

SAFETY ISSUES IN NUCLEAR HYDROGEN PRODUCTION WITH THE VERY HIGH TEMPERATURE REACTOR (VHTR)

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Abstract

Nuclear Hydrogen Production (NHP) schemes use high-temperature nuclear heat and steam to split water, thermo/electrochemically. This focuses attention on high-temperature coolants: supercritical water, gaseous helium, and liquid salts. Each has been studied within a reactor concept – the SCWR, the VHTR or the LS-VHTR respectively. Nuclear Hydrogen Production is particularly attractive for hydrocarbon upgrading in the Alberta Tar Sands, where liquid salt heat-transfer fluids have also been proposed for *in-situ* tar liquefaction. This paper considers the heat transfer characteristics of these fluids, and discusses safety issues in the context of NHP, with the gaseous helium VHTR and the liquid salt VHTR.

1. Introduction

In recent years, considerable interest has arisen in the possibility of using nuclear heat, nuclear-generated electricity and nuclear-generated steam for hydrogen production, and a significant effort is currently underway to develop the associated enabling technologies – both in Canada and internationally. Nuclear heat and steam is planned to be used for hydrogen production by water splitting with thermochemical cycles, while nuclear-generated steam and electricity will be used to produce hydrogen by high-temperature electrolysis. Nuclear Hydrogen Production, or NHP, as it has come to be called, is increasingly viewed as a strong component of the future mix of greenhouse-emission-free energy technologies. It is likely to have a role both in the short and medium term (where hydrogen will be used to upgrade and refine hydrocarbons), as well as in the long-term, when a hydrogen-fuelled transportation sector might come to exist. In either application, the technology lowers greenhouse gas emissions relative to alternatives in use today.

In Canada, a significant component of the medium-term growth in hydrogen demand (estimated to triple between now and 2020) is forecast to arise from the need for hydrogenation of (i) bitumen in the tar sands, and (ii) other lower grade hydrocarbons - for conversion into higher grade ‘sweeter’ hydrocarbons. This hydrogenation is likely to be necessary both in the pre-refining stage and during refining itself. Hydrogen will also be used (and generated) in related applications such as coal liquefaction and synthetic fuel production [1]. Currently, the hydrogen required for such processes is produced by steam reforming of hydrocarbons, mainly natural gas (other hydrocarbons are also less widely used as feedstock). Extraction of bitumen from the tar sands also requires heat – to melt it so it may flow more easily and be pumped out – this is called *in situ* tar liquefaction. This heat is currently supplied *via steam pumped directly into the wells* (in a process known as Steam Assisted Gravity Drainage, SAGD). The source of heat for making this steam is hydrocarbon combustion. Thus both the heat and the hydrogen that is required in tar sand processing are currently generated via processes that emit greenhouse gases.

Nuclear technologies currently under active development could supply both the heat and the hydrogen that is required in tar sand processing, in addition to electricity (some or all of which could

be used in hydrogen production) – without emitting greenhouse gases. (i) **Heat for Tar Liquefaction:** Nuclear-heated liquid salts (with high boiling points, high specific heats but low viscosities) could be *circulated in vertical piped loops through the tar sand wells*, for in-situ tar liquefaction [5]. (ii) **Heat for Nuclear Hydrogen Production** This could be supplied to the NHP processes (thermochemical and/or electrolytic) via a heat transfer fluid loop between the nuclear reactor and the thermo/electrochemical plant, which could be a molten salt or helium (somewhat independently of the actual coolant used in the reactor itself, which may also be a molten salt, but could be helium gas, or supercritical water). (iii) **Electricity for High Temperature Electrolysis** will be also be supplied by the nuclear reactor.

These possibilities represent potential synergies between nuclear technologies and fossil fuel technologies. They have the potential to significantly improve both the energy efficiency and the greenhouse footprint of the tar sand operations. To list the possibilities: **(I) Hydrogen Generation:** (i) Nuclear heat for thermochemical cycles, supplied via a suitable heat transfer fluid; (ii) Nuclear heat for steam generation, for use in high temperature electrolysis; (iii) Nuclear-heated steam in hydrocarbon reforming for hydrogen production (possibly as a transition technology) preventing hydrocarbon combustion and associated greenhouse gas emissions. **(II) Tar Sand Processing:** (iv) Nuclear heated thermal transfer fluids (e.g., liquid salts) for *in situ* melting of bitumen for extraction (v) Nuclear generated steam for Steam Assisted Gravity Drainage (SAGD), as an alternative, or possibly transitional technology. **(III) Other possibilities** include use of nuclear-generated process heat, steam or hydrogen for use in (vi) coal liquefaction or (vii) synthetic fuel production, and (viii) shale oil extraction. This paper will not address these last three possibilities in great detail.

It is not yet absolutely certain which mix of these technology options will ultimately be deployed. Furthermore, transitional scenarios may have a different mix from the final scenario. However, co-location of thermo-electro-chemical and nuclear plants is expected for hydrogen production. And, because this is unprecedented, a proactive safety analysis is warranted. This paper (a) reviews the nuclear technologies that have been proposed for hydrogen production; (b) examines the characteristics of proposed heat transfer fluids (needed both in nuclear hydrogen production and separately in tar sand processing); (c) scopes the safety issues that arise in these applications; (d) presents results from a quantitative risk analysis; and (e) outlines possible design modifications that could mitigate the risks.

