

Application of Bayesian Belief Networks to assess hydrogen gas retention hazards and equipment reliability in nuclear chemical plants

Fayaz Ahmed, Radiological Safety Assessor, Sellafield Ltd., Hinton House, H460, Risley, Warrington, Cheshire, WA3 6GR.

Many nuclear waste reprocessing and storage plant processes result in the generation of hydrogen gas. Radiolysis of radioactive liquors and corrosion of metallic magnesium waste are the main mechanisms for generating hydrogen in such facilities. Corrosion products such as magnesium hydroxide sludge are also formed which require storage in transportable vessels. Demonstration of sufficient reliability of systems such as purge air and ventilation extract is therefore required to protect against releases of hydrogen. Factors affecting hydrogen ignition and removal in nuclear environments as well as the identification of appropriate hazard management strategies have been the key areas of research for decommissioning and reprocessing plants. However, a knowledge gap has been identified in terms of assessing the likelihood of hydrogen retention within the sludge and waste matrix resulting in a sudden release of the gas into a vessel ullage. Hydrogen gas retention and the potential for a sudden release are affected by numerous factors such as faults leading to adverse waste disturbance. As such an appropriate technique must be applied to analyse the uncertainty from this gas behaviour. Bayesian Belief Networks (BBN) is an emerging statistical technique which allows uncertainty and dependencies between multiple variables to be taken into account in a quantified risk assessment. A BBN analysis has been undertaken to determine the key factors that would lead to disturbance of the sludge waste and the subsequent sudden release of hydrogen into the ullage space of a process vessel. The results show that the key sensitivities are adverse disturbance of the vessel sludge waste caused by faults leading to uncontrolled movements and clashes of the vessel. The benefits of applying the BBN technique to assess reliability of the purge and ventilation extract systems against radiolytic hydrogen release have also been explored. The BBN model has shown to be particularly advantageous, as it has allowed input of probability distributions of the key variables, instead of single point values, thus providing an enhanced understanding of uncertainty. Furthermore, the BBN technique has allowed updating of the probability of a known variable given a particular condition of the other variables. This updating function has enabled the key sensitivities to be determined.

Keywords

Bayesian Belief Network, sudden release, hydrogen retention, compressed air, reliability

Introduction

The generation of hydrogen gas in nuclear decommissioning and reprocessing plants presents some significant challenges which must be dealt with effectively. Corrosion of stored fuel cladding waste and radiolysis of aqueous solutions are the main mechanisms for hydrogen generation in such facilities. Understandably the effects of a potential hydrogen explosion in nuclear environments are that not only can it lead to serious operator injury, but there is also the added radiological impact. Some of the well known severe accidents in relation to hydrogen explosions in nuclear power stations, as discussed by *Gharari et al*, 2018, include the Three Mile Island, Chernobyl and Fukushima Daiichai. Such major incidents emphasise the need for robust controls against the hydrogen hazard.

Ultimately the Duty Holder of nuclear installations, including those approaching decommissioning, has the obligation to demonstrate that the hydrogen explosion hazard is appropriately managed. Preferably, the hazard should be eliminated or prevented by active protection systems. The fundamental aim of these preferred Hazard Management Strategies (HMS) is to show that the risk of hydrogen explosions is tolerable in accordance with the As Low As Reasonably Practical (ALARP) principle specified by the *Health and Safety Executive*, 2014.

The severe accident review by *Gharari et al*, 2018 clearly illustrates the complexities surrounding the hydrogen explosion mechanism. Such complexities result in a number of uncertainties and hence treatment of the hydrogen explosion as a probabilistic event in nuclear safety cases. A common symptom of these uncertainties is overly pessimistic probabilistic risk assessments. Whilst an appropriate level of conservatism in safety assessments is acceptable for Greenfield plants, this can be detrimental to existing nuclear facilities awaiting decommissioning. The reason being, complex design solutions are often difficult to substantiate, ultimately causing delays to decommissioning. This would be disadvantageous in terms of meeting the government policy for acceleration of nuclear decommissioning as discussed by the *Nuclear Decommissioning Authority*, 2016. An accurate assessment of the risk in support of the overall ALARP case for hydrogen gas explosions is therefore imperative.

The behaviour of hydrogen and identification of appropriate HMS has recently been the key area of research for nuclear decommissioning and reprocessing plants. The research work supported by London South Bank University (LSBU) used a combination of empirically derived relationships and best practice to develop a hydrogen technical guide in the form of a road map as reported by *Ingram et al*, 2001. The road map provided guidance on the assessment of hydrogen releases, the potential consequences and suggested appropriate risk mitigation and prevention techniques. The road map also advised hydrogen ignition probabilities for various decommissioning and reprocessing environments.

