5 THE COMBINED NUCLEAR/CHEMICAL COMPLEX

5.1 INTRODUCTION

Recently there has been an exceptional resurgence of interest in the nuclear power industry and the cogeneration of hydrogen from the nuclear process heat. Implementing a nuclear power industry with the cogeneration of hydrogen is considered the first phase of establishing a renewable and sustainable energy industry that is capable of supplying in the energy requirements of a growing population and economy. The benefits of the cogeneration of hydrogen are not restricted to the abovementioned factors and improving the economical feasibility of the nuclear power industry, but include the additional benefits of supplying hydrogen to the so-called hydrogen economy (as discussed in Chapter 1).

Even though the future of the nuclear power industry with the cogeneration of hydrogen appears to be bright, several barriers exist which impede the implementation of the technology. These barriers include (Golay, 1995; McDowall & Eames, 2006):

- 1. Uranium resource limitations
- 2. Economic feasibility and competitiveness
- 3. Technological barriers
- 4. Safety concerns.
- 5. Absence of applicable codes and standards
- 6. Public acceptance
- 7. Licensing

All these issues (and probably many more) need to be resolved before the technology could be implemented, with the possible exception of the uranium resource limitation which is a potential long-term barrier only if the expected increase in the nuclear (fission) power industry is realized. Even if all these issues are resolved, additional aspects to consider are those applicable to licensing, quality assurance certification and safety demonstration tests (under normal and transient conditions) required before the technology may be industrially implemented. Furthermore, since such a commercial nuclear/chemical complex does not exist, there is no operational experience or expertise in connecting these facilities (Ogawa & Nishihara, 2004; Nelson *et al.*, 2007).

It is interesting to note that almost all these issues, barriers and drivers share the common objective of safety; safety regarding the public, operating personnel, environment, equipment and production facilities. Compulsory safety requirements, specifications and governmental regulations are additional aspects that may influence the other paramount objective of economic feasibility and competitiveness. An important feature of the nuclear/chemical complex, which concerns both the economic feasibility and safety of the technology, is the distance required between the nuclear power plant (NPP) and hydrogen production facility. This is due to exceptionally high expenses associated with the transport of heat to the hydrogen facility with as little heat loss as possible, as well as the safe isolation of the facilities from each other (Yildiz & Kazimi, 2006).

The purpose of this section is to perform an extensive literature survey in order to identify all possible hazards and safety aspects associated with the nuclear/chemical complex that is responsible for the production of hydrogen by utilizing the process heat generated by an adjacent HTGR nuclear power plant. This includes investigating the following:

- The requirements of such a complex
- Interfacial equipment considerations
- Safety aspects of the combined nuclear/chemical complex
- Hazard identification

It is important to note that a complete evaluation of the processes and equipment involved with the production of hydrogen from nuclear energy is not required. Furthermore, the project does not entail any designing or simulation of the processes involved. This study is fundamentally a safety study that aims to investigate the safety aspects of the production of hydrogen from the process heat supplied by an adjacent nuclear power plant, and to identify the risks and hazards associated with combining the two critical facilities.

5.2 OVERVIEW OF NUCLEAR-HYDROGEN R&D PROJECTS

The purpose of this subsection is to investigate the nuclear-hydrogen projects currently being researched and developed in order to assess the safety and technological requirements associated with such a nuclear/chemical complex.

5.2.1 SOUTH AFRICA

PBMR is investigating a hydrogen cogeneration plant that utilizes the process heat supplied by a 500 MW_t PBMR to produce hydrogen and electricity by means of the hybrid sulphur process and a Rankine plant as is shown in Figure 5-1 below. In the near term, the primary focus is on electricity generation since the country suffered severe power shortages over the last couple of years. However, the focus may shift towards hydrogen production if the electricity situation improves. The cogeneration plant utilizes an intermediate heat exchanger to transfer heat to the secondary heat transfer loop that supplies heat to the hydrogen production plant (PCHX or DR) and Rankine plant (SG). Additionally, excess heat from the hydrogen plant is transferred to the electricity-generating loop. Figure 5-1 shows the diagram for water-splitting application with the PBMR (Greyvenstein *et al.*, 2008).

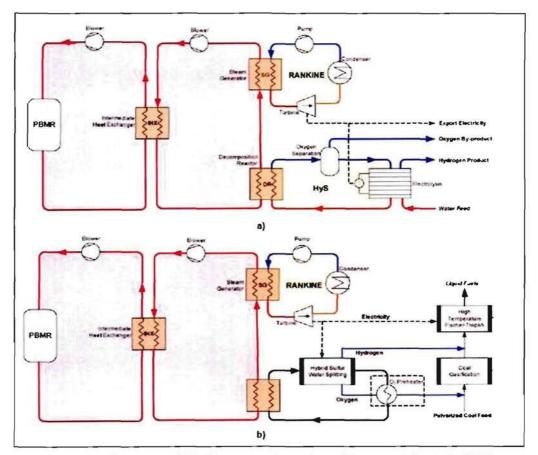


Figure 5-1: Concepts of PBMR process heat plants (Greyvenstein et al., 2008)

5.2.2 FRANCE

AREVA-NP is investigating ANTARES, which is an indirect-cycle power conversion system that can be adapted to different cogeneration schemes. The combined cycle electricity-dedicated design considers a 600 MW_t VHTR with block-type core that supplies heat to the gas and steam cycles through an intermediate heat exchanger and steam generator (Figure 5-2). Figure 5-3 shows another version of ANTARES, which is a dedicated hydrogen cogeneration plant by either the sulphur-iodine (I-S) cycle or high-temperature electrolysis (HTE; Verfondern, 2007).

