SAFETY MANAGEMENT OF A NUCLEAR-BASED HYDROGEN GENERATION SYSTEM

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Hydrogen energy is a clean energy carrier that is expected to alleviate current energy-related problems such as global climate change, air pollution and depleting fossil energy resources. Among all the hydrogen generation methods, the Canada Deuterium Uranium (CANDU) Supercritical Water Reactor (SCWR)-based Cooper-Chloride (Cu-Cl) thermochemical cycle is a promising method for future large scale hydrogen generation, due to its high energy efficiency and low temperature requirement. However, this nuclear-based hydrogen generation system leads to new challenges for system safety management. Over the past decades, Probabilistic Safety Assessment (PSA) has been used successfully in the nuclear and other industries as an important tool to assess system safety and provide information for decision-makings on design, development, operation and maintenance. In this paper, the PSA methodology is utilized and extended to provide guidelines and recommendations for safety assessment and safety management of the nuclear-based hydrogen production system.

I. INTRODUCTION

Hydrogen energy is a clean energy carrier that is expected to alleviate the current energy-related problems such as global climate change, air pollution and depleting fossil energy resources. There are many methods to produce hydrogen, such as electrolysis hydrogen production and coal gasification. The method investigated in this paper to generate hydrogen is the nuclear-based hydrogen generation based on Cooper-Chloride (Cu-Cl) thermochemical cycle. The target Nuclear Power Plant (NPP) for the Cu-Cl cycle is the Canada Deuterium Uranium (CANDU) Supercritical Water Reactor (SCWR). The relatively high efficiency and low temperature requirement make this a promising method for future large scale generation of hydrogen energy. In September 2010, the University of Ontario Institute of Technology (UOIT) in Canada launched the Clean Energy Research Laboratory (CERL) officially. The primary target of CERL is to move clean hydrogen production from laboratory to industrial use. CERL is working on the world's first lab-scale demonstration of Cu-Cl cycle for water splitting and hydrogen production.

This new method for hydrogen generation based on waste heat from CANDU-SCWR leads to new challenges for system safety analysis and management. On the one hand, the hydrogen production facility leads to external risks to the reactor safety through a direct interconnection with the reactor's coolant system. The impact of hydrogen plant on the nuclear reactor safety should be carefully studied during all life cycle stages of the nuclear-based hydrogen generation project, to avoid any potential nuclear accidents due to failures of the hydrogen plant. On the other hand, the hydrogen production facility itself encompasses a set of chemical processes which are inevitably exposed to internal process risks such as fire, explosion, and release of toxic or flammable chemicals. Both the internal and external risks from the thermochemical water splitting process should be managed through safety assessment to reduce the overall risk of the co-generation plant to an acceptable level.

Safety management involves a set of principles, analysis, regulations and decisions to prevent injuries, death and property losses, which may be caused by potential risks within a process or a product. Probabilistic Safety Assessment (PSA) has been used successfully in nuclear and other industries as an important tool to assess system safety and provide information for decision-makings on design, development, operation and maintenance of plants and facilities. In this paper, the PSA methodology is applied and extended for safety assessment and safety management of the nuclear-based hydrogen production plant. The preliminary safety assessment result of the nuclear-based hydrogen production plant is obtain to quantitatively identify potential risks in the nuclear-based hydrogen cogeneration plant.

II. DESCRIPTION OF THE NUCLEAR-BASED COOPER-CHLORIDE THERMOCHEMICAL CYCLE

The Cooper-Chloride (Cu-Cl) cycle consists of a set of chemical reactions to form a closed internal loop cycle to generate hydrogen from the thermochemical decomposition of water into hydrogen and oxygen. The intermediate copper and chloride compounds are recycled and reused within the thermochemical loop to continuously generate hydrogen without emitting pollutions and greenhouse gases. There are different variations of Cu-Cl cycle based on the thermochemical water splitting: five-step, four-step and three-step [1], and they all have the same overall reaction: $H_2O(g) \rightarrow H_2(g)+1/2O_2(g)$. The safety study in this paper is based on a four-step Cu-Cl cycle which has been integrated and demonstrated for lab scale hydrogen production in the Clean Energy Research Laboratory (CERL) at UOIT. The reactions involved in the four-step Cu-Cl cycle are shown in Table I.