Whilst the roadmap by *Ingram et al*, 2001 provided useful guidance on assessment of hydrogen explosions for a wide range of scenarios, there are still a number of knowledge gaps which are not covered by this approach. A by-product from corrosion of nuclear fuel cladding waste, namely Magnox swarf, is magnesium hydroxide sludge which can lead to hold-up of hydrogen. There are multiple factors affecting the hold-up of hydrogen in the sludge matrix and the likelihood of a

sudden release of the gas into a vessel ullage space. This release mechanism in sludge wastes has been identified as one area of uncertainty which requires analysis using an appropriate methodology.

Some reprocessing and storage operations also result in a slow release of hydrogen gas due to radiolysis of aqueous radioactive liquors. *Ingram et al, 2001* suggests that such slow releases can be managed by the use of purge air and ventilation extract systems to continuously dilute and remove the hydrogen gas. Factors affecting the reliability of the purge air and ventilation extract systems are a further area of uncertainty which require investigation.

This paper examines in detail the mechanisms for hydrogen retention specifically in sludge wastes. Factors leading to a sudden release into the ullage space of a transportable vessel used for interim safe storage of the Magnox sludge waste are also investigated. Using the Bayesian Belief Network (BBN) statistical technique detailed by *Bolstad, 2007* and *Pasman et al, 2012*, key sensitivities affecting the likelihood of the gas reaching a flammable concentration are determined. An initial application of BBNs to analyse the uncertainty from the sudden hydrogen release mechanism has been undertaken recently by the author in a separate article, *Ahmed, 2019*. However the initial BBN model by *Ahmed, 2019*, was based on a variety of wastes with a lesser capacity for hydrogen retention. Furthermore the factors leading to the sudden release of hydrogen were not previously considered. This particular article focusses on the sudden gas release behaviour in sludge wastes which represent the worst case in terms of hydrogen retention. The modelling undertaken in this article is predominantly affected by variables leading to uncontrolled storage vessel movements responsible for the sudden gas release.

A second case study in this article, which has not been previously considered, applies the BBN technique to determine the key sensitivities affecting the reliability of purge air systems for removal of radiolytic hydrogen in a process vessel. The overall aim of both case studies is to support ALARP decision making on factors which would require enhanced control to reduce the risk of hydrogen explosions.

Hydrogen release mechanisms

Chronic hydrogen generation

Underwater storage of the solid waste generated from de-canning of nuclear fuel rods, primarily metallic magnesium, or Magnox, results in corrosion of the metal to form magnesium hydroxide sludge and hydrogen gas. This continuous hydrogen generation due to Magnox corrosion is referred to as 'chronic hydrogen' in this paper. The UK government strategy for safe disposal of Magnox waste requires interim storage of the waste packages in surface stores until a suitable geological waste repository becomes available. Interim storage involves loading the waste into vessels and immersed under cover water within the vessels. Although this case study considers waste material which is predominantly sludge, there is still the likelihood of small residual quantities of metallic Magnox being present within the sludge. Therefore it is expected that the corrosion of the residual uncorroded Magnox under cover water will continue in the vessel. However the rate of hydrogen generation will decrease during storage due to the continuous consumption of the uncorroded metal.

The transportable vessel features filtered outlets to enable natural venting of the hydrogen gas. The vessel is lidded for the purpose of maintaining containment of the radioactive waste.

Hydrogen hold-up and sudden release

The continuous generation of hydrogen gas within the swarf and sludge mixture can result in expansion, or swelling, of the waste matrix. The gas hold-up and expansion behaviour and a subsequent sudden release from sludges is well recognised in literature. For example *van Kessel et al, 2002* considered the potential for methane and carbon dioxide gas, produced in sludge depots, to cause expansion of the dredging sludge. The studies by *van Kessel et al, 2002* showed that the gas bubbles produced sufficient stresses to cause cracks and channels in the sludge matrix. These channels provided a path for the gas to escape to the surface. *Van Kessel et al, 2002* also showed that the rate of flow of gases to the surface of the depot depended on the sludge shear strength. At shear strengths less than 10kPa, the gas bubbles led to expansion of the waste matrix. It is considered that the shear strength of Magnox sludge is such that waste expansion within the vessel is a credible scenario. The hydrogen will initially build within the sludge matrix, some will flow into the ullage space and vented from the vessel via the filtered outlet. Once equilibrium is reached, the hydrogen will be released into the ullage at a uniform rate resulting in a steady state hydrogen concentration, referred to as 'equilibrium hydrogen concentration'.