From a broad viewpoint, this paper contributes to the analysis of technology synergies in three major Canadian energy sectors – Nuclear, Hydrogen and Fossil Fuels. The analysis to be presented is especially relevant to tar sands development, and particularly to the theme of the 2008 CNS Annual Conference – ‘Sustainable Development through Nuclear Technologies’.

This paper is organized as follows. In **Section 2**, concepts of (i) nuclear thermochemical hydrolysis and (ii) nuclear electrolysis, for hydrogen production are presented, and the need for efficient and safe heat transfer fluids and coolants in this context is elucidated. In **Section 3**, reactor concepts, and associated coolants and heat transfer fluids that have been proposed in this context are discussed and compared. In **Section 4**, Safety Issues in Nuclear Hydrogen Production are identified, while in **Section 5**, risk-mitigating design modifications are discussed. **Section 6** is the Conclusion.

2. Nuclear Hydrogen Production

Hydrogen is the most abundant element in the universe, but reacts easily with all other elements, most commonly forming hydrides, hydrates, hydrocarbons and carbohydrates. Currently, hydrogen is commonly produced by separating it from hydrocarbons and hydrides – but these processes involve the emission of greenhouse gases, primarily carbon dioxide, coming both from the heat source and from the feedstock. Much effort is therefore currently being devoted to developing processes for hydrogen production that have a small (or zero) greenhouse footprint. This effort serves both the current spectrum of demand for hydrogen, as well as the demand from the transportation sector that is anticipated in the future.

The effort to develop greenhouse-emission free hydrogen production technologies has mainly focused on splitting water. On a global level, the impact is expected to be neutral: hydrogen is extracted from water, costing energy and releasing oxygen; then, the hydrogen is combusted, releasing energy, using up proportionate oxygen and re-creating water, which is again used for hydrogen production. A variety of energy sources are being considered for such hydrolysis – including biological energy (from algae), solar energy, wind energy, and geothermal energy. The main advantage of nuclear energy, however, is the fact that the high temperatures needed for hydrolysis can be reached easily in nuclear reactors. The maturity and versatility of nuclear technology (e.g., hydrolysis can be done alongside steam generation, electricity production, or water desalination) also increases its attractiveness, and potentially, also improves its economics.

2.1 Thermochemical Cycles

Thermochemical processes split water by trapping the hydrogen from water in a compound from which hydrogen can be easily dissociated thermally (such as Hydrogen Iodide (HI)). Several dozen cycles have been studied; two are the focus of great effort at this time: the Sulphur Iodine (S-I) cycle which employs sulphuric acid and hydrogen iodide as recycling ‘reagents’, see Figure 1; and the Copper-Chlorine (Cu-Cl) cycle which employs cuprous, cupric, and copper oxy-chloride(s) similarly; trapping the aqueous hydrogen in hydrochloric acid; see Figure 2.

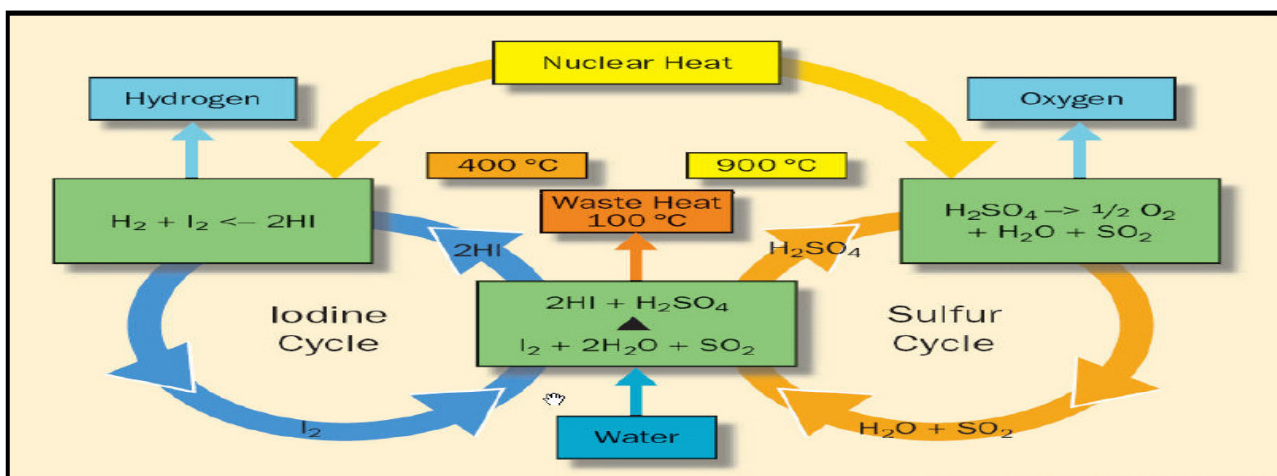


Figure 1: Concept of the S-I cycle. Heat required is drawn from a Very High Temperature Reactor (VHTR) or Advanced High Temperature Reactor (AHTR). The Supercritical Water-cooled Reactor (SCWR) outlet temperature is below the temperatures required.