TABLE I. Reactions in the Four-step Cu-Cl Cycle

Step	Reaction	Temperature(˚C)
1. Hydrogen production	2CuCl(aq)+2HCl(aq) →2CuCl2(aq)+H2(g)	<100 (electrolysis)
2. Drying	CuCl2(aq) →CuCl2(s)	<100
3. Hydrolysis	2CuCl2(s)+H2O(g)→Cu2OCl2(s)+2HCl(g)	400
4. Oxygen production	Cu2OCl2(s) →2CuCl(l)+1/2O2(g)	500

To produce 1 kg of hydrogen from the Cu-Cl cycle, 220 MW of thermal energy is required at a temperature up to approximately 530˚C. Nuclear-based hydrogen generation is a promising solution for large scale commercial hydrogen generation. One of the nuclear reactors considered as the energy source for the Cu-Cl thermochemical cycle is the Supercritical Water Reactor (SCWR). The SCWR is a Generation IV nuclear reactor which uses Super Critical light Water (SCW) as the coolant at pressure up to 25 MPa and temperature as high as 625˚C. Theoretically, coolant from the reactor outlet could be delivered directly to the hydrogen facility to power the hydrogen generation and this direct linkage of the heat transfer system would improve the thermal efficiency of the overall hydrogen generating system. However, delivering the coolant could significantly extend the NPP's containment size which introduces new regulatory complexities. Also, the NPP and hydrogen plant are operating under different pressure conditions, as up to 25 MPa in the NPP and several atmospheres in the hydrogen facility. The direct linkage could cause huge stress for the materials of piping and equipment in the hydrogen plant due to the effect of large pressure differences. Thus, an intermediate heat exchanger would be used as an interface transferring heat from the NPP's coolant system to the hydrogen generation facility to lower the pressure experienced by the hydrogen plant, as shown in Fig. 1 [2].

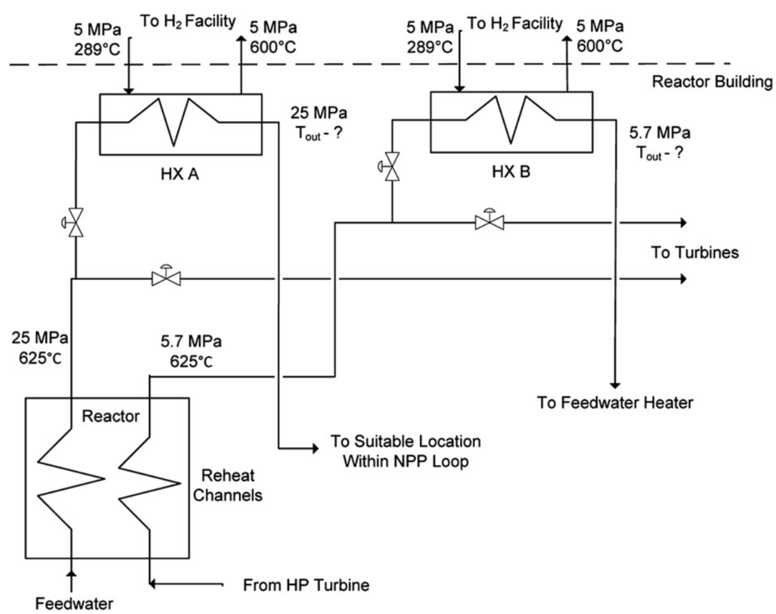


Fig. 1. Potential HX locations for SCW NPP layouts [2]

III. PSA-BASED SAFETY MANAGEMENT OF NUCLEAR-BASED HYDROGEN GENERATION

The details of the PSA-based safety management of the nuclear-based hydrogen production system are represented in this section. A preliminary safety assessment is performed first, based on currently available design information to find the major hazard and risks in the Cu-Cl cycle, and assess their impact on the plant safety. As a conceptual design phase safety study, only severe accidents are considered. The safety assessment involves the hazard identification, fault tree modeling for initiating events and event tree modeling for accident sequences. Numerical results are derived from this safety assessment, and these results are used to determine whether an additional safety system is required.

III.A. Hazard and Risk Identification of Nuclear-based hydrogen Production System

To start a PSA, hazard and risk in a process need to be identified first. The new safety challenge caused by nuclear-based hydrogen production is the major concern of this study. To identify the risk and hazard in the Cu-Cl cycle, expert judgment for nuclear hydrogen production is referred. According to the safety study in previous literature [3], the following issues would be the new risks associated with a nuclear-based hydrogen facility:

- Toxic chemical species: Hydrochloric (HCl) acid for the Cu-Cl cycle
- Hydrogen production and storage in large quantity: hydrogen safety for production and storage
- Heat transfer fluids, additional thermo-hydraulic loop in the nuclear plant, and Loss of Coolant Accident (LOCA)

Although Hydrochloric acid is mentioned as a potential risk, it is not classified as major risk in this study. This is because the hydrochloric acid has an irrigating and pungent odor even at very low concentration. It is very easy to detect when the HCl is released into the environment. Therefore, it is assumed that the operator can stop a release of HCl in early stage by shutting down the process reaction. On the other hand, HCl is an intermediate chemical in the Cu-Cl thermochemical process, where it is generated and consumed within the reactions. Although the total amount of HCl required for daily hydrogen production is very large, the actual amount existing in Cu-Cl cycle is small. Therefore, the release could have minor effect. Thus, the LOCA and the hydrogen accident are analyzed as the major risks introduced by Cu-Cl cycle by PSA-based methods in this study.