The hydrogen held-up in the sludge matrix has the potential to be released suddenly into the ullage space in the event of significant disturbance of the storage vessel. Therefore the total volume of hydrogen that could arise in the ullage under these conditions would be the equilibrium hydrogen volume plus the sudden release volume. Hence the vessel ullage hydrogen concentration can be expressed as:

Radiolytic hydrogen generation in reprocessing operations

Many process vessels in nuclear fuel reprocessing operations contain radioactive aqueous liquors. The ionising radiation associated with such liquors results in radiolysis. With alpha, beta and gamma radiation, water dissociates to form the hydrogen and the hydroxyl radical in accordance with the following chemical equation:

$$H + e^{-} + H_2O \rightarrow H_2 + OH^{-}$$

Bibler et al, 2007, demonstrate that the key variables which affect the rate of hydrogen generation from radiolysis are the ' $G(H_2)$ value' and the rate of absorption of energy by the aqueous liquor, as expressed by the following relationship:

$$Q_{\rm H} = kG(H_2)_{(\alpha)}E_{(\alpha)} + kG(H_2)_{(\gamma)}E_{(\beta/\gamma)}$$
 (Equation 2)

Where:

Q_H is the hydrogen generation rate in litres/hr.

 $G(H_2)$ value is the number of hydrogen molecules evolved per 100eV of energy absorbed by the medium. For alpha (α) and Beta Gamma (β/γ) radiation in aqueous solutions $G(H_2)$ values of 1.66 and 0.45 molecules H_2 /100eV of radiation respectively have been specified by *Bibler et al*, 2007.

 $E_{(\alpha)}$ and $E_{(\beta/\gamma)}$ is the rate of absorption of energy by the medium expressed in MeV/s. This is simply the product of the α and β/γ radioactivity content in units Becquerels and the disintegration energy in MeV. Disintegration energies of 0.662 MeV and 0.186MeV for α and β/γ radiation respectively have also been reported by *Bibler et al*, 2007. Typically in process vessels with aqueous liquors consisting of predominantly β/γ radioactivity, the $E_{(\beta/\gamma)}$ value of 8.13 x10⁻¹³MeV/s is known.

K is a dimensional constant with a value of 1.44×10^{-15} when $E_{(\alpha)}$ and $E_{(\beta/\gamma)}$ is expressed in MeV/s.

Using Equation 2 and the associated parameter values as discussed above, the radiolytic hydrogen generation rate in a process vessel containing β/γ radioactivity can be calculated as:

 $Q_{\rm H} = 0.0527 \ \text{l/hr} [1.44 \times 10^{-15} \times 0.45 \times 8.13 \times 10^{-13}]$

Hazard Management Strategies

Continuous release of chronic hydrogen in sludge waste storage vessels

The chronic hydrogen generation rate in interim storage vessels containing waste, which is mainly sludge with small quantities uncorroded Magnox, is typically less than 0.11/hr. Only a small proportion of this type of waste could result in rates up to 11/hr. As suggested by *Ingram et al*, 2001, the key Hazard Management Strategy (HMS) against chronic hydrogen generation in vessels is that a sufficient ullage gap between the cover water level and the vessel lid is maintained. This ensures that the hydrogen generated is diluted and vented from the vessel lid filtered outlets via the process of diffusion.

The filtered vents in the vessel lid are designed to maintain a steady state hydrogen in air concentration, in the vessel ullage to less than the Lower Flammable Limit (LFL) of 4% v/v. The filter performance under normal operations is such that at a chronic hydrogen generation rate of 11/hr, the equilibrium hydrogen in air concentration in the ullage is 2% v/v. Thus provided that the filter performance remains effective, the chronic hydrogen accumulation hazard in the vessel is adequately controlled. It is realised that this HMS is reliant on the control of the ullage volume. Expansion of the waste due to gas hold-up has the effect of decreasing the ullage volume such that this could result in a higher than the expected ullage hydrogen concentration. Therefore the vessel fill operations are required to ensure that the ullage volume allows for potential waste expansion such that the hydrogen concentration is below the LFL.

Sudden release of held up hydrogen in sludge waste storage vessels

Whilst the vessel design is based on the management of continuous chronic hydrogen generation, the HMS for a sudden release of held up hydrogen in the sludge waste is to prevent the onset of occurrence this hazard. The filtered vents are only effective for the dilution of chronic hydrogen. The only credible means for a sudden release would be if the vessel transfer movements result in a significant disturbance of the waste matrix. Thus the HMS is to ensure that the risk of adverse disturbance mechanisms is negligible.

Radiolytic hydrogen generation in aqueous liquor reprocessing vessels

For processes that result in a slow release of hydrogen, such as the radiolytic hydrogen generation in reprocessing vessels, the recommended hazard management strategy by *Ingram et al*,2001 is to provide ventilation extract systems to control the ullage hydrogen concentration. The ventilation extract systems would comprise a compressed purge air supply into the ullage space and extract fans to provide the driving force for continuous removal of the hydrogen in air mixture from the ullage space. The purge air is often supplied into the vessel via level control pneumercator pipework. Back-up and emergency compressed air supply systems would also be provided for the purpose of enhanced reliability of the forced ventilation system. Figure 1 shows the arrangement of a typical purge air supply and ventilation extract system for a hydrogen gas generating process vessel.

