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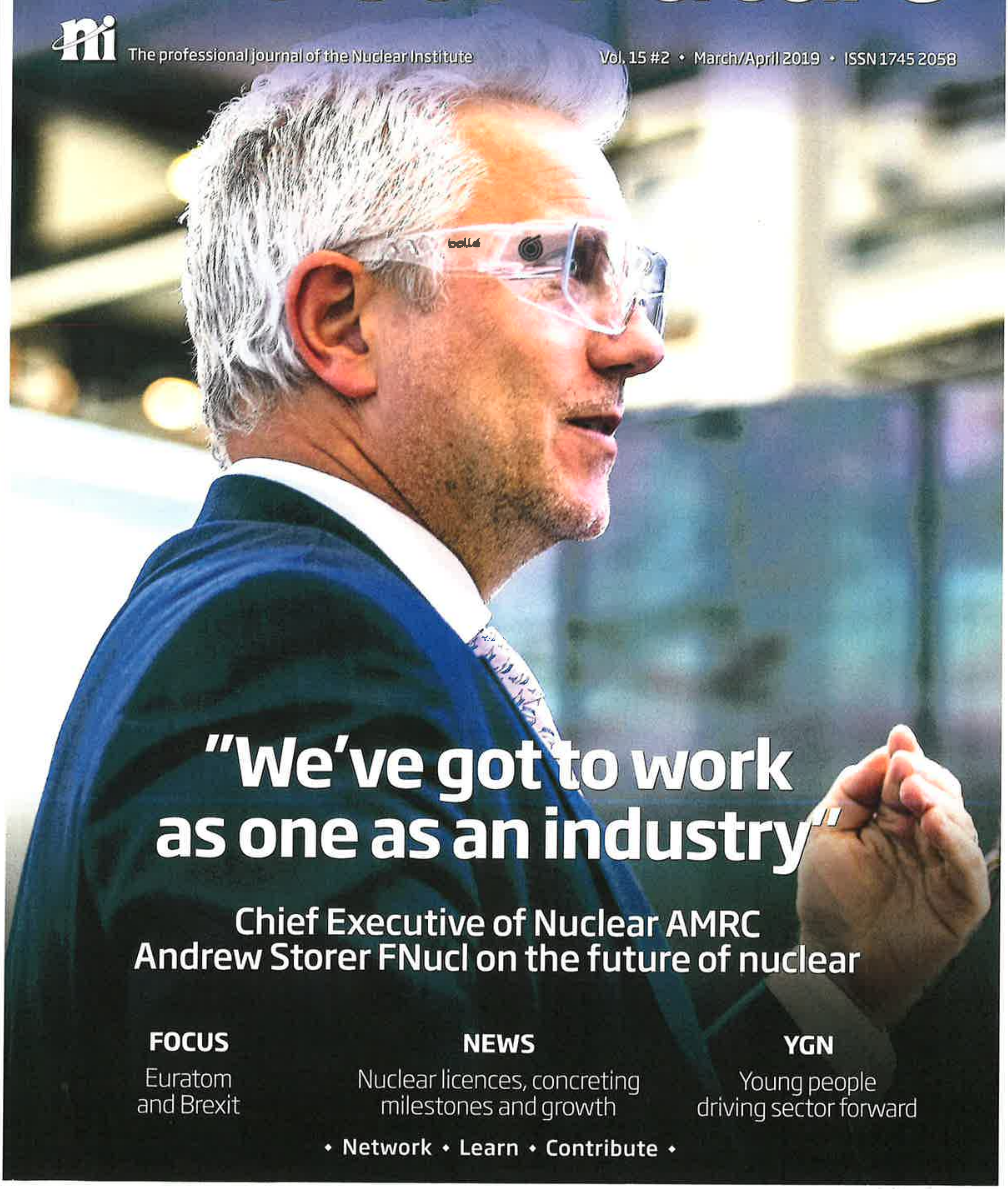

"We've got to work as one as an industry"

Chief Executive of Nuclear AMRC Andrew Storer FNucl on the future of nuclear

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and Brexit

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Nuclear licences, concreting
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Managing hydrogen gas hazard uncertainty

SUMMARY

- There is an uncertainty associated with the risk from hydrogen gas generated in nuclear waste processing and decommissioning operations.
- Bayesian belief networks provide an improved means of assessing the risk of hydrogen explosions.
- The Bayesian belief network technique has been applied in a case study to identify the main sensitivities associated with hydrogen generation in a vessel ullage space.
- The updating capability of Bayesian belief networks has shown that gas hold-up and discontinuous release are the key factors affecting hydrogen concentration in the vessel ullage.