III.B. LOCA Analysis

Loss of Coolant Accident (LOCA) is an accident that coolant is released from the heat transport system. When LOCA occurs, the nuclear reactor will loss part or total of the cooling ability to keep the reactor core in a stable state and it is the possible cause of some severe nuclear accident such as reactor core damage. The cause and effect of coolant release must be studied carefully to ensure a safer design.

A double pipe intermediate heat exchanger is proposed as an interface for transferring thermal energy from the reactor coolant system to the hydrogen production plant [4]. The heat exchanger would be interfaced with the no-reheat alignment of coolant cycle or the single reheat alignment of the coolant cycle. In both linkage options, a location on the coolant loop downstream of the reactor and upstream of the turbine would be a suitable location for the heat exchanger. In this case, SCW is the operating fluid on the primary loop of heat exchanger. For the single reheat cycle, a second available location would be downstream of the steam reheat channels and the operating fluid is the superheated steam.

In both linkages, the reactor coolant is bypassed to the heat exchanger to power the hydrogen production and then mixed back with the reactor coolant main stream. Although the heat exchanger thermal hydraulic behavior could be different between these two linkage options due to different operating fluid property, the safety performance of these two systems could be similar, since both systems share the same control and instrumentation structure. For plant balance, the amount of coolant delivered to the hydrogen plant is determined based on the electrical and hydrogen generation demand, which is controlled by a control valve located at the downstream of the reactor core. The heat exchanger located at the downstream of the reactor in a no-reheat coolant cycle is selected for the PSA study in this paper. The heat exchanger loop for hydrogen production consists of:

- Two isolation valves: isolate the heat exchanger from the NPP coolant system
- Pipes
- Control valve: control the flow rate of heat exchanger primary loop
- Intermediate heat exchanger: double pipe or tube and shell
- Mix valve: mix the operating fluid from downstream of heat exchanger with NPP coolant main stream

The Piping and Instrumentation Diagram (P&ID) of the heat exchanger is shown in Fig. 2.

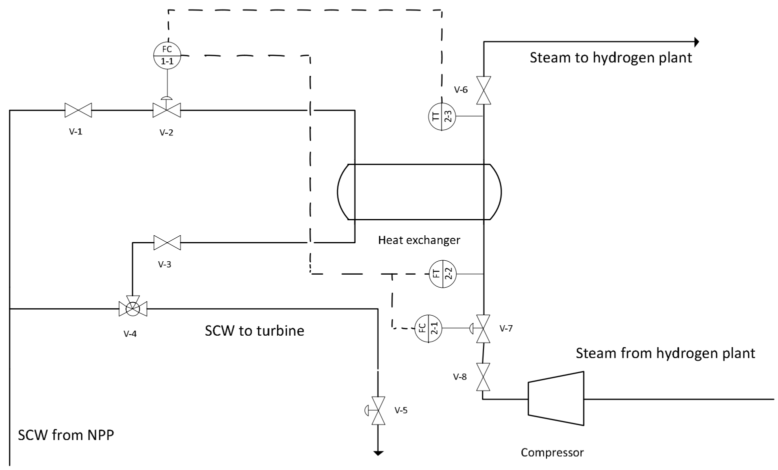


Fig. 2. P&ID of the Heat Exchanger

III.B.1. LOCA Probability Identification by FTA

LOCA is caused by any leakage in the heat transfer interface piping. To identify the initial event frequency of a LOCA due to hydrogen production, a fault tree is developed to calculate the LOCA probability as a top event. The fault tree is shown in Fig. 3. The failure rates of the basic events are taken from [5, 6], as shown in Table II. In particular, the piping failure rate is related to the length of the pipe. According to the current Cu-Cl design specifications, the heat exchanger is located in the nuclear containment building. Thus, it is assumed the pipe length of the heat exchanger primary loop is in the range of 100 meters. In this analysis, the value for pipe length is taken as 100 meter.