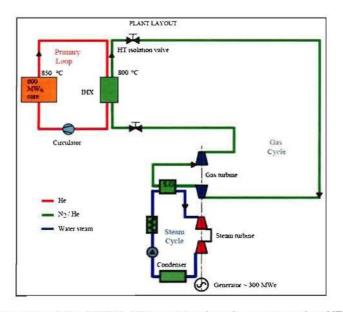


Figure 5-2: Principle of the AREVA-NP combined cycle cogeneration HTGR (Copsey, 2005 as illustrated in Verfondern, 2007)

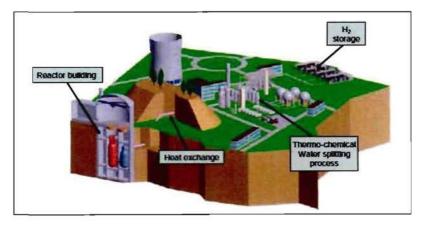


Figure 5-3: Potential arrangement of a dedicated 600 MWt VHTR for H₂ production at a rate of 1 kmol/s (Anzieu, 2005 as illustrated in Verfondern, 2007)

5.2.3 JAPAN

JAERI (now JAEA) developed a nuclear steam reforming system to produce hydrogen by utilizing the process heat supplied by a 30 MW₁ prismatic block-type HTTR. This design uses an intermediate heat exchanger to transfer heat to the steam reformer as is shown in the following figure (Figure 5-4; Verfondern, 2007). However, the focus has recently shifted towards the GTHTR300C cogeneration plant utilizing a block-type 600MW₁ HTGR. A part of the heat (168 MW₁) supplied by the reactor is used to produce hydrogen through the I-S cycle and the remainder utilized for electricity generation. The GTHTR300C cogeneration plant is illustrated in Figure 5-5 and utilizes an IHX for heat transfer (Verfondern, 2007).

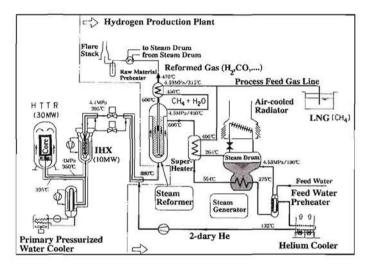


Figure 5-4: HTTR/SMR plant (Verfondern, 2007)

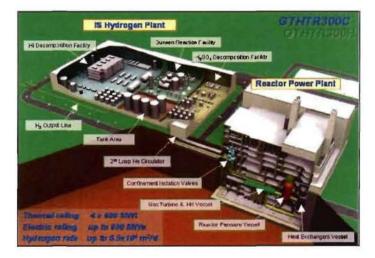


Figure 5-5: GTHTR300C plant (Yang, 2005 as illustrated in Verfondern, 2007)

5.2.4 KOREA

In 2004, the Korean government started the Nuclear Hydrogen Development and Demonstration (NHDD) project. The nuclear power plant is intended to be a VHTR with either a 600 MW₁ block-type core or a 400 MW₁ pebble bed core. The hydrogendedicated plant utilizes both HTE and the I-S cycle to produce hydrogen, while the Methane-Methanol-Iodomethane thermochemical cycle (MMI) is also under consideration. Figures 5-6 and 5-7 illustrate the Korean design (Verfondern, 2007).

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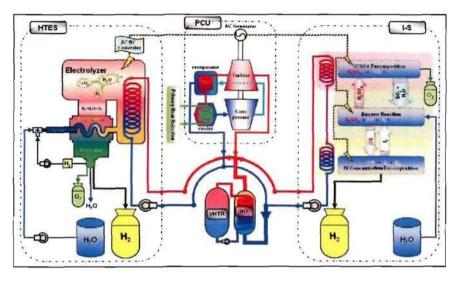


Figure 5-6: Korean NHDD plant (Lee, 2005 as illustrated in Verfondern, 2007)

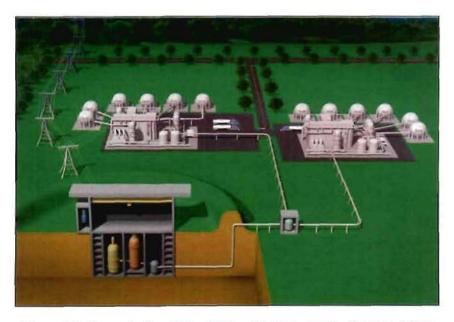


Figure 5-7: Korea design (Shin, 2005 as illustrated in Verfondern, 2007)

5.2.5 USA

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The United States are currently designing a "Next Generation Nuclear Plant" (NGNP) to generate electricity and produce process heat for hydrogen production via the I-S cycle or alternatively HTE. However, the hybrid sulphur process also receives significant R&D from industry and the government. Several nuclear plants are being considered, including the modular helium reactor (MHR or H2-MHR), the molten salt-cooled AHTR and the STAR-H2 reactor, which is a heavy metal-cooled, mixed U-TRU-nitride fuelled fast reactor. Since molten salt-cooled and liquid metal-cooled reactors fall outside the scope of this investigation, consider the H2-MHR, which is based on the GT-MHR and has a power output of 600 MWt (Schultz *et al.*, 2003). Figures 5-8 and 5-9 illustrate the concept of the H2-MHR (Verfondern, 2007).

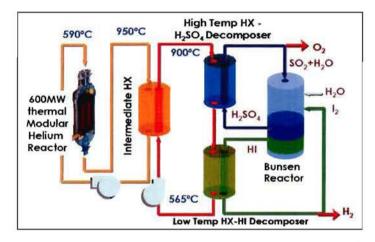


Figure 5-8: Concept of the US H2-MHR combined with the I-S cycle (Verfondern, 2007)

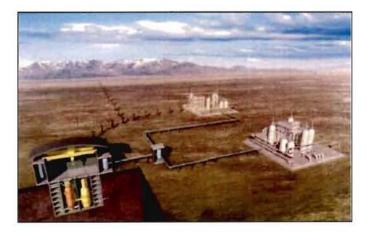


Figure 5-9: H2-MHR combined with both I-S cycle and HTE (Verfondern, 2007)

5.2.6 DISCUSSION

Considering the nuclear-assisted hydrogen production technologies discussed in the previous subsections, certain aspects and commonalities arise including:

- 1. Use of an intermediate heat exchanger (IHX)
- 2. Underground placement of the nuclear reactor
- 3. Physical separation of the plants by a safety distance
- 4. Construction of an earthen mound between the facilities
- 5. Storage of the product(s) at the outer perimeters of the plant
- 6. Earthquake-mitigation equipment for the nuclear reactor in most cases
- 7. Most are cogeneration plants
- 8. Presence of a high-temperature (HT) isolation valve in some designs

While the IHX is used to "isolate" or provide a barrier between the primary and secondary systems, points 2 to 5 physically separate the nuclear plant from the chemical facility. These design aspects are employed to mitigate potential hazards from propagating from one plant to the other, as well as to conform to regulations. Similarly, the governing authorities also require earthquake-mitigation equipment. While cogenerating plants are more efficient and offer flexibility regarding operations, the HT isolation valve is a design modification as a result of safety analyses. Current regulations regarding the separation distance between the facilities are based on quantity distance (QD) relationships and are almost inconceivable for any thermally assisted hydrogen production option. From a thermal-hydraulic perspective it would be beneficial if the two facilities were as near as possible to each other, whereas from a safety and regulatory perspective an increased distance between the facilities is preferred (Smith et al., 2005). However, most regulations offer options to decrease this distance if risk analyses are performed in order to prove that the attendant risk is sufficiently low. To this extent, the presence of physical barriers such as an earthen mound between the facilities and underground placement of critical systems are being investigated. These aspects are discussed in more detail in this chapter, but the first aspect to consider is that of the compatibility of the plants.