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1. INTRODUCTION

The nuclear waste processing and decommissioning sector faces a key challenge of managing the risk from the generation of hydrogen gas. The properties of hydrogen are unique in that its flammable concentration band is wide and the energy required for ignition is very low. As such the likelihood of ignition of this gas is higher in comparison with other combustible gases [1]. The regulatory framework for nuclear installations and legislation require that the risk of hydrogen explosions is 'as low as reasonably practicable' (ALARP). This suggests that the quantified risk assessment must be plausible. An inaccurately assessed risk can potentially lead to decision making which is not fit for purpose.

Most hydrogen management strategies for storage and processing of nuclear waste, under normal plant operations, employ measures for instance control of ullage volume. Sufficient ventilation extract routes are also required to prevent formation of flammable gas mixtures in vessels. However parameters such as ullage volume can vary and are dependent on other variables including the potential for hydrogen hold-up leading to waste matrix expansion. Such variations and dependencies can lead to an uncertainty in quantification of the risk of hydrogen explosions.

In this paper an emerging statistical technique known as Bayesian belief networks (BBN) is explored as an improved means of quantifying the risk from hydrogen generation during processing of nuclear waste. The BBN technique is applied in a case study to predict the likelihood of a flammable mixture of hydrogen in air developing in the ullage space of a transportable process vessel containing intermediate level wastes (ILWs).

2. APPROACHES TO QUANTIFIED RISK ASSESSMENTS FOR NUCLEAR SAFETY CASES

Standard industry practice for a quantified risk assessment [2, 3] of an accident scenario is to initially conduct hazard and operability studies and identify credible faults at a facility. This is followed by a hazard analysis. Fault tree analysis (FTA) and event tree analysis (ETA) are often undertaken in support of hazard analysis to assess the quantified risk. A major disadvantage of both the FTA and ETA methods is that they are unable to adequately represent the uncertainty and dependencies between factors in complex systems such as hydrogen generation.

BBN [3] is an alternative technique which can provide a means for overcoming the limitations of FTA and ETA, allowing uncertainty and dependencies between different factors to be taken into account. The main advantages of the BBNs are that they can use distributions rather than single probability values thus allowing an uncertainty analysis. Furthermore they can be used to update the likelihood of an event based on new evidence thus improving model accuracy.

3. BAYESIAN BELIEF NETWORK METHODOLOGY

3.1 Bayes theorem

The Bayesian network methodology is a graphical means for modelling relationships between the causal variables and effects of a particular event. Essentially it uses a statistical hypothesis known as the Bayes theorem which is based on the concept of conditional probability.

The term conditional probability can be defined as the probability of a hypothesis given the occurrence of another event. In terms of hydrogen safety, the hypothesis may be "what is the probability of a hydrogen explosion?" The use of conditional probability allows this uncertainty to be resolved by making use of a piece of evidence that affects the likelihood of the hypothesis. So the question would be phrased as "what is the probability of a hydrogen explosion given that a flammable hydrogen concentration arises in the vessel ullage space?" Based on these concepts, Bayes theorem [4] is expressed as:

$$P(A|B) = (P\ (A)\ x\ P(B|A))/P(B)$$

Equation 1

Where:

- P(A) and P(B) are the probabilities of observing events A and B independently of each other.
- P(A) is termed as the "prior probability" of the hypothesis before allowing for any evidence.
- P(A|B) is a conditional probability which represents the likelihood of observing event A given that B is true.
- P(B|A) represents the probability that event B occurs given that A is true.

Effectively Bayes theorem provides the relationship between the prior probability P(A) before any evidence is available and the likelihood of hypothesis A when evidence B has been allowed for, i.e. P(A|B).

Equation 1 can be applied to a typical hydrogen safety issue concerning the likelihood of a flammable hydrogen in air concentration > 4%v/v arising in the ullage space of a vessel. It is hypothesised that the probability of ullage concentration being > 4%v/v is P(A) and the probability of occurrence of a high hydrogen generation rate is P(B). In this case a high hydrogen generation rate relative to a fixed ullage volume is assumed to result in a flammable hydrogen concentration. The following hypothetical example illustrates how Bayes theorem can be used to update the likelihood of flammable hydrogen concentration with new evidence.