LOCA can be classified as Small Break Loss of Coolant Accident (SBLOCA) and Large Break Loss of Coolant (LBLOCA), depending on the leakage size in the coolant system. Based on the FTA, the SBLOCA due to hydrogen production is 1.33E-6/h and the LBLOCA frequency is 9.02E-8/h. This corresponds to 1.17E-2/y for SBLOCA and 7.9E-4/y for LBLOCA. The result is in the same range of LOCA value in other CANDU NPP [7]. In this study, the SBLOCA and LBLOCA are combined together as an overall LOCA event, because the SCWR safety system performance is similar in both scenarios [8]. The total occurrence frequency of LOCA is 1.27E-2/y.

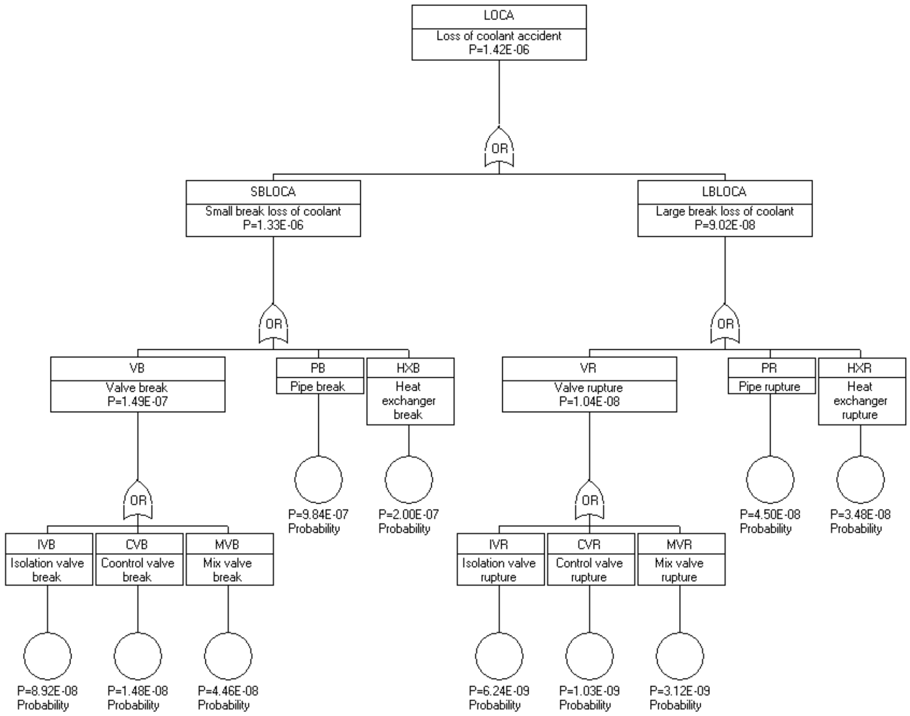


Fig. 3 Fault Tree for LOCA

TABLE II. Failure Rate for Heat Exchanger Primary Loop

Code	Description	Available Values*	Value used	Comment
IVB	Isolation valve break	NUREG: 4.46E-8/h E&R: 1.0E-8/h	8.92E-08/h	Two isolation valves in loop
CVB	Control valve break	NUREG: 1.48E-8/h E&R: 1.0E-8/h	1.48E-08/h	
MVB	Mix valve break	NUREG: 4.46E-8/h E&R: 1.0E-8/h	4.46E-08/h	
PB	Pipe break	NUREG: 6.89E-10/h-ft E&R: 3.0E-9/h-ft	9.84E-7/h	100m
HXB	Heat exchanger break	NUREG: 2.0E-7/h E&R: 1.0E-7/h	2.0E-7/h	Tube leakage
IVR	Isolation valve rupture	NUREG: 3.12E-9/h E&R: 1.0E-10/h	6.24E-9/h	Two isolation valves in loop
CVR	Control valve rupture	NUREG: 1.03E-9/h E&R: 1.0E-10/h	1.03E-9/h	
MVR	Mix valve rupture	NUREG: 3.12E-9/h E&R: 1.0E-10/h	3.12E-9/h	
PR	Pipe rupture	NUREG: 1.38E-10/h-ft E&R: 3.0E-11/h	4.5E-8/h	100m
HXR	Heat exchanger rupture	NUREG: 3.48E-08/h E&R: 1.0E-9/h	3.48E-08/h	Tube Rupture

* *NUREG* [5]*: NUREG/CR-6928 Industry-Average Performance for Components and Initiating Events at U.S. Commercial Nuclear Power Plants.*
E&R [6]*: Component external leakage and rupture frequency estimates.*

III.B.2. LOCA consequences modeling with event tree analysis (ETA)

The response of main reactor safety system in the event of LOCA is modeled with the event tree. The event tree analysis of CANDU-SCWR is based on the safety study of CANDU-SCWR described in [8], the event tree is shown in Fig. 4.