5.3 COMPATIBILITY OF THE PLANTS

The production of hydrogen by means of a nuclear/chemical complex can only be successful if the nuclear power plant is compatible with the hydrogen production

facility regarding technical and safety requirements. The general requirements for combining the two facilities include effective heat transfer to the chemical plant with minimum reduction of the coolant temperature, minimizing the pressure drop in the coolant and heat transfer loops, using chemically inert coolants, reducing power-to-flow discrepancies in the reactor and capital costs (Yildiz & Kazimi, 2005). Furthermore, the hydrogen facility imposes several requirements on the nuclear power plant including (Forsberg, 2003):

- 1. Reactor power
- 2. Peak temperature and temperature range of delivered heat
- 3. Providing a low-pressure interface with the hydrogen production processes
- 4. Isolating the nuclear plant from the chemical plant
- 5. Tritium contamination

These issues concern the safety and feasibility of the technology and consequently require thorough assessment if the technology is to be successful.

5.3.1 POWER, HEAT AND TEMPERATURE

The considered nuclear power plants have a power output of approximately 400 - 600 MW_t and supply energy in the form of hot helium gas at a temperature in the range of 900 to 950 $^{\circ}$ C, which complies with the requirements of thermally assisted hydrogen production technologies. The peak temperatures of the delivered heat should be minimized in order to reduce thermal and pressure stresses. Figure 5-10 illustrates the temperature of delivered heat for various nuclear reactor concepts (Forsberg, 2003).

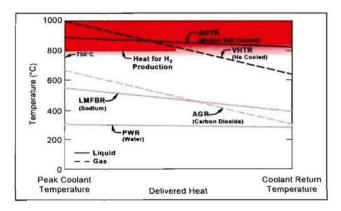


Figure 5-10: Temperature of delivered heat for some reactors (Forsberg et al., 2004)

While some of the reactor concepts discussed in the previous subsection prefers the VHTR design, the remaining options consider Generation 4 (Gen-IV) modular helium reactors. Although not illustrated in Figure 5-10, Gen-IV MHRs, especially those of annular core design, deliver heat with reduced temperature fluctuations across the core (radial direction) and conform to the requirements of power, heat and temperature. Moreover, the modular concept of HTGRs increases its compatibility with hydrogen production plants with regard to power and economics of scale of operation since several HTGR modules can supply heat (and electricity) to the hydrogen facility.

5.3.2 PRESSURE

The pressure at the nuclear/chemical interface is a very important aspect and affects the feasibility and the safety of the complex. According to Forsberg (2003), the pressure at the interface should be low since the chemical reactions in the hydrogen facility go to completion at low pressures; low pressures would minimize the risk of pressuring the chemical plant as well as minimizing the high-temperature materials strength requirements. Furthermore, the low pressure would also minimize the potential risk of heat exchanger failure and release of toxic gases if the chemical plant becomes over-pressurized (Forsberg, 2003). However, a decrease in pressure from the primary to the secondary circuit allows for higher tritium transport to the process side. Furthermore, if the pressure difference between the circuits is substantial, it would result in severe material strength requirements regarding the IHX. Lastly, the PBMR cogeneration plant reaches an economic optimum at a relatively large pressure of approximately 7 MPa in the secondary circuit (PBMR, 2008).

5.3.3 ISOLATION

The chemical plant should be isolated from the nuclear power plant to ensure safe operation of both facilities and in order to adhere to governmental regulations. The term isolated includes the required distance between the facilities as well as system isolation by means of an IHX. The IHX "isolates" the processes from one another, minimizes the risk of contamination (such as tritium) and restricts hazardous events from progressing from one facility to the other (Forsberg, 2003). Initial studies based on probabilistic safety assessments (PSA) show that a minimum spacing of 60 to 110 meters must exist between the hydrogen facility and the nuclear plant (Sherman,

2004). However, Smith et al. (2005) found that the minimum separation distance should be at least 110 m (this study is discussed in more detail at the end of this chapter). The distance between the plants depends on the presence of flammable materials (natural gas, methane and hydrogen) and is usually determined by quantity-distance (QD) relationships according to the applicable governing authority, which differs significantly from country to country.

5.3.4 TRITIUM CONTAMINATION

The production of tritium occurs in the core during normal operation as a ternary fission product, neutron bombardment of the helium coolant and by activation reactions of lithium and boron in the graphite components. Unless damaging of the fuel particles' coating occurs, the tritium produced during the fission process does not escape from the particles to pollute the system (less than 10⁻⁶ percent of the inventory escapes through the coatings; Kugeler, 2005). The tritium released from the damaged fuel coatings and produced from uranium contamination of the core graphite, escape into the coolant where most impurities (including tritium) are removed by the helium purification systems. However, a small amount of tritium is able to transport to the process side by permeating through the heat exchanger tubes into the process streams (Verfondern & Nishihara, 2004a).