Upon introduction of the purge air to the vessel, the objective is to ensure that the equilibrium hydrogen concentration, C_{eq} (%v/v) in the ullage space is always below the LFL. Ideally the hydrogen concentration under normal operations is required to be no greater than 25% of the LFL, i.e. 1%v/v. The ullage equilibrium concentration can be estimated knowing the radiolytic hydrogen generation rate Q_{H} and the purge air supply flow rate Q_{air} as follows:

$$C_{eq} = 100Q_{\rm H}/(Q_{\rm H} + Q_{\rm air})$$
(Equation 3)

Thus if Q_{air} is kept as high as possible a low equilibrium concentration can be achieved. Typically for a hydrogen generation rate of 0.0527 l/hr and purge air flow rate of 18l/hr, a low equilibrium concentration of 0.29% v/v would be achieved. Should

the normal purge air supply fail, then the timescale, t, required to reinstate the purge air before exceeding the 1%v/v limit in an ullage volume, U, can be calculated according to the relationship:

$$t = [U/Q_{\rm H}] \ln[(100 - C_{\rm eq})/(100 - 1)]$$
(Equation 4)

Therefore for a hydrogen generation rate, Q_H of 0.0527 l/hr, a typical ullage volume, U of 500l and an equilibrium concentration of 0.29% v/v, the time required to reinstate the purge air supply before the desired 1% v/v is exceeded would be 67.6 hours. Hence the back-up purge air supply systems would need to be reinstated within this timescale to ensure that there is sufficient margin in time for rectification before the actual LFL limit is reached.



Figure 1: Typical ventilation extract system for removal of radiolytic hydrogen in a reprocessing vessel

Identification of dependent variables affecting sudden and radiolytic hydrogen release scenarios

Variables affecting sudden hydrogen release

Each of the hazard management strategies identified against chronic hydrogen generation and sudden hydrogen release has multiple dependent variables associated with it. Figure 2 illustrates the dependent variables, numbered 1 to 15, and how they interact with each other. As shown, the ullage hydrogen concentration is dependent on the ullage volume. However the ullage volume is affected by the waste expansion, or swelling, due to hydrogen hold-up. Thus a change in the waste swelling fraction not only affects the volume of hydrogen available for a sudden release but it can also result in a reduction of the ullage volume which has an impact on ullage concentration. All dependencies, interactions and their associated likelihood probabilities must therefore be taken into consideration when quantifying the risk from hydrogen hold-up and sudden release. This is discussed in the following sub-sections.

(Equation 5)

Vessel ullage and hydrogen hold-up volume

The vessel ullage volume is affected by the volume of hydrogen held in the sludge and swelling of waste matrix (variables 7 and 8) such that the ullage volume decreases with increasing waste expansion volume.

This particular case study assumes that the interim storage vessel contains predominantly sludge with small quantities of uncorroded Magnox. This type of waste has the ability to expand by a relatively large volume fraction in the range 15-20%. A probability distribution of 99% can be applied for the expansion fraction being in the range 15-20 % by volume. The hydrogen hold-up volume (variable 8) can be calculated knowing the vessel waste volume of 1000 litres (variable 4) and the waste swelling fraction (variable 7) using equation 5:

Hydrogen hold-up volume (1) = Vessel waste volume (1) x Waste swelling fraction



Figure 2: Dependent variables affecting ullage concentration in a waste storage vessel

The vessel ullage volume (variable 2) is derived knowing the vessel internal volume, the total volume of vessel contents, which are both constants, and the hydrogen hold-up volume (variable 8) using equation 6:

Vessel ullage volume = Vessel internal volume - total vessel content volume - hydrogen hold-up volume (Equation 6)

Total ullage hydrogen volume

The total ullage hydrogen volume can be derived knowing the equilibrium hydrogen volume (variable 1) and sudden hydrogen release volume (variable 9) using equation 7:

Total ullage hydrogen volume (l) = Equilibrium H_2 volume (l) + Sudden H_2 release volume (l) (Equation 7)

Equilibrium hydrogen volume

As discussed earlier, based on the chosen vessel and its lid filter design, the maximum equilibrium hydrogen concentration using the worst case chronic hydrogen generation rate of 1 l/hr is known to be 2% v/v. However it is considered that approximately 85% of the probability distribution corresponds to a low hydrogen generation rate of 0.1 l/hr which results in an equilibrium concentration of up to 0.25% v/v. On this basis a probability distribution of 85% for the equilibrium hydrogen concentration (variable 3) being in the range 0 to 0.25% v/v is considered reasonable. Knowing the vessel ullage volume (variable 2) and the equilibrium hydrogen concentration (variable 3), the equilibrium hydrogen volume (variable 1) can be derived using the equation:

Equilibrium hydrogen volume (l) = Vessel ullage volume (l) x Equilibrium H₂ concentration (% v/v)/100 (Equation 8)

Sudden release volume

The sudden release volume of hydrogen into the vessel ullage space (variable 9) is dependent on the amount of hydrogen held up which is available for a release (variable 8) and the fraction of this volume released (variable 14), as represented by the equation:

Sudden H_2 release volume (l) = Hydrogen hold-up volume (l) x Fraction of held-up H_2 released

(Equation 9)

The hydrogen release fraction of release is dependent on factors that could cause adverse disturbance of the vessel contents during transfer for interim storage. A failure of the vessel handling crane Programmable Logic Control (PLC) could lead to an uncontrolled lowering or raising of the vessel (variable 11) hence cause disturbance of the waste content. The second cause is the crane zoning system limit switch failure (variable 12) leading to a loss of zoning functionality and potential clash of the vessel with other building items. Either of the two failures, variables 11 or 12, could lead to disturbance of the vessel contents and a sudden release. If an adverse sudden release occurs, expert judgement is that up to 20% v/v of the held up hydrogen could be released into the ullage.