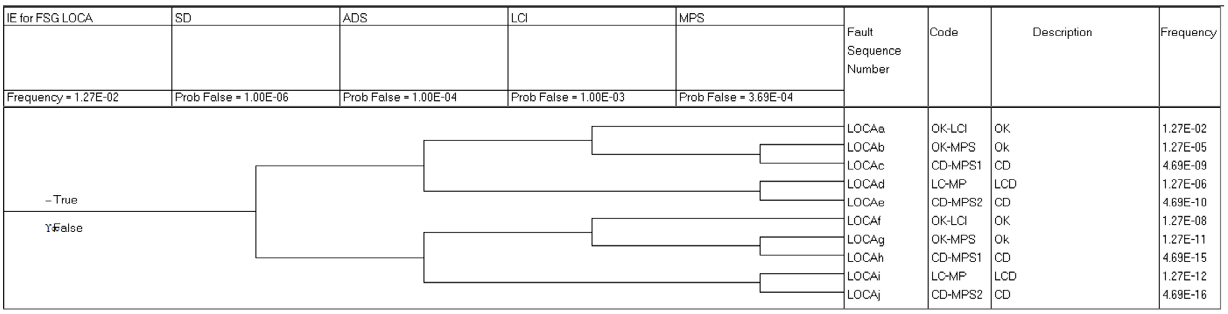


Fig. 4. Event Tree Model for LOCA

Compared with existing CANDU reactors, which have a positive Coolant Void Reactivity (CVR), the CANDU-SCWR has a negative coolant voiding which slows and eventually stops the fission process in a LOCA. Similar to other CANDU reactors, two independent shutdown systems will activate to ensure a minimal loss of inventory prior to trip. The residual heat generated from the LOCA is removed by the emergency cooling system which consists of pumped or gravity-fed Automated Depressure System (ADS) and Low Pressure Core Injection (LCI). An ADS is capable of sustaining blowdown cooling for a period of some 10s of seconds for rapid depressurization, and the LCI supplies water to the reactor core during emergency cooling conditions. If all the safety system failed, the use of a passive moderator cooling is the last line of defense to keep the core cool in the case when cooling capability is lost. The summary of LOCA outcomes is shown in Table III.

TABLE III. Summary of LOCA Outcomes

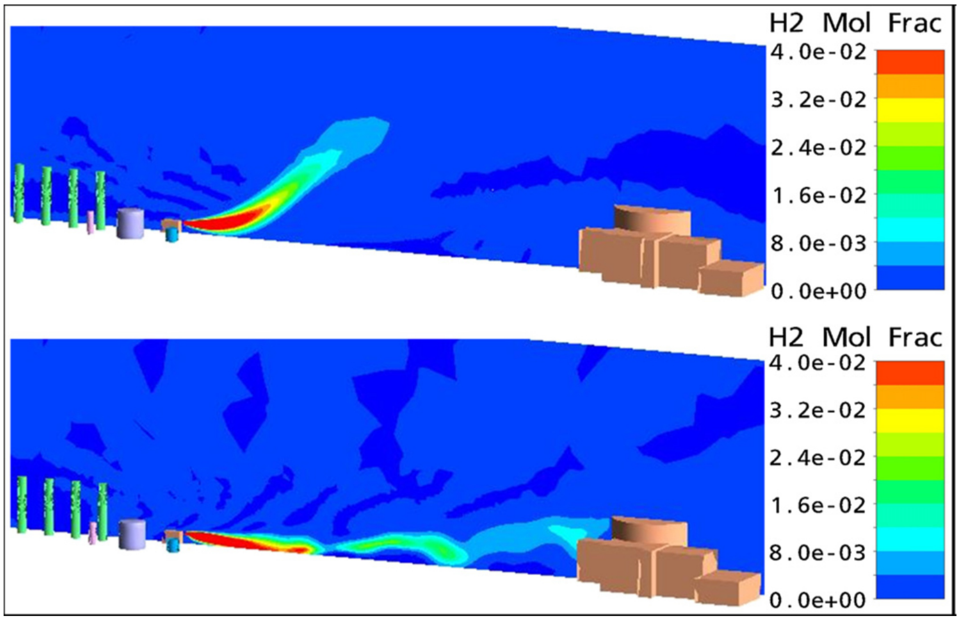
Outcome Code	Outcome Description	Outcome Frequency
CD-MPS1	CD	4.69E-09
CD-MPS2	CD	4.69E-10
LC-MP	LCD	1.27E-06
OK-LCI	OK	1.27E-02
OK-MPS	OK	1.27E-05