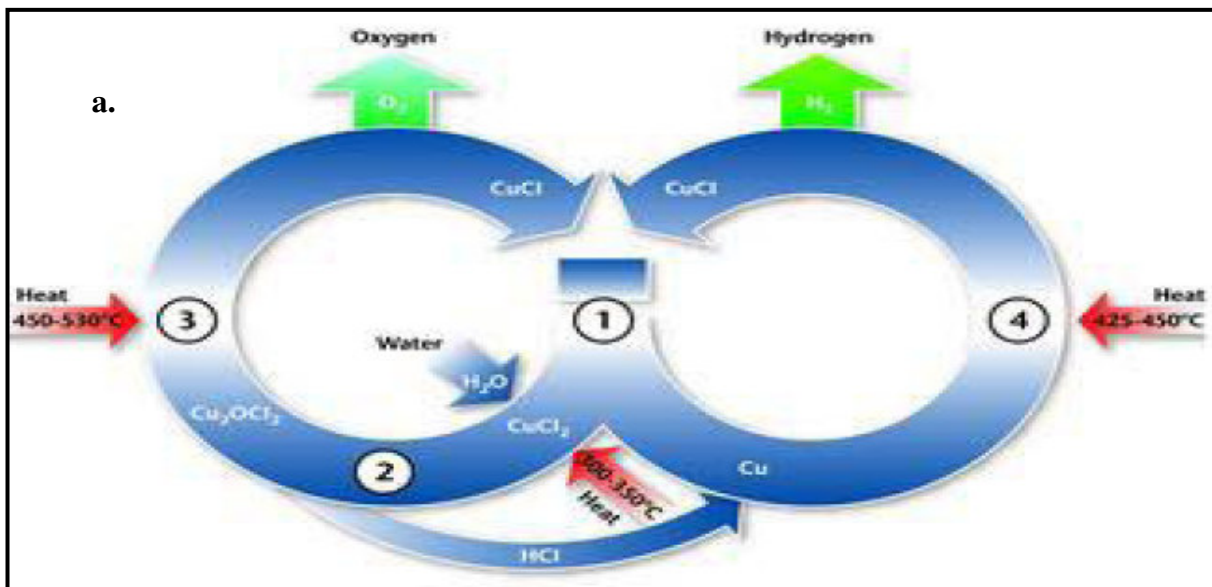


Figure 2: Conceptual Picture of the Copper Chlorine Cycle (adapted from Rosen 2006). The heat that is required in the three steps can be obtained from the CANDU-SCWR, whose concept envisages an outlet temperature of 650 C; the VHTR or AHTR can also supply the heat required.

2.2 High Temperature Electrolysis

The High Temperature Electrolysis (HTE) concept involves the use of nuclear generated high temperature steam at temperatures greater than 900 C, feeding into a special solid oxide fuel cell (SOFC) with porous electrodes and a gas-tight electrolyte, being run in reverse mode.

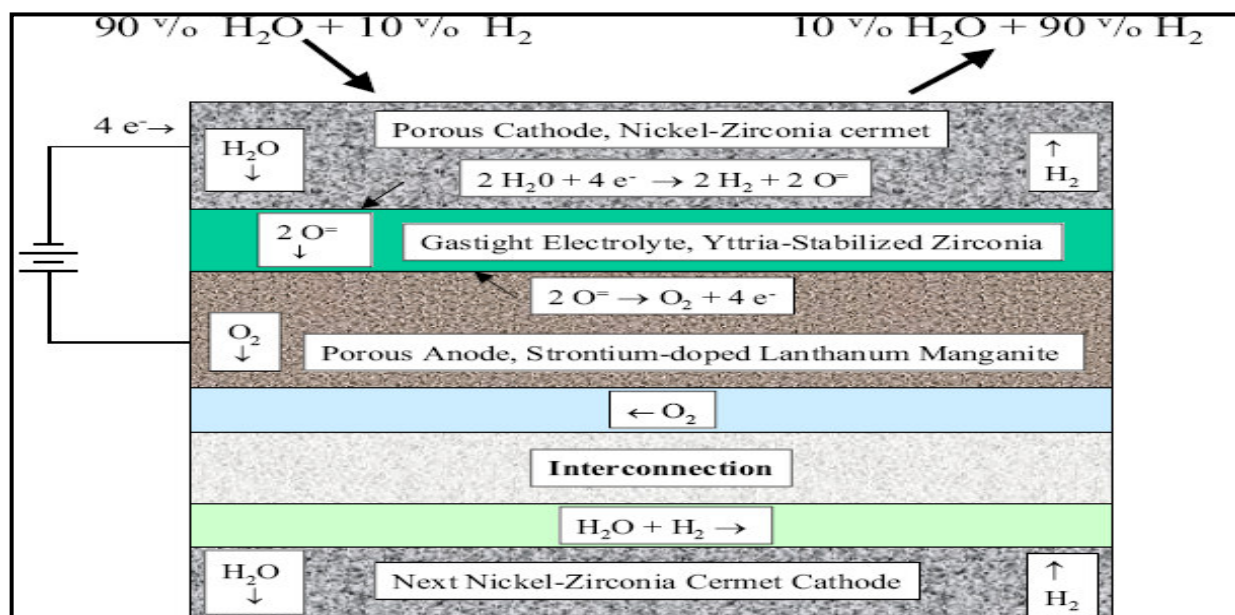


Figure 3: Conceptual Schematic of a High Temperature Electrolytic Fuel Cell. The input is steam with a 10% mixture of hydrogen at a temperature around 900 C, while the output is 90% hydrogen. Many such cells will be run at any future production facility. Other fuel cell concepts for high-temperature electrolysis are also being considered.