Variables affecting radiolytic hydrogen concentration in aqueous liquor reprocessing vessels

Section 2 of this paper discussed that the method for control of hydrogen concentration in enclosed process vessels where the hydrogen generation rate is relatively low, is to use ventilation extract systems. Ideally, as part of normal operations, the ventilation system should be sufficiently reliable so that it can maintain a hydrogen concentration of 1%v/v in the ullage space. Section 2 demonstrated that in reprocessing vessel containing aqueous liquor, with an ullage space of typically 500l and a radiolytic hydrogen generation rate of 0.0527 l/hr, the timescale for reinstatement of the purge air to the vessel, upon failure of the normal supply, would be 67.6 hours before the 1%v/v concentration limit is exceeded. In this section the variables that affect the availability of the purge air supply and the associated interactions are identified. Potential failure probabilities of the plant components are also identified for the purpose of determining the overall reliability of the system.

The ventilation system would consist of a factory purge air supply delivered to the vessel via the pneumercator level instrument pipework. The driving force for extraction of the hydrogen in air atmosphere from the ullage space is the ventilation extract system (Figure 1). This particular case study assumes that the ventilation extract fans remain operational. Only failures affecting the availability of the purge air supply system are considered. A failure of the normal factory air supply would be indicated to the operators by the pressure instrumentation alarms due to a loss of the delivery air pressure and operators would be required start the either of the two standby air compressors, A and B. Thus for a complete loss of this back up protection system to occur, either of the following events would need to occur:

- The pressure instrument has to fail, or;
- the operator has to fail to start the back-up compressors, or;
- both of the back up compressors fail mechanically, or;
- a loss of building power supply has to occur.

Assuming that the operators are well trained, and they will follow clear instructions for the start of the compressors, a low human error probability of 1 in 100, i.e. 0.01 is considered appropriate. Upon loss of power to the back-up air compressors, A and B, a second team of operators would be required to start the emergency diesel powered air compressor. Where multiple teams of operators are involved, the operator error probability is limited to a Human Performance Limiting Value (HPLV) of 1E-4. For a failure to changeover to the diesel compressor, a power failure and either a failure of this compressor or the operators failing to take action would have to occur. For the occurrence of the Top Event, i.e. a complete unavailability of the purge air system, the normal purge air supply, the back up and the emergency diesel compressor systems would have to fail coincidently. The list of failures of all the primary events discussed above and their known failure probabilities are summarised in Table 1.

| Primary Event ID | Description | Failure |
|---------------------|--|-------------|
| Event ID | | probability |
| FACTAIR | Failure of normal factory compressed | 8E-3 |
| | air supply to the process vessel | 01-5 |
| HE1 | Human error probability of operator | |
| | failing to start back up compressors A | 0.01 |
| | or B | |
| PRINST | Pressure detection instrument for | |
| | factory compressed air fails to alarm | 7.5E-3 |
| HPLV | Human Performance Limiting Value | 117.4 |
| | for different operating teams | 1E-4 |
| COMPA | Mechanical failure of back up air | |
| | compressor A | 0.245 |
| COMPB | Mechanical failure of back up air | |
| | compressor B | 0.245 |
| POWER | Loss of power to back up | |
| | compressors A and B | 4.3E-03 |
| HE2 | Human error probability of a second | |
| | operator failing to start back up | 0.01 |
| | emergency diesel compressor | |
| DCOMP | Mechanical failure of diesel | |
| | compressor. | 0.357 |

Table 1: List of Primary failure modes and probabilities for compressed air supply to a process vessel

Dependency modelling using Bayesian Belief Networks

The previous section 4 discussed the dependent variables affecting sudden release of held up hydrogen from the sludge matrix in a vessel used for transfer and interim safe storage of the waste. In a second case study, variables affecting radiolytic hydrogen release in a stationary process vessel containing aqueous radioactive liquors were also discussed. Having established these dependencies and their initial likelihood probabilities, a suitable modelling technique needs to be used to quantify the probability of exceeding the flammable hydrogen concentration in the vessel ullage. The modelling technique would also need to be able to predict key sensitivities that affect the hydrogen concentration in the vessel ullage. In the case of radiolytic hydrogen generation in reprocessing vessels, the aim is to identify a modelling technique that can enable prediction of the key sensitivities that affect the reliability of the purge air system used for dilution of the gas.