In Table IV, the state OK-LCI implies the reactor core successfully gets long-term cooling from the LCI after the reactor shutdown. In the situation when the LCI fails and Moderator Passive Circulation System (MPS) works, the core can still be kept cool (OK-MPS). In the situation where ADS fails and MPS works, limited core damage is caused due to the delay in core cooling from MPS (LC-MP). Core damage occurs when the MPS fails together with either the ADS or the LCI (CD-MPS1 CD-MPS2). In summary, the severe core damage probability due to the interfacing with hydrogen production is 5.1e-9/y.

The quantitative safety goal for Core damage frequency (CDF) in Canadian NPP is stated by RD-337 standard as [9]: “the sum of frequencies of all event consequences that can lead to significant core degradation is less than 10-5 per reactor year”. The LOCA PSA result is significantly lower than the safety target of the nuclear design basis failure frequency. This PSA result implies that the additional risks for nuclear safety caused by the linkage with hydrogen plant can be effectively eliminated by the nuclear safety system. Therefore, no extra safety system is required to control risks from heat interface between NPP and the hydrogen plant.

III.C. Hydrogen Accidents

The hydrogen accident such as fire and explosion is possible to cause a nuclear-safety-related accident if the blast wave of hydrogen explosion carries enough energy that destroys the safety barriers of the NPP. However, the impact of the hydrogen event on the NPP highly depends on the amount of hydrogen storage inside the generation plant and the separation distance between NPP and hydrogen plant. The hydrogen release in the Cu-Cl process has been modeled with Computational Fluid Dynamics (CFD) [10] and the flammable hydrogen cloud distribution is shown in Fig. 5. The CFD analysis draws the conclusion that at a separation distance of 100m the explosion caused by hydrogen leakage to the open atmosphere has limited effect on the NPP.



Hydrogen molar fraction after 500s of release from the high pressure pipe with 1 m/s wind (Top) and with 10 m/s wind (Bottom). The wind direction is from left to right. The red colour identifies the hydrogen cloud with a volumetric concentration equal or larger than 4%.

Fig. 5. Hydrogen Release CFD

Because the layout of the nuclear-based hydrogen plant is still under the design phase, it is difficult to estimate how much hydrogen gas would be stored in the hydrogen facility and what is the final separation distance between the two plants. However, in accordance with the inherently safer design philosophy [11], the risks in the process could be reduced or eliminated by minimizing the quantities of hazardous material. Therefore, it is assumed in this study that the future design of the hydrogen plant will significantly reduce the amount of hydrogen in the hydrogen plant to achieve a better inherent safety, by immediately deliver the hydrogen gas to the hydrogen storage facility located at a safety distance from both the hydrogen plant and the NPP. Based on this assumption, the amount of hydrogen involved in a hydrogen event is limited. Only the release from continuous hydrogen generation is analyzed.

III.C.1. Hydrogen accidents overview

Hydrogen is a flammable, colorless, tasteless and odorless gas. As a hazardous resource in the process industry, hydrogen has unique properties, such as ease of leaking, wide range of combustible mixture and low-energy ignition. The production, distribution and use of hydrogen as a primary energy source pose new safety challenges.

In the Cu-Cl cycle, hydrogen is generated with the CuCl/HCl electrolysis reaction. The main equipment used for hydrogen generation is the electrolyzer, in which the hydrogen gas is produced at the cathode. The lab scale hydrogen production with electrolyzer has been demonstrated at AECL [12]. In future large scale hydrogen production systems, the industrial electrolyzer will consist of many individual electrolysis cells. The reactant is delivered into each reaction cell evenly, and the hydrogen is generated at the cell's cathode. Hydrogen gas is collected from the cells through pipes inside the electrolyzer and delivered to storage and distribution facilities. The hydrogen storage and distribution facilities are located at a safe distance away from both the hydrogen plant and the NPP.

III.C.2. Initiating event identification for hydrogen accidents

The initiating event for a hydrogen accident is the release of hydrogen gas from the hydrogen production reactor. Hydrogen would release from any leakage of piping and equipment or from the loss of containment of the elecrtolyzer due to reactor overpressure. Fig. 6 shows the fault tree for the hydrogen release. The failure rates of basic events are taken from [5, 6, 13], as shown in Table IV. The pipe length is assumed in the range of several 10 meters, due to the fact that a hydrogen explosion will have minor effect within that range [10].