Hypothetical example

A series of 3000 process vessels is considered in which one incident of a high hydrogen concentration in the vessel ullage was previously observed. This gives a prior probability P(A) = 1/3000 or 0.03%. If it is assumed that upon experimental trials, a high hydrogen generation rate was observed in 7% of the vessels, indicated by gas bubbles observed at the liquor surface, then P(B) = 0.07. If it is also assumed that the probability that the hydrogen generation rate would have been high given that a flammable hydrogen concentration arose, then P(B|A) = 1. Applying Bayes theorem signifies that if a high hydrogen generation rate is detected in a vessel, the probability of a flammable concentration would rise from 0.03% to 0.43% (P(A |B) = (1 x 0.0003)/0.07 = 0.43%). This clearly shows the probability updating capability of Bayes theorem confirming that the probability of finding a flammable atmosphere has increased owing to new evidence. Equation 1 above applies Bayes theorem to a straight forward uncertainty analysis with only two variables. However when a large number of events and causal combinations are involved, the Bayesian algorithm for estimation of the likelihood of the hypothesis would become extremely complex and difficult to calculate manually. Software systems such as Netica [5] are available commercially based on the application of Bayes theorem, which enable modelling of complex hypotheses in the form of a cause and effect network. This is commonly referred to as the Bayesian belief network (BBN).

3.2 Process for Bayesian belief network analysis

A BBN is a directed acyclic graph which identifies believed relations, i.e. cause and effect, between a group of variables relevant to a hypothesis. A typical hypothesis modelled in a BBN in terms of hydrogen safety would be the risk of a hydrogen explosion occurring in the ullage space of a vessel.

During construction of the BBN, if there is a cause and effect interaction between two variables or nodes, the two nodes are linked by an arc. For example in Figure 1 an arc from nodes E1 and F3 to node E2 indicates that the random variables E1 and F3 (often termed as 'parent nodes') cause the random variable E2 ('child node').

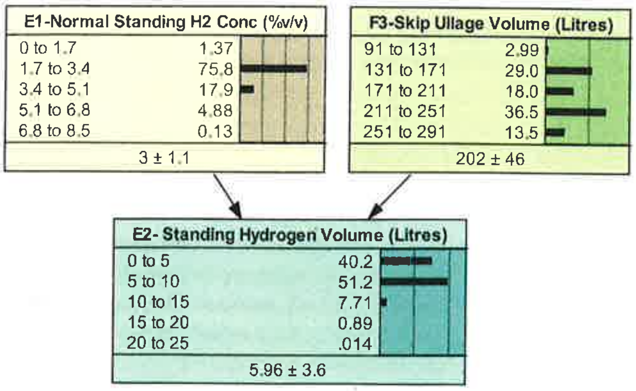

FIGURE 1: Simple Bayesian network with two parents and one child node

One node is used for each variable, which may be either 'discrete' or 'continuous'. A discrete node consists of a set of possible states, for example the answer to a question i.e. 'yes' or 'no' or a state which is 'true' or 'false'. A continuous node consists of a range of values which may be defined as a probability distribution, e.g. Normal Distribution. All three of the nodes in Figure 1 are continuous, defined by probability distributions with mean values of 3%v/v, 5.96 litres and 202 litres.

Conditional probability tables are specified within the network, derived using experimental data, mathematical model equations or expert opinion. A conditional probability distribution for a child node indicates the probabilities of the node which are dependent on the values of its parent node. Figure 2 illustrates the generic process for construction of a BBN.

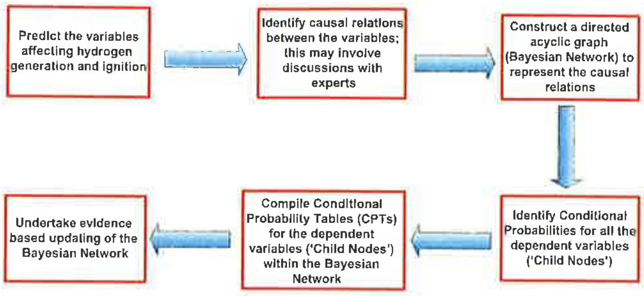

FIGURE 2: Generic process for Bayesian belief network analysis

4. CASE STUDY

Application of Bayesian belief networks to assess hydrogen concentration in a process vessel containing intermediate level wastes.