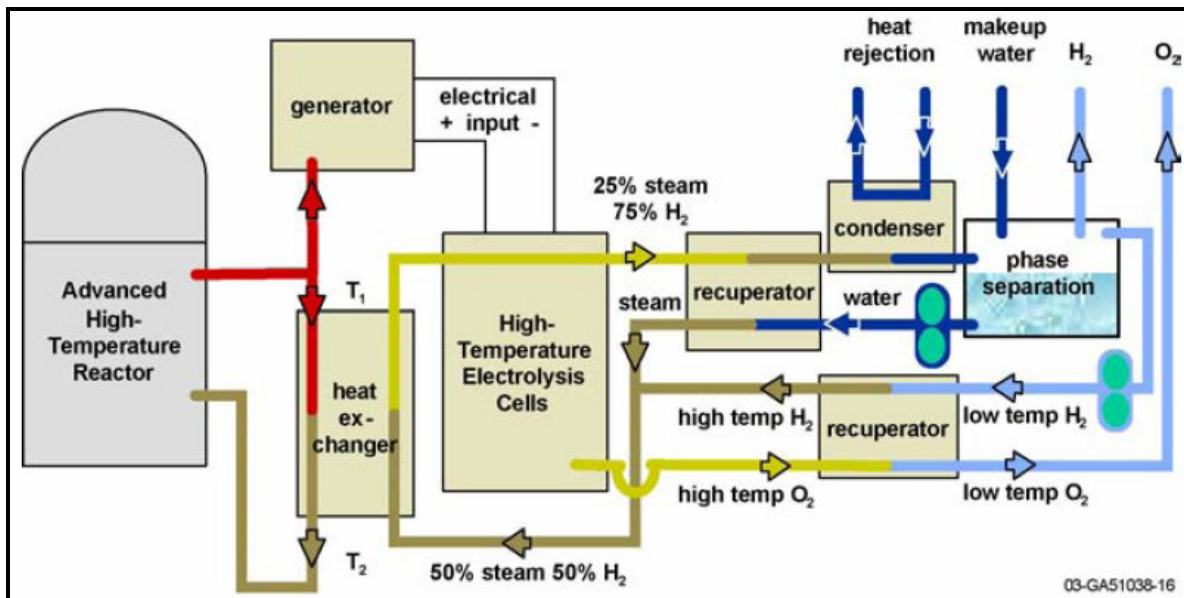


Figure 4: Conceptual Schematic of the High Temperature Electrolysis (HTE) scheme for Nuclear Hydrogen Production. The reactor provides high temperature steam, and the electrolysis cells use nuclear generated electricity as well. While an AHTR is indicated, a VHTR could also be used.

2.3 Need for High Temperature Heat

The need for high-temperature steam (temperatures > 900 C) is clear in both the thermochemical and electrolytic schemes for splitting water. Although the schemes will work at lower temperatures, the efficiency of both processes exceeds 50% only at a temperature of around 900 C (see Figure 5). The dependence of efficiency with temperature is particularly strong for the S-I process.

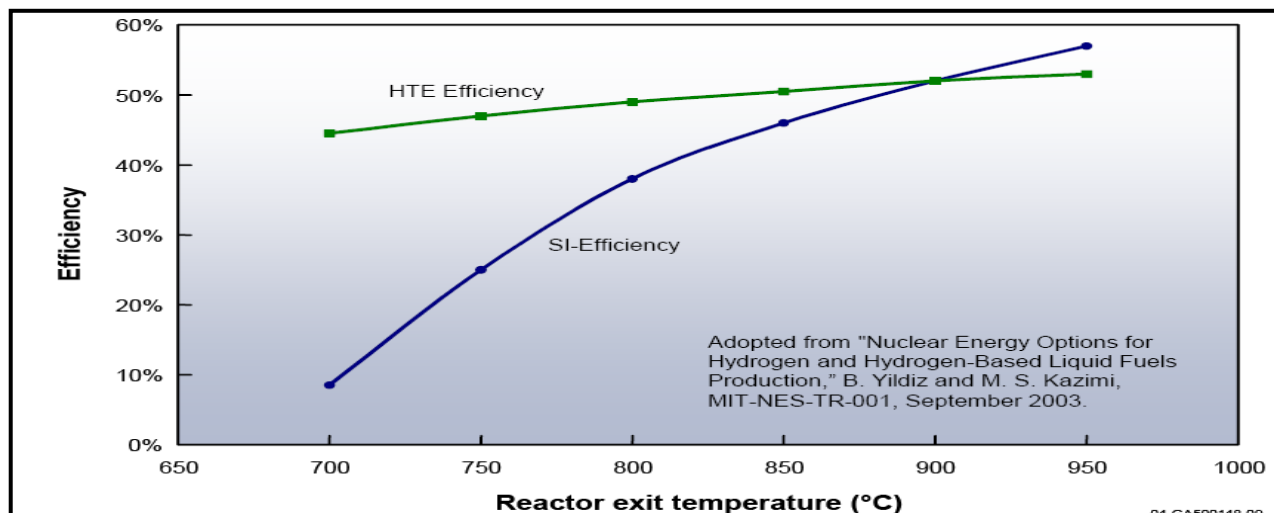


Figure 5: Dependence of Hydrogen Production efficiency on Reactor outlet temperature. Heat at the temperatures required will likely be supplied from a Very High Temperature Reactor (VHTR) or an Advanced High Temperature Reactor (AHTR).