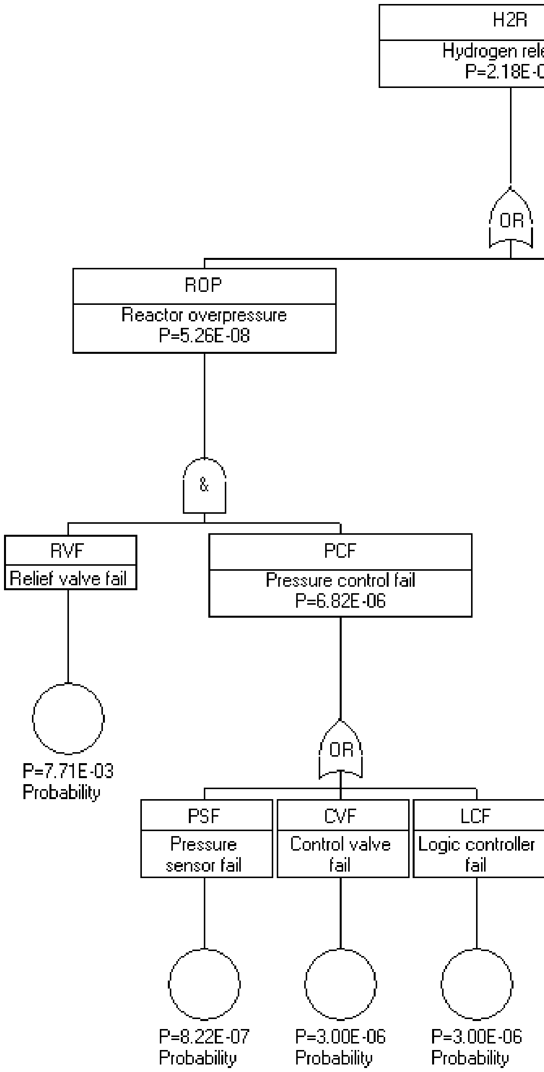


Fig. 6. Fault tree model for the hydrogen release

TABLE IV. Failure Rate for Hydrogen Generation

Code	Description	Available Values*	Value Used	Comment
PSF	Pressure sensor fail	NUREG: 8.22E-7/h	8.22E-07/h	
CVF	Control valve fail	NUREG: 3.0E-6/h	3.0E-06/h	Fail to control
LCF	Logic controller fail	WSRC: 3.0E-6/h	3.0E-06/h	
RVF	Relief valve fail	NUREG: 7.71E-3/d	7.71E-3/d	Failure per demand
PB	Pipe break	NUREG: 6.89E-10/h-ft E&R: 3.0E-9/h-ft	9.84E-7/h	100m
HRB	Hydrogen reactor break	WSRC: 1.0E-7/h	1.0E-7/h	Pressurized tank
PL	Pump leak	WSRC: 1.0E-6/h	1.0E-6/h	External leak
VL	Valve leak	NUREG: 4.46E-8/h E&R: 1.0E-8/h	4.46E-08/h	

***NUREG: NUREG/CR-6928 Industry-Average Performance for Components and Initiating Events at U.S. Commercial Nuclear Power Plants.*
E&R: Component external leakage and rupture frequency estimates.
WSRC [13]*: WSRC-TR-S3-262 Savannah river site generic data base development (u).*

The initiating event probability of hydrogen release is 2.18E-6/h, i.e., 1.9E-2/y. From FTA, the hydrogen leakage in the piping and equipment contributes most to cause a hydrogen release, while the risk of hydrogen release due to reactor overpressure is effectively mitigated by the relief valve.

III.C.3. Hydrogen events modeling with ETA

The consequence of a hydrogen leakage is modeled with the event tree as shown in Fig. 7. The event hydrogen release is selected as an initiating event for the ETA, and a frequency of $1.9e^{-2}$/y is assigned to it according to the result of the FTA. The event tree is adopted from [14]. The assessment is based on worst case scenario, where the hydrogen release rate is 3.6 kg/s.

Hydrogen is extremely flammable gas. Thus, according to the classification criteria described in [14], hydrogen is classified as category 0 material. As a category 0 gas, the direct ignition probability given a hydrogen release is 0.2 with a rate less than 10 kg/s. If a direct ignition does not happen, a flammable cloud is formed due to the continuous hydrogen release and a delayed ignition would take place. The probability of delayed ignition is $1\text{-}P_{\text{direct ignition}}$, which is 0.8 in this ETA. If the cloud does not ignite immediately, the released hydrogen will disperse to open atmosphere. The hydrogen release has no effect for safety in this scenario. On the other hand, if a delayed ignition occurs, the outcome of the event would be an explosion or a flash fire based on other conditions in the delayed ignition. The conditional probability of an explosion is 0.4 given a delayed ignition.