4.1 Case study description

The BBN technique was applied to the following plant case study to predict the likelihood of a flammable hydrogen in air concentration developing in the ullage space of a transportable process vessel. The case study considers a process vessel, referred to as the 'skip' hereafter, which contains ILWs for interim safe storage. The waste comprises a mixture of Magnox, cover liquor and magnesium hydroxide sludge which arises from underwater corrosion of the magnesium metal. Hydrogen gas releases within the skip due to continuous corrosion of Magnox and radiolysis of the skip aqueous liquor. The skip lid consists of filtered outlets to enable the hydrogen to be vented to atmosphere while retaining airborne particulate. The skip is housed in an outer box for the purpose of providing containment. A schematic of the skip is shown in Figure 3.

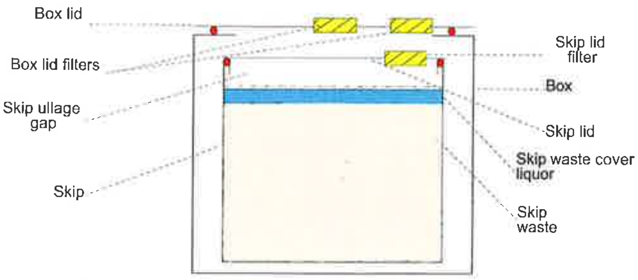

FIGURE 3: Schematic of the ILW skip

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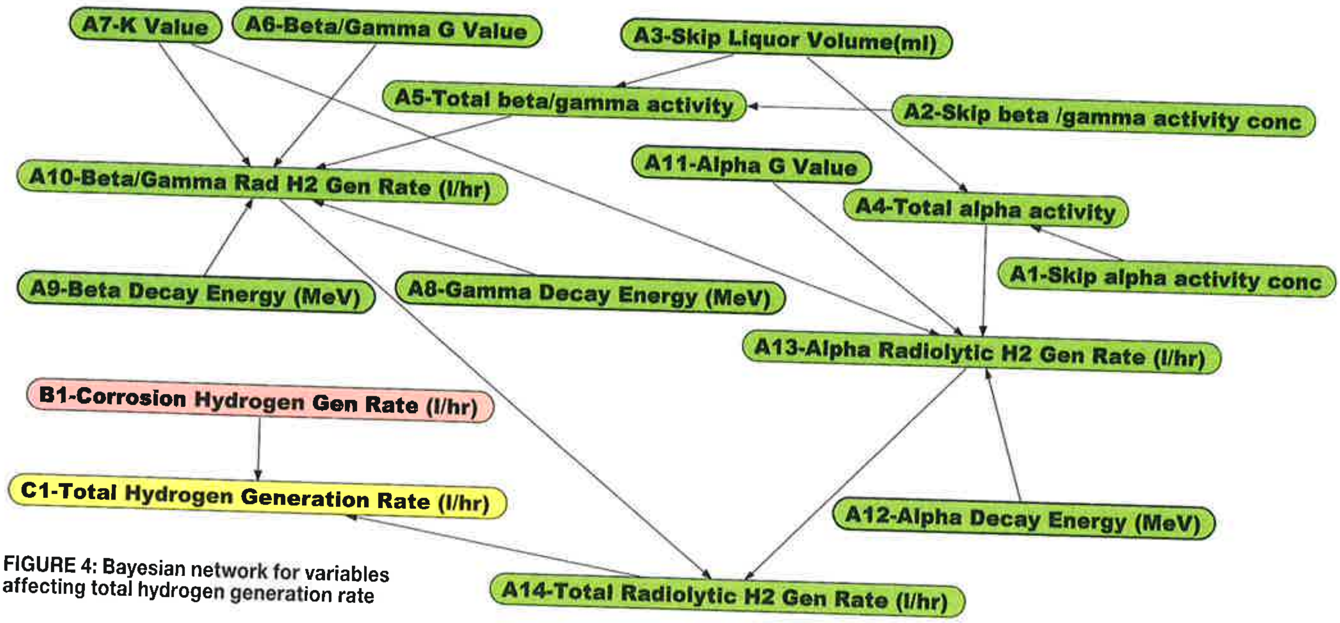

FIGURE 4: Bayesian network for variables affecting total hydrogen generation rate

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A knowledge gap has been identified in terms of hydrogen hold-up within the sludge and waste matrix and the potential for a discontinuous release of the gas into the ullage during skip handling. A discontinuous release of hydrogen refers to a mechanism whereby the hydrogen generated via metal corrosion and radiolysis is not released from the waste bed at the continuous rate at which it evolves. Instead, hydrogen is able to build up as bubbles and pockets of gas within the waste matrix. Beyond a certain point the forces constraining the movement of these gas bubbles are overcome and a sudden release of a significant proportion of the held-up gas can occur. Such a release could occur during skip movements resulting in a concentration of hydrogen in the ullage space that is close to or above the lower flammable limit.