5.3.5 DISCUSSION

The thermally assisted hydrogen production technologies are compatible with hightemperature nuclear reactors as related to power, temperature, heat delivery, pressure and (to some extent) isolation. However, this is from a conceptual viewpoint since no commercial plant exists where they have actually been coupled and some very important aspects have been identified that still require significant R&D. One of these aspects is the selection of materials that are able to withstand the extreme temperatures, pressures and corrosive environments they will be applied to. From an engineering perspective, these are very high temperatures and at the limit of current engineering technologies. With regard to the heat transfer system, the size (up to 2400 MWt), temperature (~800 $^{\circ}$ C), and distance (500 to 1000 m) are substantially beyond industrial experience (Forsberg, 2003). Another aspect is that of isolation, which includes process isolation and physical separation of the plants. Process isolation is achieved (to some extent) by employing IHXs, however, some of the tritium and hydrogen is still able to transport through the tubes of the heat exchanger to "contaminate" the other loop. This is particularly possible at the high temperatures and pressures that the IHX is proposed to operate at. Three options exist regarding tritium contamination, the reactor can be designed to minimize tritium production, tritium can be trapped in the coolant or heat exchangers can be specially designed to minimize tritium transport. Additionally, high-temperature bake-out of the graphite during the manufacturing process can reduce the amount of lithium impurities in the graphite (Forsberg, 2003). Regarding the physical separation distance between the plants, if the distance is to be substantial, it would increase the material requirements of the heat transfer loop as well as the corresponding economic investment and technological feasibility (if heat loss is considered). Therefore, the next issue to be discussed is the interfacial equipment and connection technologies proposed to address these concerns.

5.4 INTERFACIAL EQUIPMENT

The interfacial equipment forms the "connection" between the nuclear and chemical facilities as well as functioning as a barrier between the two processes. The IHX, nuclear steam reformer, heat transfer ducts and (to some extent) the high-temperature isolation valve fall into the category of interfacial equipment.

5.4.1 THE INTERMEDIATE HEAT EXCHANGER

In the IHX, the primary He system exchanges heat with the secondary He circuit to transfer heat from the nuclear plant to hydrogen production facility such that the facilities remain relatively "isolated". Figure 5-11 illustrates the concept of the IHX.

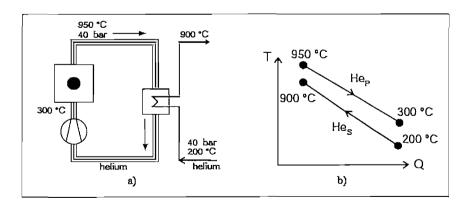


Figure 5-11: Primary circuit of a modular HTGR with IHX a) Principle flow diagram, b) T-Q diagram for use of nuclear heat (Verfondern, 2007)

The combination of the nuclear reactor with the chemical processes requires a decoupling between the primary circuit and the heat utilization system (secondary circuit) for the following reasons (Verfondern, 2007):

- Separation of the nuclear plant for safety reasons (vice versa contamination)
- Limitation of radioactive contamination of the product (i.e., tritium)
- Exclusion of ingress of corrosive process media into the primary circuit
- Near-conventional design of heat utilization system
- Ease of maintenance and repair of heat utilization system
- Exclusion of contamination of high industrial investments

According to Verfondern (2007), the IHX establishes a physical barrier between the nuclear and process heat plant such that the heat application facility may be conventionally designed and repair works to be conducted under non-nuclear conditions. Furthermore, under normal conditions the IHX prevents the primary coolant from entering the process plant as well as the process gases from being transported through the reactor systems and reactor containment (Verfondern, 2007). While Germany has done significant R&D regarding the IHX in the past, Japan, France and the USA are actively investigating "new" IHX concepts.

5.4.1.1 GERMANY

In Germany, the employment of an IHX was suggested within the PNP project and several IHX concepts were tested in the KVK experiments. The concept of combining an IHX with a reactor similar to the HTR-Modul is illustrated in Figure 5-12 and would involve a side-by-side arrangement of a reactor and IHX vessel for each modular unit. The thermal power of the nuclear reactor and of the IHX were to be limited to 170 MW_t due to the requirement of self-acting decay heat removal if a total loss of active cooling accident was to occur. Figure 5-12 illustrates this concept with an IHX of helical-tube or U-tube design (Verfondern, 2007).

The Prototype Plant Nuclear Process Heat Project (PNP) focussed on the nuclearassisted steam gasification of hard coal and considered the use of an IHX. Within the PNP project, a facility for large component testing (KVK) was constructed and successfully operated by INTERATOM (Harth, 1990 as cited in Verfondern, 2007). The KVK facility included a heating system with a total thermal power of 10 MW that heated helium to 950°C at 4.0 MPa. Furthermore, the plant also tested hot gas ducts

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of large diameter, a steam generator, valves for hot helium and other components such as hot headers or auxiliary plants for gas purification (Verfondern, 2007). The KVK plant tested two IHX components namely a helical-tube bundle constructed by the Steinmüller company and an U-tube bundle constructed by the Balcke-Dürr company (Verfondern, 2007). Figure 5-13 shows these IHX components (note: figure for illustration purposes only due to the legends being in German and no English versions being available).

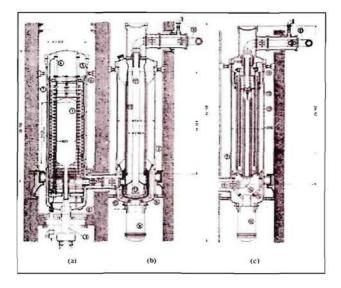


Figure 5-12: Arrangement of (a) a reactor based on the HTR-Modul-type and (b) Helical-IHX or (c) U-tube-IHX (IA, 1983 as illustrated in Verfondern, 2007)

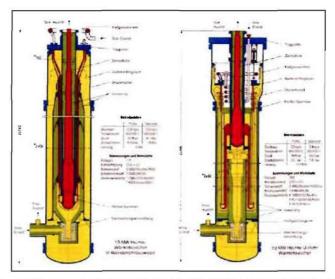


Figure 5-13: Two IHX components tested in KVK: (left) Helical tube bundle and (right) U-tube bundle (Verfondern, 2007)

5.4.1.2 JAPAN

The IHX employed at the HTTR project is a vertical, helically coiled, counter-flow type heat exchanger. As quoted from Verfondern (2007):

"The primary helium enters the IHX through the inner pipe of the primary concentric hot gas duct attached to the bottom of the IHX. It flows upwards outside the tubes transferring the nuclear heat of 10 MW to the secondary helium cooling system and flows back through the annular space between the inner and outer shells. The secondary helium flows downwards inside the heat transfer tubes and flows upwards in the central hot gas pipe through the hot header".

Figure 5-14 shows a schematic illustration and a photograph of the He-He IHX in the HTTR. In principle, a similar IHX is to be used in the GTHTR300C cogeneration plant due to the success of the HTTR project (Verfondern, 2007).