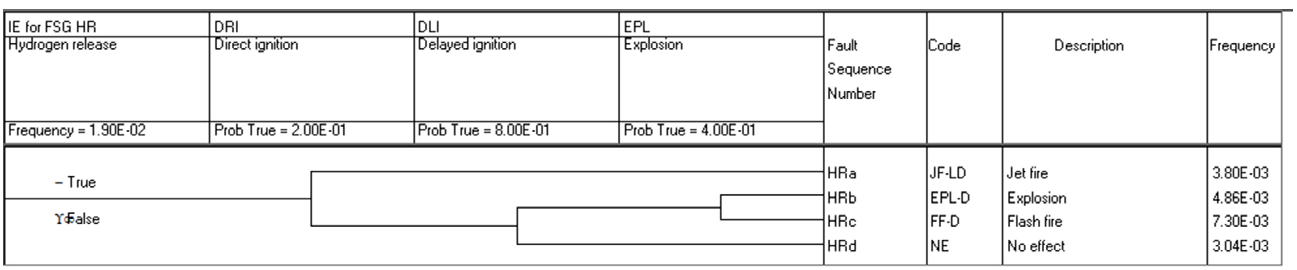


Fig. 7. Event Tree Model for Hydrogen Accident

As can be seen from Fig. 7, in the NE state, which has a probability of 3.04e-3, neither the direct ignition nor delayed ignition occurs, so the released hydrogen will disperse to the environment and the hydrogen plant is kept safe. Limited damage would be made from a jet fire (JF-LD), it is assumed the emergency response to the jet fire, such as fire extinguishing and emergency shutdown will stop the release and prevent the hydrogen plant from further harm. The worst case is the explosion (EPL-D) and flash fire (FF-D), when large amount of hydrogen ignition occurs. The outcome distribution for the hydrogen events is shown in Fig. 8.

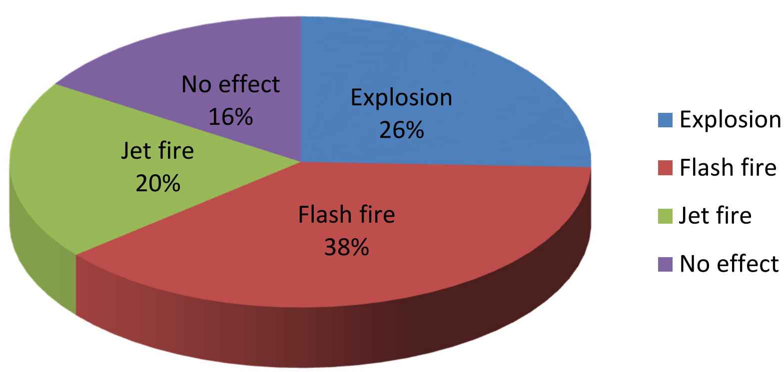


Fig. 8. Outcome Distribution of Hydrogen Release

Given a hyrogen release, there is only 16% chance to avoid an accident. In more than 60% of the hydrogen release case, a severe hydrogen accident will occur in delayed ignition. The hydrogen release is the major risk in a nuclear-based hydrogen generation.

IV. CONCLUSIONS

Nuclear-based hydrogen generation is a promising technique for large scale hydrogen production in the future. The linkage of the NPP and the hydrogen facility introduces new safety challenges for the co-generation plant. A safety study has been performed in this paper. Two major safety concerns, LOCA due to direct heat transfer loop and hydrogen event from Cu-Cl cycle, are analyzed based on PSA. The initiating event occurrence probabilities have been derived from the FTA, with a probability of $1.27E^{-2}$/y for LOCA and $1.9E^{-2}$/y for hydrogen release. Based on the FTA result, the accident sequences are modeled with ETA to derive all possible outcomes and their probabilities. The ETA shows that the nuclear safety issues due to the LOCA in the heat transfer loop can be handled by the nuclear safety systems, and therefore have minor impact on the nuclear reactor. On the other hand, the hydrogen event is the major risk in the nuclear based hydrogen generation plant, which can lead to a severe accident with a probability of $1.2E^{-2}$/ year. Therefore, it is recommended that a Safety Instrumented System (SIS) to be designed as an independent protection layer for the hydrogen plant, in order to mitigate the risks of hydrogen release.

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