3. Reactor Types, Coolants and Heat Transfer Fluids

The high temperatures to which steam must be heated in order for hydrogen production to occur at acceptable efficiency cannot be achieved in PWRs, PHWRs, or BWRs, which have a maximum achievable temperature of about 300 C. This upper limit in temperature is an inherent design constraint, and also limits the overall thermal efficiency of these reactors.

Therefore, alternative designs which allow the coolant to be heated to higher temperatures must be investigated, both to increase the reactor thermal efficiency and to allow the hydrogen production to occur at acceptable efficiency. The major complicating factor in low-boiling liquid coolants such as water is the existence of phase changes and the consequent dual phase flow which can occur within the operating range with serious safety implications. Gas coolants are free of this constraint, and may be safely heated to very high temperatures. Chemical reactivity of the gas is a serious issue; thus helium, an inert gas, is preferred. This leads to the basic variant of the **VHTR** (Very High Temperature Reactor) a helium cooled concept. Alternatively, water cooled reactors may be operated in the supercritical water regime (though this requires increases in both operating temperatures and pressures) leading to the Supercritical Water-cooled Reactor concept, **SCWR** [2]. A third option that has more recently acquired credibility is to use high boiling liquid salts. Flinak (which is a LiF-KF-NaF mixture in the ratio 46.5-11.5-42) melts (or freezes) at 454 C and boils at 1570 C. Thus it is liquid at normal pressure in the range 454 C – 1570 C. Its specific heat-density product, a measure of heat carrying capacity, is about the same as that of water, 1 cal/g. A reactor concept that has been developed with this coolant has been given the acronym '**AHTR**' for Advanced High Temperature Reactor, also called **LS-VHTR**.

The CANDU-SCWR concept has been scoped at an outlet temperature of 650 C, while the RPV-SCWR concept is scoped at an outlet temperature of about 500 C [2]. The CANDU-SCWR can thus provide steam at the temperatures required for the Cu-Cl cycle, while either the gas-cooled VHTR or the liquid salt cooled LS-VHTR, can supply heat at the temperatures required for both the S-I cycle and the Cu-Cl cycle. The basic heat transfer loops in a future nuclear hydrogen production facility would look as shown in Figure 6.

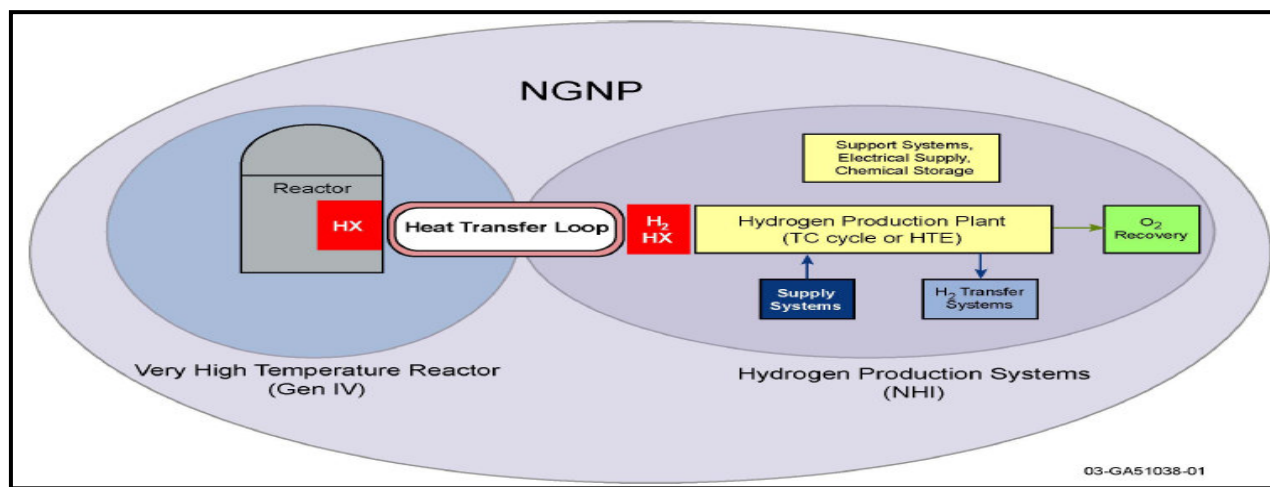


Figure 6: Conceptual schematic of nuclear hydrogen facility clearly showing the heat exchangers and heat transfer loops (Sherman 2006).

3.1 Heat Transfer Fluids

Since heat must be transferred from the nuclear reactor to the thermochemical or electrochemical facility that produces the hydrogen, heat transfer fluids must be considered. Heat transfer fluids must have high heat capacity, low viscosity, undergo no phase change in the temperature range of interest, and be chemically inert. Safety considerations indicate that a minimum distance be maintained between the nuclear reactor and the thermo-electro-chemical plant, to prevent an event, such as a spill, explosion, or fire in the chemical plant impacting the nuclear reactor. Thermal fluids should be able to flow a considerable distance at low pumping power. Ideally, there should be no possibility for any adverse interaction between the thermal fluid and the neutron radiative flux, or piping and heat exchanger materials

Gases like helium are automatic candidates for heat transfer fluids because of their chemical inertness. However, liquid salts have low viscosity and can flow with minimal pumping power. They also have low density and high specific heats which further commend them. Using helium as either a coolant or a heat transfer fluid also raises the possibility of tritium generation through neutron activation of He-3, but Li in Flinak can also undergo activation to release tritium.