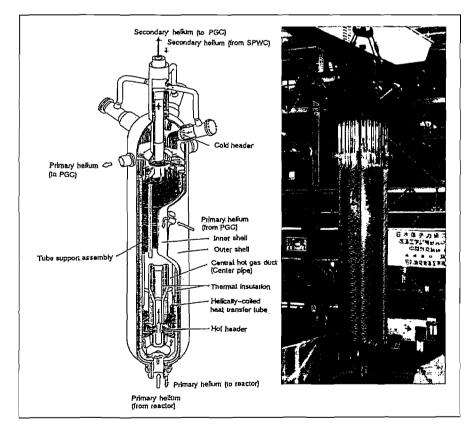


Figure 5-14: Schematic and photograph of the He-He IHX in the HTTR (Verfondern, 2007)

5.4.1.3 USA (H2-MHR)

In the US H2-MHR project, the IHX is the so-called Printed Circuit Heat Exchangers (PCHE) developed by the Heatric Company. As quoted from Verfondern (2007):

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"A heat exchanger module is composed of metal plate layers containing alternately coolant channels for the primary and for the secondary fluid flowing (e.g.) counter to each other (top right). The flow channels with a semicircular profile (top left) are chemically edged into the plates using a technique similar to that for printing electrical circuits. This manufacturing technique makes complex streams possible".

The following figure (Figure 5-15) illustrates the concept of printed circuit heat exchangers (Verfondern, 2007).

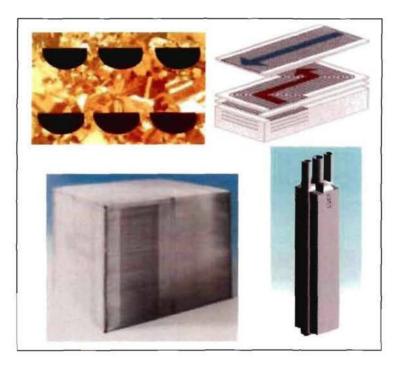


Figure 5-15: Printed Circuit Heat Exchanger, PCHE (HEATRIC as illustrated in Verfondern, 2007)

PCHE designs are highly compact, highly robust, have high thermal efficiencies and allows operating pressures of up to 50 MPa and temperatures of 900°C to be realised. Moreover, the basic modules can be adjusted to construct heat exchangers of any desired scale (Verfondern, 2007).

5.4.1.4 FRANCE

The new compact IHX designs currently under investigation in France for the ANTARES project are the plate machined heat exchanger and the plate fin heat exchanger (Breuil, 2006 as cited in Verfondern, 2007). Figure 5-16 illustrates the two plate fin heat exchangers (PFHEs) under development, namely the Brayton energy design and the Nordon design. The Brayton Energy design has wavy or straight fins on a flat support plate, while the Nordon design employs a different type of fins, serrated offset strip fins, on a support plate (Verfondern, 2007).

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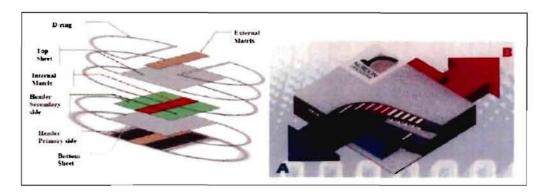


Figure 5-16: Two variants of plate fin IHX Brayton Energy design (left) and Nordon design (right) (Breuil, 2006 as illustrated in Verfondern, 2007)

Figure 5-17 shows a potential design of an IHX vessel for ANTARES containing eight plate-type IHX modules in symmetrical arrangement (Verfondern, 2007).

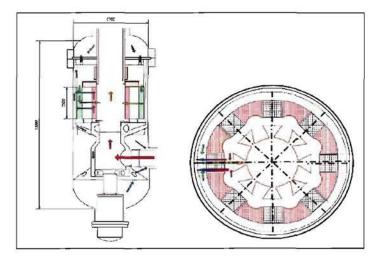


Figure 5-17: IHX vessel with integrated plate IHX modules (Breuil, 2006 as illustrated in Verfondern, 2007)

5.4.2 NUCLEAR STEAM REFORMER

The nuclear steam reformer is a helium-heated steam reformer in which the catalytic steam reforming of light hydrocarbons takes place to produce hydrogen. In the German approach (Figure 5-18), the steam reformer is directly coupled with the primary circuit, while the Japanese approach (Figure 5-19) involves the use of an IHX. Verfondern (2007) states that the helium-heated steam reforming process is well understood and tested on a large scale.

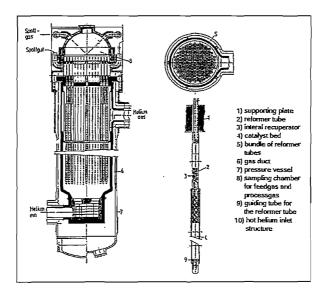


Figure 5-18: Technical concept of a helium-heated steam reformer connected to a modular process heat HTGR (Verfondern, 2007)

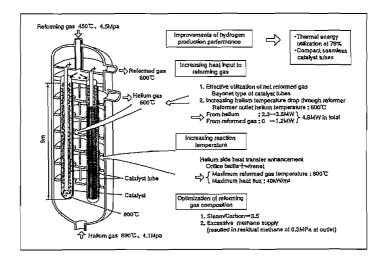


Figure 5-19: New concept helium-heated steam reformer for the HTTR/SMR (Verfondern & Nishihara, 2004)

5.4.3 HOT GAS DUCT

The hot gas duct transports the hot helium leaving the core to the steam generator, gas turbine, nuclear steam reformer or intermediate heat exchanger depending on its application. The hot gas duct forms part of the nuclear system and should be designed accordingly. The temperature of hot helium is generally in the range of 700° C - 950° C at 4 - 7 MPa, which result in high material and construction requirements for this component, especially considering operating lifetimes of 40 - 60 years. Moreover, it should be designed to minimize pressure drops and temperature losses and be able to withstand accidents like depressurization of the primary circuit. Compensation for cold and hot states and tightness are other important requirements. Furthermore, the components must be designed to withstand vibrations and loads from earthquakes. Figure 5-20 shows the concept of a hot gas duct for a 200 MWt modular HTR (Kugeler, 2005).