The Bayesian Belief Network (BBN) technique reported by *Bolstad, 2007* and *Pasman et al, 2012*, can provide a powerful means of modelling uncertainty and dependencies between multiple variables, such as those illustrated in Figure 2. Some of the main advantages of BBNs include:

- a) Causal variables and the relationships between them are set out in a network structure so that their interactions are clearly observable.
- b) Uncertainty in the causal variables is expressed more accurately using distributions rather than single point probability values
- c) The posterior distribution for an event, e.g. the likelihood of hydrogen concentration reaching the LFL, expresses in a simple way the predicted outcome or likelihood of that event.

Various types of data, e.g. discrete or continuous, can be used in the network. They can be derived on the basis of experiments, historical observation or expert judgement.

d) Expert systems can be developed to aid the decision making process as part of the risk assessment process.

The principles of the BBN technique are based on the use of conditional probabilities of the dependent variables. In the context of the hydrogen release model illustrated in Figure 2, the probability of the ullage concentration exceeding the LFL would be conditional to the probability of a sudden release occurring. Similarly the likelihood of a sudden release occurring is conditional to the probabilities of hydrogen hold-up and significant disturbance of the vessel due to process control failure.

Mathematically the Bayesian theory and the conditional probability concept can be explained by considering the probability of a hypothesis A given that an event B has occurred. In statistical terminology this conditional relationship between events A and B is expressed as P(A|B) which can be expanded as follows:

$$P(A|B) = P (A \text{ and } B)/P(B), \text{ i.e. } P(A \cap B)/(P(B))$$
(Equation 10)

The term $P(A \cap B)$ in Equation 5 is the probability of both the events A and B occurring. This may also be expressed as the probability of A times the probability of B given that the hypothesis A occurs, i.e.:

$$P(A \cap B) = P(A) \times P(B|A)$$

Similarly $P(A \cap B)$ can also be expressed as the probability that event B occurs times the occurrence of hypothesis A given that event B has occurred, i.e.:

$$P(A \cap B) = P(A) \times P(B|A) = P(B) \times P(A|B)$$
 (Equation 11)

Therefore solving for P(A|B) gives the following equation which represents Bayes Theorem (Bolstad, 2007):

$$P(A|B) = (P(A) \times P(B|A))/P(B)$$
 (Equation 12)

Where:

- P(A) and P(B) are the probabilities of events A and B occurring
- P(A) is referred to as the 'Prior' probability of the hypothesis before any evidence is available
- P(A|B), is referred to as the 'Posterior' which is a conditional probability for the likelihood of observing hypothesis A if B is true.
- P(B|A) is the probability of event B occurring if A is true.

Equation 12 above applies Bayes Theorem to a hypothesis with only two variables A and B. Often an uncertainty analysis such as that shown in Figure 2 is affected by many dependent variables such that the Bayesian algorithm would be difficult to compute by hand. Accordingly software systems such as Netica by *Norsys* have been developed commercially which allow Bayesian modelling in the form of Bayesian Belief Networks (BBN).

The key features of a BBN can be summarised as follows:

• Causal parameters, often referred to as the 'Parent' nodes which lead to an effect, i.e. the 'Child' node are connected together by an arc from the Parent to the Child.

- The nodes can be 'Discrete' i.e. having two or more outcomes e.g. True or False or 'Continuous' represented by a scale or a probability distribution.
- For the Child nodes, Conditional Probability Tables (CPTs) are completed based on existing data or expert opinion
- Equations representing the relation between the Parent and Child nodes can also be used by the BBN software to derive the CPT automatically particularly when multiple Parents affect a single Child.
- A feature unique to BBNs is that their updating capability allows a 'reverse analysis' to determine the effect on primary node, i.e. nodes with no Parents, probabilities conditional to the top event probability.

Case Study 1 - Uncertainty analysis of hydrogen hold-up and sudden release using Bayesian Belief Networks

Using the methodology discussed above and the Netica software for Bayesian networks, a BBN analysis was carried out for the hydrogen hold-up and sudden release in a lidded vessel containing predominantly Magnesium hydroxide sludge. The objective was to determine the key sensitivities that affect the sudden hydrogen release from sludgy wastes and the resultant impact on the vessel ullage concentration. Netica software was used to replicate the cause and effect variables identified in Figure 2, treating each of the variables 1 to 15 as a separate node. The quantification of Bayesian network was based on the Prior probability distributions discussed for each of the nodes in Section 4. The nodes ullage volume (Node 2), total ullage hydrogen volume (Node 10), equilibrium hydrogen volume (Node 1) and sudden hydrogen release volume (Node 9) are all continuous nodes. The derivation of the Conditional Probability Tables (CPTs) data for these nodes was undertaken within the BBN automatically by the software. This involved application of equations 6, 7, 8, 9 and the Prior probability distributions of the associated Parent nodes discussed in Section 4.