Heat transfer fluids are also needed in the *in situ* liquefaction of tar sands. Currently, steam serves as a heat transfer fluid in the SAGD (Steam Assisted Gravity Drainage) process. However, the attractive properties of liquid salts make them worth considering for this purpose. Forsberg (2007) has proposed the schematic shown in Figure 7. Liquid salts heated in a nuclear reactor are circulated through vertical subsurface heat transfer loops and then pumped back to the surface. The liquefied bitumen/tar can then be pumped up from separate wells. The great advantage of the process over SAGD is that liquid salts can flow down by gravity, and do not need to be pumped in as steam is currently.

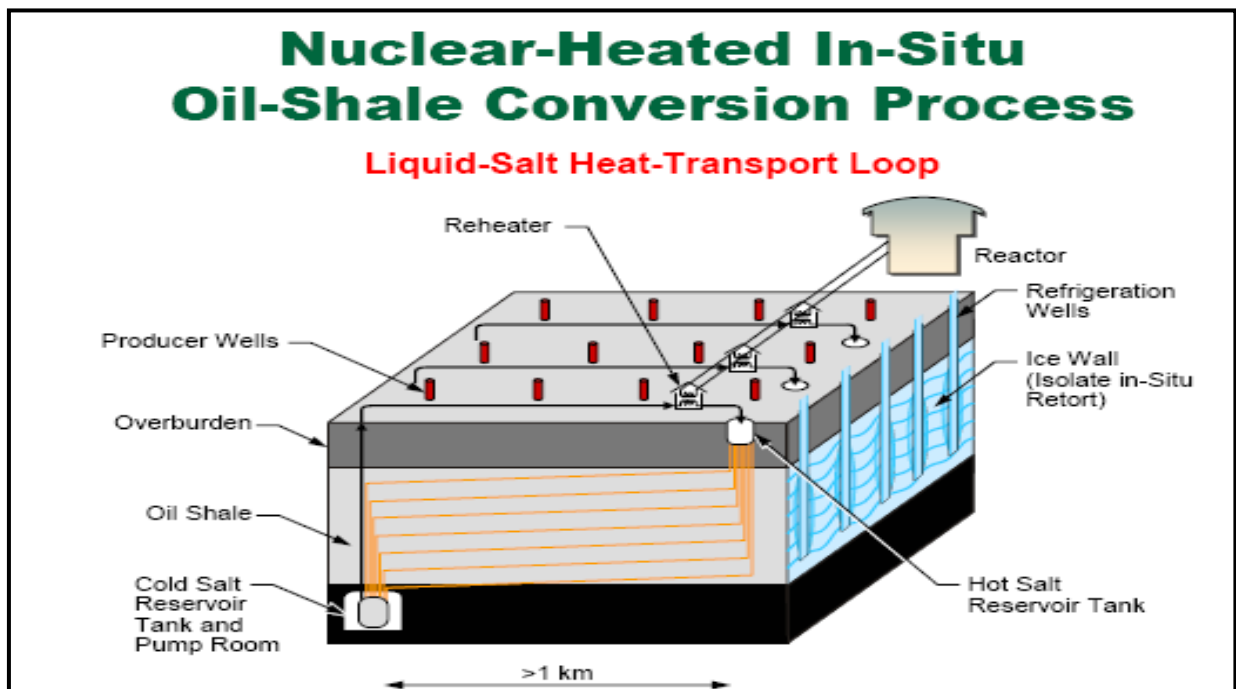


Figure 7: Conceptual Schematic of an *in situ* liquefaction process using liquid salts as heat transfer fluids (Forsberg 2007). Similar technologies may have applications in the Alberta sands.

	Helium	S-CO ₂	Flinak (LiF-NaF-KF)
Formula Wt (g/mol)	4.003	44.01	41.2
Melting Point (°C)	-	-	454
Density (kg/m ³)	2.859	31.06	1528
Heat Capacity (kJ/kgmol-°C)	20.78	1.285	88.35
Viscosity (Pa s)	2.703e-3	5.161e-5	1.963e-4
Thermal Conductivity (W/m-°C)	0.2517	0.07885	0.8612
Phase	Gas	Gas	Liquid

Table 1: Properties of three alternative heat transfer fluids. Flinak is seen to be less viscous, more conductive, and denser, with greater heat capacity than Helium. S-CO₂ is supercritical carbon-dioxide, some of whose *heat transport* properties resemble those of supercritical water.

4. Safety Issues in Nuclear Hydrogen Production (NHP)

There are two main conceptually separable parts of the nuclear hydrogen production facility – the nuclear reactor itself (which supplies heat, steam and electricity), and the thermo-electrochemical plant, which produces the hydrogen. Safety issues arise in both contexts separately, and have been the focus of considerable analytical effort. Traditional nuclear safety analyses have considered reactor core safety and safety of the thermal hydraulic loop, separately and together. Chemical safety analyses have considered the probability of fires, leaks, spills and explosions in chemical plants intended for a wide variety of uses.