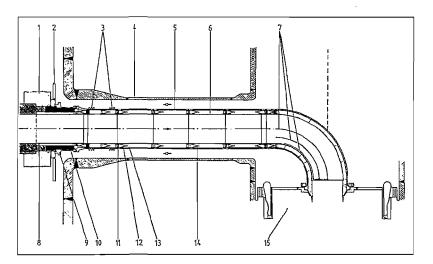


Figure 5-20: Hot gas duct of a 200 MWt modular HTR (Kugeler, 2005)

In Figure 5-20, the following is applicable (Kugeler, 2005): 1) graphite structure 2) core barrel 3) compensator 4) connecting vessel 5) bearing tube 6) metallic liner 7) guiding plate 8) graphite tube 9) intermediate flange 10) fibre insulation 11) – 13) insulation material 14) metallic plates 15) steam generator.

Most hot gas duct concepts are coaxial with hot gas flowing in the inner pipe and the cold gas in the outer annular space (see Figure 5-21). According to Kugeler (2005), the principle of coaxial ducting of hot gas has been tested in several large helium test-facilities (EVA II-, HHV-, EVO-, KVK-plant) and can be considered a proven

technology. Figure 5-21 illustrates the hot gas duct of the EVO-Plant as well as giving details on fibre insulation (Kugeler, 2005).

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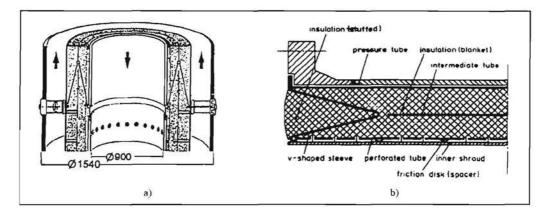


Figure 5-21: Details of insulation systems for hot gas ducts: a) hot gas duct of EVOplant (helium-temperature of 750°C), b) detail of fibre insulation (Kugeler, 2005)

5.4.4 HIGH-TEMPERATURE ISOLATION VALVE

The high-temperature isolation valve proposed to be used in the Japanese projects is still under development and will be used to mitigate thermal deformations that may occur at the high operating temperatures required for the processes associated with the utilization of the nuclear heat (Verfondern & Nishihara, 2004a). Figure 5-22 illustrates the concept of the HT isolation valve.

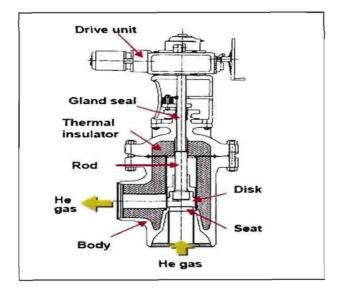


Figure 5-22: High Temperature Isolation Valve (Verfondern & Nishihara, 2004a)

The driving force behind the development of the HT isolation valve probably lies in a PSA related study conducted on the combined HTTR/SMR complex for hydrogen production, which is discussed in a subsequent section of this report.

5.4.5 DISCUSSION

In light of the overviews regarding interfacial equipment, it can be concluded that:

- the concept of the IHX has been tested extensively in the KVK experiments, but several innovative IHX concepts are being investigated
- the helium-heated steam reforming process is regarded as a medium term option and is well understood and tested on a large scale
- the principle of coaxial ducting of hot gas has been tested in several large helium test-facilities and can be considered a proven technology
- the HT isolation valve still require significant R&D

With regard to the IHX, its application to the size of operation and the environment in which it is to be employed in nuclear cogeneration plants still have to be verified. Particularly materials development at the desired high temperatures and pressures require investigation, especially considering their extensive operational lifetimes and the contamination hazards that are associated with heat exchanger failure. Additionally, alterations such as using coated heat exchanger tubes to reduce tritium transport to the chemical process require investigation under appropriate operating conditions. Even though the helium-heated steam reforming process is well established, the helium-heated reactors associated with the other thermochemical processes still require significant R&D. However, the principle of heat exchanger reactors is well understood and established in the chemical industry. Coaxial ducting of hot gas as it is to be used in cogeneration plants also need validation tests under the expected operating conditions and environment. In conclusion, since the requirements of a combined complex and interfacial equipment are known, the next issue to be discussed is that of the safety of the combined complex, which include identification of hazards.

5.5 SAFETY ASPECTS OF THE COMBINED COMPLEX

The safety aspects of the combined complex, as it is related to this study, are the safety aspects of concern when a hydrogen production facility is coupled to a nuclear plant such that either the inherent safety of the nuclear plant or the safety of the chemical plant is compromised, specifically due to the connection of the plants. Therefore, the purpose of this section is not to investigate the safety of nuclear plants or stand-alone hydrogen production facilities, but rather the identification and evaluation of additional hazards that arise due to coupling of the two critical facilities.

5.5.1 HAZARD IDENTIFICATION

A preliminary identification of the hazards associated with such a combined nuclear/chemical complex is:

- 1. Presence of hazardous components (flammable and radioactive)
- 2. Release of radioactive or other hazardous components
- 3. Release and evolution of a flammable gas cloud
- 4. Ignition and combustion of a flammable gas cloud
- 5. Heat radiated and over-pressures generated during combustion
- 6. Tritium contamination of the product
- 7. Hydrogen ingress into the reactor core
- 8. Thermal turbulence induced by upsets in the chemical plant
- 9. Chemical plant is considered a heat sink of the complex
- 10. Interfacial equipment failure
- 11. Hydrogen embrittlement and decarburization

While hazards 1 to 6 are primarily potential dangers to humans (operating personnel and consumers), hazards 7 to 11 may influence the operation of the nuclear reactor. However, except for hazard 6, which is only a potential danger to consumers and operating personnel, all the hazards may affect the safe and/or continuous operation of the plant. The extents to which these hazards are able to influence the safety of the complex depend on plant specifics (mitigation measures and design characteristics), the degree to which they occur (for instance the complete or partial release of hazardous inventory), the layout of the complex (distance between facilities) and the location of the hazard (if not specified).