Figure 3 provides results of the Bayesian analysis, which can be summarised as follows:

- The probability of hydrogen hold-up volume being in the range 100-150 litres is 90%.
- The probability that the ullage equilibrium hydrogen volume is in the range 0-2 litres is 94.1%.
- At a probability of 93.2% the sudden hydrogen release volume is in the range 0 to 13.3 litres.
- With the above predicted Prior likelihoods of causal variables, the probability of the ullage hydrogen concentration being in the range 0-4% v/v, i.e. up to or below the LFL, is 36.9%.



Figure 3: Bayesian network for vessel ullage hydrogen concentration with Prior probability values

The results in Figure 3 indicate that in comparison with the equilibrium hydrogen volume, the sudden release volume results in a far greater proportion of the gas in the ullage space. This release mechanism is accountable for 71% of the ullage concentration population being above the 4% v/v LFL. Therefore if the likelihood of factors resulting in a sudden release is decreased, this should result in a direct reduction of the ullage concentration. As shown in Figure 3, the variables which have a direct effect on the likelihood of a sudden release are the primary nodes 11 and 12, crane PLC and zoning system failures.

The Bayesian updating feature is able to apply abductive reasoning to update the posterior probability of the primary nodes which are conditional to the occurrence of the top event. In other words, the Bayesian technique can undertake a 'reverse

analysis' to determine the effect on Parent node probabilities conditional to the top event probability. Such information is extremely beneficial when undertaking a sensitivity analysis. The reason being, it provides an understanding of how significantly the failures of system components would need to reduce in order to achieve a specified reduction in the likelihood of the top event.

Obviously the desired outcome for the Bayesian model in Figure 3 would be if the probability density for ullage hydrogen concentration (Node 15) is 100% in the 0-4% v/v range. Accordingly, the Netica software updating feature was applied to the top event Node 15, by instantiating the distribution of the 0-4% v/v concentration range to 100% probability (Figure 4). As discussed in the preceding paragraph, this updating was undertaken to identify the Parent nodes with the greatest sensitivity. The updated BBN in Figure 4 shows that in order for the ullage hydrogen concentration to keep within the 0-4% v/v range, the likelihood of the sudden release, Node 13, would need to decrease from the original 6.83% to 0.9% probability. Figure 4 shows that this reduction is only possible if the Primary nodes 11 and 12, i.e. crane PLC and zoning system failure probabilities reduce from the original 0.14% and 6.7% (Figure 3) to 0.02% and 0.9% respectively.

The updated BBN in Figure 4 also shows that the ullage concentration is less sensitive to the vessel ullage volume (Node 2) and equilibrium hydrogen volume (Node 1) as there is only a marginal change in the probability distributions for these two variables. This reaffirms the conclusion that the key sensitive factors which present the greatest impact on the vessel ullage hydrogen concentration are the crane PLC and vessel transfer zoning system failures. If the unavailability of these systems is reduced to 0.02% and 0.9% the risk of hydrogen concentration in the ullage space exceeding the 4% v/v LFL could be reduced to negligible.



Figure 4: Updated Bayesian network for vessel ullage hydrogen concentration

Case Study 2- Application of Bayesian Belief Networks to assess reliability of a purge air ventilation system for radiolytic hydrogen management

A BBN analysis was also undertaken to determine the unavailability, i.e. the probability of failure of the purge air delivery system for dilution of radiolytic hydrogen generation in a process vessel. The BBN is based on the key variables, their interactions and the individual primary event failure probabilities discussed in Section 4 and summarised in Table 1. The purpose of this BBN analysis is to determine how reliable the combined purge air delivery plant is and also to establish the key sensitivities that contribute to the unavailability of the system. Given that the purpose is to model the reliability of the system, it is considered that each item of equipment or operator error can be either in a failed or operational state. Hence the Bayesian Network for this scenario models all of the parents and children as discrete nodes with the failure being either in a 'true' or 'false' state. The BBN based on the Prior failure probabilities of the primary nodes, is given in Figure 5 with numerical results for each node presented as a bar chart. Netica software converts all probability values of the primary events, as given in Table 1, to percent probabilities. Therefore all probability values shown in Figure 5 are percent probabilities.



Figure 5: BBN for unavailability of the purge air system with prior failure probabilities of the primary events

The results show that the unavailability of the purge air system is 1×10^{-6} . The actual failure probability value is stated as 0+ in the node title 'Top Event', for the true state in Figure 5. This is due to a default in the Netica software such that all low probability values below 1×10^{-5} are stated as 0+. The results also show that the back- up compressor failure ('BACKUP') and emergency diesel compressor system failure ('EMRGF') at 7.65% and 0.16% probability respectively are accountable the Top Event probability of 1×10^{-6} .