While the SCWR is a truly new concept that has not been previously constructed, *variants* of both the helium-cooled VHTR and the liquid salt cooled VHTR that have been constructed in the past. A body of operating and safety knowledge about them thus exists. Further analyses would undoubtedly be required as the current variants move toward deployment. In the case of the helium-cooled VHTR, both thorium and uranium fuel cycle power reactors were built in the past, but all of them were subsequently shut down due to poor operating performance. Both Japan and China are currently operating helium-cooled high-temperature test reactors (HTTRs) and both are also building full-fledged power reactors of this type. Japan has also operated a test hydrogen production facility drawing heat from a 30 MW helium-cooled test reactor, and is well advanced on the path to running it on a commercial scale.

Though co-location of hydrogen production facilities with nuclear power plants is unprecedented, hydrogen is already used in BWRs and PWRs for controlling the pH of the coolant light water; thus hydrogen and nuclear technologies are no strangers. Hydrogen is stored on site at LWR locations, and is pumped directly into the LWR coolant loops as required. The primary safety concern from hydrogen is the risk of fires and explosions. An event database exists from which probabilities may be estimated. A number of safety codes exist which govern the quantity, location, and proximity that hydrogen may be stored in near a nuclear reactor, motivated by the need to prevent damage to the nuclear reactor core from such hydrogen

explosions and fires. The analysis that underlies the development of such safety codes and standards will largely continue to apply to future nuclear hydrogen production facilities. Some of these codes are valid regardless of where the produced hydrogen is used – since hydrogen is a flammable and volatile gas, there is a body of safety codes that govern its storage and use in *any* context.

However, qualitatively new risks in nuclear hydrogen production (NHP) will arise from the following sources.

- (i) Proximate location of a Chemical Plant to a Nuclear Plant. The plant might have not only hydrogen but also toxic chemical species, possibly under high temperature and pressure, such as Sulphur dioxide for the S-I process and Hydrochloric Acid for the Cu-Cl process.
 - a. These will tend to corrode their containment systems, and their leakage may also have toxic effects. This introduces a new risk into the entire equation. Chemical plant explosions do occur, and in some ways, chemical plants need to be more closely supervised than nuclear plants. This incremental risk needs to be analysed thoroughly.
 - b. Hydrogen itself is non-toxic but extremely flammable, which is the source of most of the risk from hydrogen. While a full scope safety analysis of the NHP-complex is needed, a safety analysis of smaller scope that deals with the incremental risk created by generating and storing hydrogen on-site – based on the historical record of hydrogen explosion and deflagration events in LWR locations can be carried out more easily, and results of such a study have been published. (Smith *et al* 2005)
- (ii) Local production of Hydrogen in large quantities (as opposed to merely storage). This may require that codes established for hydrogen storage on site be re-visited for possible modification. Piping requirements for the hydrogen that is produced may be needed. Some of the hydrogen that is produced may come in the form of tritium, and this is more especially the case for High Temperature Electrolysis, which has a significant tritium yield
- (iii) Issues arising from Heat Transfer Fluids. The effect of having new heat transfer loops is akin to introducing an additional thermo-hydraulic (or thermo-gasdynamics) loop into the combined plant. In fact, even in the simplest conception, there will actually be at least two additional thermo-hydraulic loops. This increases the plant complexity significantly, and LOCAs or Loss of Thermal Fluid Accidents must be analysed.
 - a. Helium – if used as a coolant and heat transfer medium, could lead to tritium contamination in the hydrogen product. This will happen by activation of the He-3 isotope, which will absorb a thermal neutron and emit a proton, and H-3, tritium.
 - b. While Helium is chemically inert, it can embrittle metals and alloys by diffusing into grain boundary locations. The effect this has on overall plant systems structural stability is a significant issue.
 - c. Flinak – if used as a coolant, the lithium component of Flinak has a non-negligible cross section for converting to tritium on neutron activation. (Ohashi 2007). In addition, Flinak and related salts have corrosive properties which must be balanced against their usefulness as heat transfer fluids.

5. Risk-Mitigating Design Modifications

While a broad scoping analysis uncovers a number of safety issues, a smaller scope study examining a number of risk mitigating design modifications was carried out by a Probabilistic Safety Analysis by the Idaho National Laboratory in the US. The results are summarized in Table 2.

Case	Description	Mean Core Damage Frequency (per year)	Mean Chemical Dispersion Frequency (per year)
Nominal	No mitigating features and the separation is set at 60m	7.00E-06	3.00E-03
1	Varying the Distance between Nuclear and Hydrogen Production Facilities	See Note below	3.00E-03
2	Constructing an earthen barrier between the nuclear and chemical facilities	4.00E-10	3.00E-03
3	Constructing the Nuclear Facility primarily underground	4.00E-10	3.00E-03
4	Constructing Blast Panels near the Chemical Facility to dampen overpressure events	4.00E-10	3.00E-03
5	Constructing the Chemical Facility primarily underground	8.00E-11	8.00E-04
6	Moving the Nuclear Plant Control Room offsite	7.00E-06	Releases no longer impact control room

Table 2: Impact of Specific Risk Mitigating Design Modifications on Nominal Case (adapted from Smith et al 2006). **Note:** The direct core damage risk decreases monotonically with distance to about 80m, but flattens out before declining further. Beyond 80 m, the indirect core damage occurring from hydrogen explosions which first damage other parts of the nuclear plant becomes a significant component of total risk. Both risks decline significantly beyond 100m.