The primary mitigation measure regarding aspects 1 to 5, with regard to flammable substances, is that of physical separation of the plants and includes safety distances, employment of physical barriers between the facilities and underground placement of critical systems. The principle behind physical separation of the plants is that when the plants are separated by a sufficiently large distance, the hazards associated with the release and/or combustion of flammable or hazardous components at the chemical facility will not have a significant impact on the safety or operability of the nuclear plant. Since the release of radioactive material from the nuclear plant is considered the most hazardous event that could occur, metrics such as core damage frequency (CDF) and large early release rates are used as factors that constitute the safety of the complex when LWRs are involved. The extremely remote probabilities of these events to occur when Gen-IV HTGRs are involved make these metrics essentially meaningless and risk-based methodologies such as examined in NUREG 1860 should be considered. NUREG 1860 is a relatively "new" "regulation" (published in December 2007) in which the feasibility of implementing a risk-informed and performance-based regulatory structure for future plant licensing is evaluated. Although it is clear that current nuclear regulations, which is LWR-based, will not be efficient or effective in regulating HTGRs, the lack of studies performed with NUREG 1860 in mind necessitated that this investigation focus on current, active nuclear regulations.

The combined complex should be designed and constructed such that no possible hazard originating from one of the plants could propagate to the other plant to affect its safety. As the nuclear plant should always be considered as inherently safe, the onus of responsibility may probably lie more towards the nuclear facility, especially considering problems regarding licensing by the nuclear regulators and public acceptance of the technology. Therefore, the nuclear plant must be designed to withstand any hazard originating from the chemical facility (even though it will be a concomitant effort of both of the facilities to improve and promote safety). As related to the presence of flammable and toxic substances at the chemical facility, the nuclear facility should be able to address issues regarding heat radiation and peak overpressures generated by the combustion of a flammable gas cloud, as well as the possibility of hazardous substances released at the chemical facility to reach and enter the nuclear building. Given that it is extremely improbable that a hazardous substance released at the chemical facility could travel to the nuclear plant (due to their dispersion characteristics and a sufficient safety distance between the plants), the impacts of heat radiation and peak overpressures are more applicable. To this

extent, the outer concrete structure of the nuclear plant, as well as the systems contained within it should be able to withstand the impacts of heat radiation and peak overpressures without compromising the safety and operability of the nuclear plant. Given that Gen-IV nuclear plants are designed to withstand the impacts of an airplane crash and significant earthquake, it will probably be able to withstand the impact of the peak overpressures generated by a hydrogen or methane explosion or deflagration. However, since the standard mitigation measure regarding hydrogen and hydrocarbon fires is to stop the supply of the flammable component and let the fire burn out, how would the nuclear facility be affected by prolonged exposure to high intensity heat radiation? Would the integrity of the concrete structure or the operability of the nuclear systems be compromised during such an event? Considering the characteristics of heat radiation and the flammable components involved, aspects such as the emissivity, amount, concentration and release rate of the flammable substances, as well as the ambient conditions, presence of physical barriers, exposure period and distance to the fire will play vital roles in answering the above-stated questions. These aspects involve design aspects and are specific to the plant being evaluated and will therefore not have a universally applicable answer. However, if the appropriate mitigation measures are employed, the consequences of this hazard can be moderated to such an extent that it will not affect the safety of the nuclear plant or the feasibility of the combined complex.

Tritium contamination of the chemical process and hydrogen ingress into the nuclear cycle occur due to hydrogen's and its isotopes' ability to permeate through intact metals, especially at the proposed operating conditions of high temperature and pressure. This hazard involves process isolation and is to be achieved by the employment of an IHX to form a physical barrier between the chemical and nuclear heat transfer loops. However, the effectiveness of this isolation depend significantly on the materials of construction of the IHX, the pressure difference between the nuclear and chemical heat transfer loops and alterations to the IHX such as the use of coated heat exchanger tubes to reduce permeation of hydrogen and its isotopes through the walls of the heat exchanger.

In cogeneration nuclear plants a (significant) portion of the heat generated in the nuclear reactor is used by the chemical plant to drive the endothermal chemical reactions required for hydrogen production. Therefore, the chemical production plant may be considered as a heat sink of the nuclear plant since it "removes" heat from the nuclear cycle. If the chemical process plant were unavailable, it would result in a

significant amount of heat energy to be "recycled" back to the nuclear reactor and consequently, the heat removal and reactivity control mechanisms will be required to manage the discrepancy in heat removal. This could result in a nuclear reactor scram event in which the reactivity in the nuclear reactor is reduced to such an extent that the fission process is stopped and only decay heat is generated. However, this outcome is unlikely since Gen-IV HTGR designs are able to handle load rejection without scram and the reactor is able to adjust quickly to lower demand, particularly if a steam dump valve is included in the secondary system.

A related event to consider is that of thermal turbulences in the nuclear system due to upsets in the chemical plant. In this event, the chemical process is not necessarily unavailable to remove heat but due to complications in the chemical process it is not able to remove all the heat it was designed to remove from the nuclear cycle. Therefore, this event would not result in a nuclear reactor scram but may challenge the control and operability mechanisms of the nuclear plant. With regard to these events that result in thermal turbulences in the nuclear system, coupling of the nuclear and chemical plants pose a more significant complexity regarding control and operability than the steam or gas cycles in electricity dedicated nuclear power plants. This is obviously due to the more complex nature and setup of the chemical production facility in which more events could result in thermal turbulences or plant unavailability (more components and equipment and "complex" chemical reactions and separation technologies). However, simulations such as the HTR-Modul, PNP and HTTR/SMR projects indicate that these events are manageable if appropriate mitigation measures are taken. Interfacial equipment failure is considered a very hazardous event since it may result in one of more of the following events:

- chemical process gases or radioactive material being released into the environment,
- radioactive contamination of the chemical process,
- process gases entering the nuclear cycle,
- depressurization accident in the nuclear cycle,
- thermal turbulences in the nuclear cycle or
- insufficient heat transfer to the chemical process.

Therefore, failure of the interfacial equipment may be an initiating event of many of the hazards identified at the start of this subsection and correspondingly makes interfacial equipment of critical importance. Most of these issues can be addressed by appropriate material selection and proper design such that a rupture of any of these equipment is extremely unlikely.

The last hazard identified is that of hydrogen embrittlement, which is another hazard that can be mitigated by proper materials selection and development. Since hydrogen embrittlement and the accident phenomena associated with flammable and combustible substances were addressed in the previous chapter (Chapter 4), the next issues to investigate are process isolation to limit hydrogen and tritium transport, thermal turbulences due to upsets in the chemical plant, release of flammable substances inside the reactor building and physical separation requirements.