This case study investigates the application of the BBN methodology to identify key sensitivities which would affect the likelihood of a flammable hydrogen in air mixture forming due to continuous and discontinuous releases. The application of the BBN technique to hydrogen generation in nuclear decommissioning environments has recently been reported by London South Bank University [6]. However reference [6] focussed on factors that affect hydrogen generation rate due to corrosion. This paper takes into consideration the uncertainty from other variables, primarily hydrogen gas hold-up and discontinuous releases.

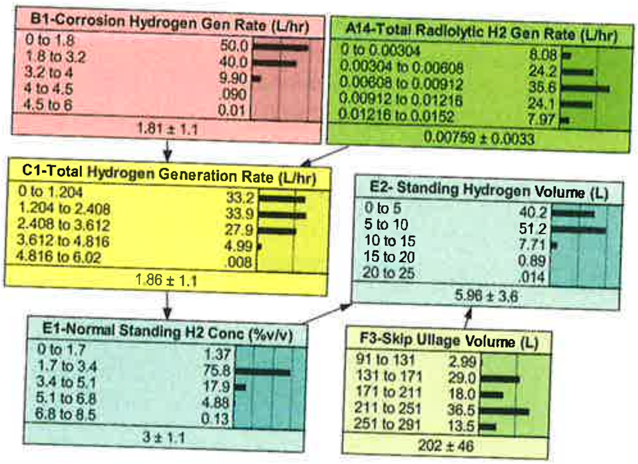

FIGURE 5:
Quantified Bayesian network for uncertainty analysis of hydrogen concentration in ILW skip (continuation from Figure 4 Node A14)

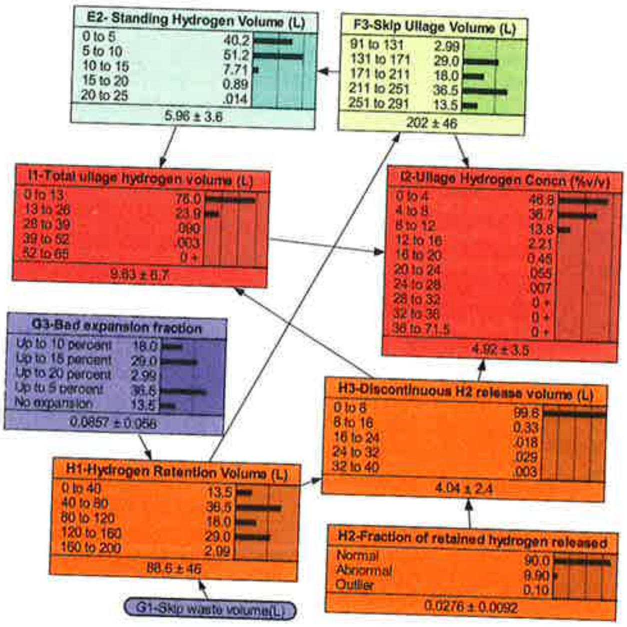

FIGURE 6:
Quantified Bayesian network for uncertainty analysis of hydrogen concentration in ILW skip (continuation from Figure 5 node F3)

4.2 Identification of the key variables and dependencies for the Bayesian network

Initially the key variables, i.e. the parent and child nodes which affect hydrogen concentration in the skip ullage, were identified. This was achieved through discussions with a team of specialists in the field of sludge and hydrogen gas behaviour. A BBN cause and effect diagram was then constructed using Netica software [5]. For the purpose of presentation in this paper, the BBN is split into three figures as shown in Figures 4, 5 and 6. The key variables and dependencies in these figures are discussed below.

Hydrogen generation rate

The main mechanism of hydrogen generation in skips of ILWs was considered to be corrosion of the waste. Whilst radiolysis of the skip liquor was deemed a secondary mechanism, it was still considered beneficial to explore this additional source of uncertainty.