Whilst a low unavailability of 1×10^{-6} indicates that the combined normal and back up purge air system is highly reliable, it is still useful to identify the main failure modes contributing to the Top Event and to determine how significantly the individual failures would need to increase if the Top Event occurs indefinitely. Thus by instantiating the Top Event probability to 100%, using the Netica updating feature, Figure 6 shows the posterior probabilities of each of the nodes. It can be observed from Figure 6 that the primary events 'Loss of factory air (FACTAIR)' and 'POWER' present the greatest contribution to the occurrence of the Top Event.



Figure 6: Updated BBN for unavailability of purge air system

Conclusions

Hydrogen gas generation due to corrosion of fuel cladding waste and, to a lesser extent, radiolysis of aqueous solutions presents a significant challenge in nuclear decommissioning and reprocessing operations. Both of these release mechanisms and the associated hazard management strategies result in complex interactions and uncertainties. Hold-up of hydrogen gas and a subsequent sudden release from sludge wastes is a key example in this respect. Slow release of hydrogen gas from radiolysis of aqueous solutions also requires robust hazard management strategies such as highly reliable purge air ventilation systems.

A BBN analysis has been carried out for a study relating to a sudden release of hydrogen in a lidded storage vessel containing radioactive magnesium hydroxide sludge. A BBN analysis for a second case study to assess key sensitivities affecting the availability of a forced ventilation system for dilution of radiolytic hydrogen was also undertaken. From the analysis it is concluded that the main factors which present the greatest impact on the vessel ullage hydrogen concentration following a sudden release of hydrogen are the crane PLC and vessel transfer zoning system failures. For the second case study, it is demonstrated that the loss of factory air and power supply to the emergency diesel air compressor present the greatest contribution to the unavailability of the forced ventilation system.

Nomenclature

| Ceq | Equilibrium hydrogen in air concentration, %v/v |
|---------------------------|--|
| E _(a) | Rate of absorption of alpha radionuclide decay energy by the medium, MeV/s |
| Ε(β/γ) | Rate of absorption of beta gamma radionuclide decay energy by the medium, MeV/s |
| $G(H_2)_{(\alpha)}$ | Number of hydrogen molecules evolved per 100eV of alpha decay energy absorbed by the medium |
| $G(H_2)_{(\beta/\gamma)}$ | Number of hydrogen molecules evolved per 100eV of beta gamma decay energy absorbed by the medium |
| k | dimensional constant, 1.44x10 ⁻¹⁵ |
| P(A) | Probability of occurrence of hypothesis A |
| P(A B) | Conditional probability for the likelihood of observing hypothesis A given that B is true |
| P(B) | Probability of occurrence of event B |
| P(B A) | Probability of event B occurring given that A is true |
| Qair | Purge air volumetric flow rate, l/hr |
| Qн | Radiolytic hydrogen generation rate, l/hr |
| t | Time required to reinstate purge air before exceeding 1%v/v hydrogen concentration limit |
| U | Vessel ullage space volume, l |

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References

- 1. Ahmed, F., 2019, Managing hydrogen gas hazards, *Nuclear Future, Journal of the Nuclear Institute*, Fuel Cycle theme, Issue 15.2.
- 2. Bibler N.E, Pareizs J.M, Fellinger T.L, Bannochie C.J., 2007, Measurement and Prediction of Radiolytic Hydrogen Production in Defence Waste Processing Slurries at Savanah River Site, WM 07 Conference, 1-15.
- 3. Bolstad, W. M., 2007, Introduction to Bayesian Statistics, Second Edition, ISBN-0-470-14115-1.
- 4. Gharari, R., Kazeminejad, H., Mataji, N., Kojouri, A., Hedayat A., 2018, A review on hydrogen generation, explosion, and mitigation during severe accidents in light water nuclear reactors, *Int J Hydrogen Energy*, 43:1939-1965.
- 5. Nuclear Decommissioning Authority, 2016, Strategy, Effective from April 2016, Nuclear Decommissioning Authority, 2016. ISBN 9781474130431, Available At: https://www.gov.uk/government/publications.
- 6. Health and Safety Executive, 2014, Office for Nuclear Regulation, Safety Assessment Principles for Nuclear Facilities, Revision 0, 2014 Edition. Available at www.onr.org.uk/saps/saps2014.pdf.
- 7. Ingram, J.M., Kempsell, I.D., Wakem, M.J., Fairclough, M.P.,2001, Hydrogen explosion An example of hazard avoidance and control, *Hazards XVI*, IChemE Symposium Series No. 148:523-539.
- 8. Van Kessel, T., van Kesteren, W.G.M., 2002, Gas production and transport in artificial sludge depots, *Waste Management*, 22:19–28.
- 9. Netica, Norsys, Available at www.norsys.com.
- 10. Pasman, H.J., Rodgers, W.J., Risk assessment by means of Bayesian networks; A comparative study of compressed and liquefied H₂ transportation and tank risks, 2012, *Int J Hydrogen Energy*, 37:17415-17425.