, B09201.

DSRL (2015): Dounreay Site 1977 shaft explosion: http://www.dounreay.com/decommissioning/shaft-and-silo/1977-shaft-explosion. Dounreay Site Restoration Limited. Accessed 12/03/15

Lee, J.H.W. and Chu, V.H. (2003). “Turbulent jets and plumes - A Lagrangian approach”. Kluwer Academic Publishers.

NASA. (1997). “Safety Standard for Hydrogen and Hydrogen Systems”. *NSSS 1740.16*, National Aeronautics and Space Administration.

NDA. (2014). Business Plan: Financial year beginning April 2014 to financial year ending March 2017. Nuclear Decommissioning Authority, UK.

ONR (2011). Japanese earthquake and tsunami: Implications for the UK nuclear industry Final Report HM Chief Inspector of Nuclear Installations. Office for Nuclear Regulation, UK.

Pritchard, D. K., Hedley, D. and Eaton, G. T. (2004). “Hydrogen explosion trials for British Nuclear Fuels”. *BNFL Report RP/LPSERP-000/PROJ/00082/A*.

Scorer, R. S. (1957). “Experiments on convection of isolated masses of buoyant fluid”. *Journal of Fluid Mechanics*, 2, pp 583-594.

Venetsanosa, A.G. , Papanikolaoua, E., Delichatsiosa, M., Garciac, J., Hansend, O.R., Heitsche, M., Huserf, A., Jahng, W., Jordanh, T., Lacomei, J.-M., Ledinj, H.S., Makarovk, D., Middhad, P., Studerl, E., Tchouvelevm, A.V., Teodorczykn, A., Verbeckek, F., Van der Voorto, M.M. (2009). “An inter-comparison exercise on the capabilities of CFD models to predict the short and long term distribution and mixing of hydrogen in a garage” *International Journal of Hydrogen Energy*, 34, pp 5912-5923.