Reference 7 considers that the rate of radiolytic hydrogen generation from radioactive liquors is directly proportional to the amount of decay energy absorbed by the liquid and the experimentally determined G(H2) values. The G(H2) value represents the number of molecules of hydrogen produced for every unit of decay energy, MeV, absorbed. Using this concept from reference [7], the hydrogen generation rate due to radiolysis can be expressed as:

$$Q_H = kG(H_2)_{(\alpha)}E_{(\alpha)} + kG(H_2)_{(\beta\gamma)}E_{(\beta/\gamma)}$$

Equation 2

Where:

- Q_H = radiolytic hydrogen generation rate at room temperature and pressure, (litres/hr)
- $G(H_2)_{(x)}$ = G-value, the number of molecules of hydrogen evolved per 100 eV of radiation x, where x is alpha (α) or beta gamma (βγ) radiation
- $E_{(x)}$ = rate of absorption of energy x by the liquid/solid (MeV/s)
- K = dimensional constant (1.44×10^{-15}).

The value of E in MeV/s was obtained by multiplying the activity quantity in Becquerels by the α and βγ decay energy in MeV. G values of 1.66 and 0.45 Molecules/100eV are known for α, and βγ nuclides in aqueous liquors [7]. The skip α and βγ activity quantity was determined from the activity concentration, knowing the skip liquor volume. Hence, knowing the G value, α and βγ activity quantity, decay energy and the k value, the Bayesian model used Equation 2 to predict the distribution of the alpha and beta gamma radiolytic hydrogen generation rates. The alpha and beta gamma radiolytic hydrogen generation rates were summed to obtain the total radiolytic hydrogen generation rate. These results are shown in node A14 of Figure 5.

Corrosion hydrogen generation rate was considered to be primarily dependent on the proportion of un-corroded Magnox. The probability distribution as shown in Node C1 of Figure 5 with a mean rate of 1.86 L/hr was considered appropriate by the hydrogen and sludge specialist team. As the total radiolytic hydrogen generation rate of 0.0076 L/hr is very small in comparison with the rate due to corrosion, it is confirmed that the BBN results are less sensitive to the variables affecting the former mechanism. For this reason the quantified results of the parent nodes affecting radiolytic hydrogen generation rate are not shown in Figure 4 and only a cause and effect structure is presented.

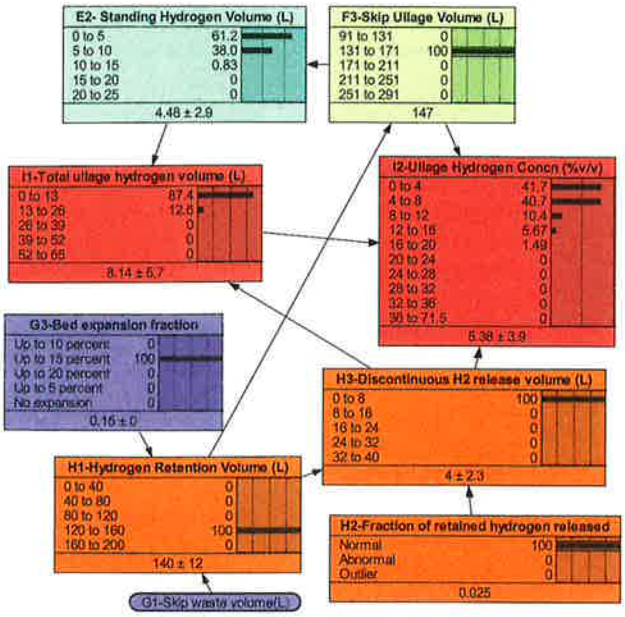

FIGURE 7:
Updated Bayesian network for uncertainty analysis of hydrogen concentration in ILW

Hydrogen retention and discontinuous release

The hydrogen discontinuous release volume from the skip waste matrix is dependent on the volume of hydrogen retention (Figure 6 Node H1) and the degree of waste swelling, if sludges are involved. Filter performance also affects the volume of hydrogen accumulating in the ullage.

Skip ullage volume

Intuitively for a given volume of hydrogen released, its concentration in the ullage is inversely proportional to the ullage volume. The ullage volume is dependent on:

- degree of waste swelling leading to a reduction in ullage volume,
- total volume of the skip contents, assuming the skip is full.