Engineering safety analyses carried out by INL (Smith *et al* 2005) have concluded that separation distances between the chemical plant and the nuclear reactor in the range of 60 m to 120 m should be adequate to reduce core damage risk from hydrogen overpressure events leading to detonation – to below 1E-9 (Smith *et al.*, 2005). This also has notable implications for the heat transfer fluid that is ultimately employed to transfer heat between the nuclear reactor and the chemical plant. If a liquid salt heat transfer fluid like Flinak is employed, it will be able to flow at low pumping pressure, thus consuming less energy, while high pressures will be required if helium is used. Use of helium also

raises the probability of tritium contamination of product hydrogen. Risk arising from the new core designs or thermalhydraulic loops in the nuclear plant itself were not analysed, but addressing those risks is a major component of the Generation IV Program anyway and so will be separately addressed. A combined PSA will then need to be carried out for the joint risk probability in the nuclear-thermochemical complex as a whole.

Other recommendations from the INL study include

- A 100 kg on-site limit for hydrogen storage;
- Double-walled pipes for hydrogen transport, for piping hydrogen away as soon as it is produced;
- Location of the nuclear plant control room outside the dispersion zone for chemical release. This design modification does not change the core-damage risk, but it reduces the possibility of exposing control room personnel to toxic gases to negligible levels. Compare the Nominal Case with Case 6 in Table 2 – the core damage risk remains $7\text{E-}6$ in both cases but the annual chemical dispersion frequency impacting personnel falls from $3\text{E-}3$ to zero.

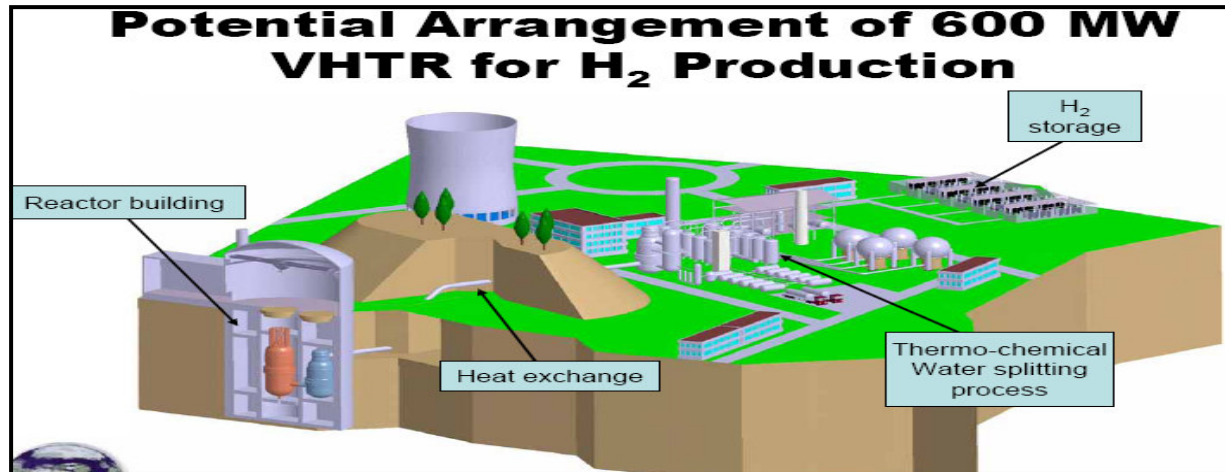


Figure 8: Artist's conception of the potential layout of a future hydrogen production facility using nuclear heat from a VHTR, incorporating most of the risk mitigation measures outlined – an earthen barrier, a separation distance, and an underground core. (Adapted from Verfondern 2006).

These recommendations were made in light of the historical record of using hydrogen in and around LWR facilities, from which an event tree and an associated fault tree were developed for a probabilistic safety analysis (PSA) to be carried out. Those risks which *directly* emanate from the chemical plant itself are not considered in this analysis, except to note that locating the control room outside the dispersal zone of chemicals from the chemical plant will reduce the risk of toxic effects to control room personnel to negligible levels. This kind of safety code is already in use in certain chemical contexts, and if nuclear-thermochemical cycles of the kind discussed are eventually commercialized, it may well become part of the culture of nuclear plants as well.

6. Conclusion

This paper has examined and reviewed several technology options for Nuclear Hydrogen Production, and compared and contrasted their characteristics and associated reactor options. It has scoped out the safety issues that are likely to be important in this context, and considered design modifications and safety codes that could mitigate the risk. It has explained some of the implications for three of Canada's major energy industries – Fossil Fuel, Hydrogen and Nuclear, and identified emerging synergies between them. It has provided quantitative estimates of the risk from hydrogen storage and chemical dispersion events, and summarized the design modifications for mitigating the special class of safety risks that arise. The types of further analyses that will be needed have also been identified.

This paper is a contribution to forward-looking technology foresighting; safety analysis; R&D issue identification; and analysis of nuclear technology options for reducing greenhouse gas emissions in the fossil fuel industry. The paper will meet the author's goals if it succeeds in communicating the excitement of building the full-scope synergies between nuclear, hydrogen and fossil fuel technologies, while also signalling the need for a careful safety analysis.

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