4.3 Key findings from case study

Figures 5 and 6 show the quantified results of the BBN analysis. The model is based on the following prior nodes which required input of probability distributions using expert opinion:

- skip alpha activity concentration (Node A1),
- beta/gamma activity concentration (Node A2),
- skip liquor volume (Node A3),
- corrosion hydrogen generation rate (Node B1),
- skip waste volume (Node G1),
- release fraction of retained hydrogen (Node H2).

The conditional probabilities for all the remaining nodes were calculated by the network using equations based on the dependencies between variables as discussed above. The main results from the BBN analysis are that at a skip mean ullage volume of 202L (Figure 5 Node F3) and a total mean ullage hydrogen volume of 9.6L (Figure 6 Node I1), a hydrogen concentration of <4%v/v at 47% probability is predicted. The contribution from discontinuous

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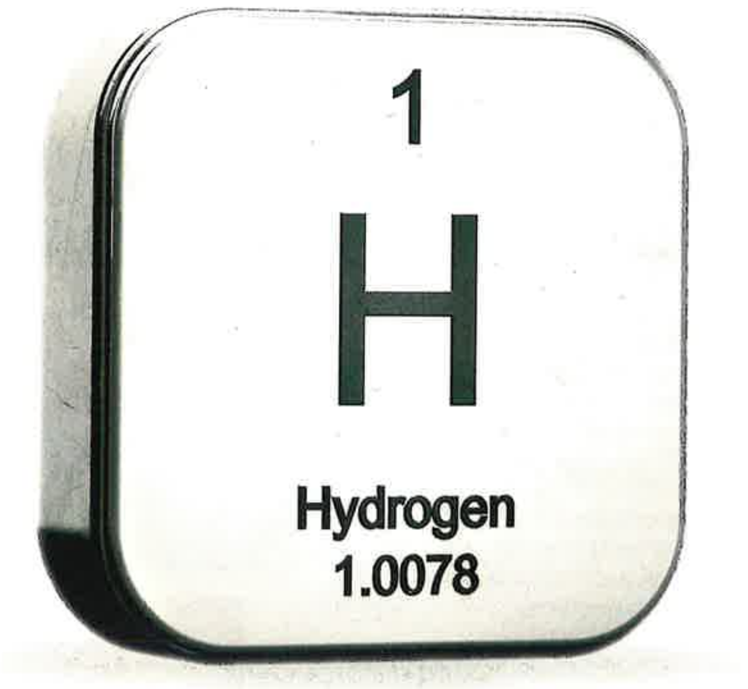

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release is a key sensitivity affecting hydrogen concentration.

One of the distinct features of BBNs is that they can incorporate evidence observed for a particular case to update the predictions of the network, thus enabling a sensitivity analysis to be performed. Utilising the Netica software updating function, the model was rerun with observed values of the key parameters, as identified by the specialist team (Figure 7). The observed values included a discontinuous release fraction of 0.025 (Node H2) and an ullage volume of 147L (Node F3). Figure 7 is also based on corrosion hydrogen generation rate of 2L/hr, by updating Node B1 of Figure 5 to this value. At these observed values, Figure 7 Node I2 predicts a mean ullage hydrogen concentration of 5.38%v/v. This increase in hydrogen concentration is as expected due to the higher hydrogen generation rate used in comparison with the analysis in Figures 5 and 6.

5. CONCLUSIONS

Using a case study for a process vessel containing ILWs, the Bayesian belief network technique has been applied to undertake an uncertainty analysis of hydrogen concentration in the vessel ullage. The key sensitivities affecting hydrogen concentration are the rate of hydrogen generation, hold-up of hydrogen gas within the waste matrix and the subsequent discontinuous release as well as the factors that govern ullage volume.

By using best estimates of the prior distributions of the Bayesian input nodes, it has been demonstrated that the contribution from discontinuous release is a key sensitivity affecting hydrogen concentration. If the likelihood of discontinuous release is reduced by minimising waste disturbance and the waste processing time prior to interim storage, then the probability of exceeding the lower flammable limit could be shown to be negligible.

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Acronyms

ALARP	As low as reasonably practicable
BBN	Bayesian belief network
ETA	Event tree analysis
FTA	Fault tree analysis
ILW	Intermediate level waste

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Fayaz Ahmed has over twenty years of experience in radiological safety assessments. He is currently undertaking part time PhD research at London South Bank University. His area of research is the application of novel quantified risk assessment techniques to hydrogen hazard analysis for nuclear chemical plants.

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