5.5.2 TRITIUM AND HYDROGEN TRANSPORT

As mentioned previously, tritium is the radioactive isotope of hydrogen and is produced in the ruclear reactor as a temary fission product, by neutron bombardment of the helium coolant and by activation reactions of lithium and boron impurities in the graphite components. Since tritium is radioactive, no product that has a tritium concentration above the applicable legal limits may be sold to or used by the public. Concerning nuclear assisted hydrogen production technologies, tritium is sufficiently contained within the TRISO coated fuel particles (only 10⁻⁵ percent of the inventory escapes), however, a small amount of damaged particles release their fission product inventory into the coolant. Even though all HTGR nuclear reactors and next-generation concepts contain helium purification systems (removal of tritium and other impurities from the primary system), a small amount of tritium lingers in the primary system, a fraction of which is able to permeate through the heat exchanger tubes to the chemical process. The tritium production and release rate into the helium coolant for the 170 MW, process heat HTR-Modul are shown in the following table (Table 5-1) and indicates that approximately 1/3 of the tritium produced in the reactor system enters the helium coolant (Verfondem, 2007).

Tritium source	Tritium production rate [10 ³ Bq/s] ([%])		Tritium release rate into coolant [10 ³ Bq <i>ls</i>] ([%])	
	Initial phase	Equilibrium	Initial phase	Equilibrium
Fission	898 (14)	1245 (51)	89 (4)	126 (12)
Li-6	4721 (76)	846 (34)	1413 (66)	529 (52)
He-3	628 (10)	367 (15)	628 (30)	367 (36)
Total	6247 (100)	2458 (100)	2130 (100)	1022 (100)

Table 5-1: H ₃ production and release for the 170 MW _t HTR-Modul (Verfond	ern, 2007)
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At the high temperatures associated with nuclear assisted thermochemical production technologies, hydrogen and its isotopes are highly permeable through the heat exchanger tubes and therefore require mitigation. The following figure (Figure 5-23) illustrates the transport paths of hydrogen and tritium (Note: HT denotes H_3 in this figure only) for the HTTR/SMR project (Verfondem, 2007).

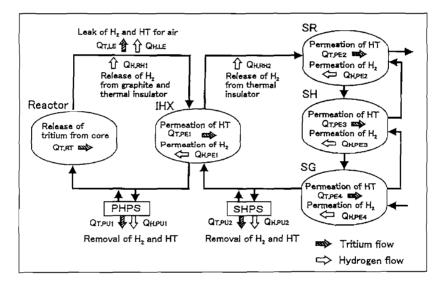


Figure 5-23: Tritium ("HT") and hydrogen balance in HTGR H₂ production system (Verfondern, 2007)

In this figure, SR, SH, SG, PHPS and SHPS represent the steam reformer, super heater, steam generator, and primary and secondary helium purification systems respectively. According to Verfondem (2007), there are three approaches to reduce the tritium concentration in the chemical process section and the resultant products. These are:

- 1. Oxide layers on the heat exchanging surfaces
- 2. A gas purification system to remove tritium from the primary circuit

3. An intermediate circuit purified by a sweep gas

The presence of oxide layers on the heat transfer surfaces and the use of coated IHX tubes significantly reduce the permeability of tritium, as can be seen for the HTTR/SMR system in the following table (Table 5-2; Verfondern & Nishihara, 2004a)

	Tritium			
Tube surface		Purification	Hydrogen	concentration
IHX	SR, SH, SG	rate [kg/h]	release	hydrogen [Bq/g]
Clean	Clean	200	No	89.5
Clean	Oxidized	200	No	20.5
Coated	Oxidized	200	No	8.9
Defect	Oxidized	200	No	12.0
Coated	Oxidized	400	No	5.3
Clean	Oxidized	800	No	8.5
Coated	Oxidized	200	Yes	6.8

Table 5-2: Tritium permeation for steady-state operation of the HTTR (Verfondern & Nishihara, 2004)

As mentioned previously, all HTGRs have helium or gas purification systems to remove tritium and other impurities from the primary circuit. However, the addition of "getter materials" for hydrogen and tritium into the purification system could improve its efficiency and reduce the permeation of tritium to the product and the amount of hydrogen present in the primary circuit (Verfondern, 2007). Getter materials are components that have a high affinity for certain components such that when they are injected into the system, they immediately "combine" with that component upon contact, thereby changing its characteristics (physically or chemically) and essentially removing it from the system or increasing its probability to be removed from the system.

The ingress of hydrogen into the primary circuit is also of concern since it causes corrosion of the graphite structures and the corresponding release of carbon into the primary circuit. While corrosion affects the integrity of the graphite structures, the transport of carbon in the helium circuits could lead to carbon deposition on the surfaces of high temperature alloys, which in turn could result in changing their material properties (Verfondern, 2007).

5.5.3 THERMAL TURBULENCES

A HTGR supplying process heat to drive endothermal chemical reactors will exhibit thermal turbulences due to the chemical reactions being dependent on the amount and temperature of heat supplied to the chemical reactor. Obviously, the heat required by the chemical reactor is also dependent on the flow rate of reactants into the reactor at the appropriate concentrations. Therefore, if there is a change in the heat supplied to the chemical reactor (due to a nuclear reactor scram or any failure resulting in a heat transfer transient) it will affect the chemical conversion process. In case of a nuclear reactor scram, no heat will be transferred to the chemical process (the decay heat is removed by the applicable nuclear safety systems) or the transfer of heat will decrease dramatically (only decay heat generated in the nuclear reactor). The most probable event during a nuclear reactor scram is that no heat will be transferred to the chemical process, which should result in the instantaneous cut-off of the reactants to the chemical reactor for safety, operability and economic reasons. Heat transfer transients during normal operation of the nuclear reactor system will be very low since the HTGRs will supply heat at a near constant high temperature with small variation in peak temperature (requirement of the combined complex as discussed previously). However, more likely events to initiate thermal turbulences in the combined complex are those due to transients or failures in the chemical process. Considering the SMR process, this could be due to a change in the flow rate of either feed gas (methane) or water to the steam reformer. However, this aspect could be extended to the supply of reactants to the endothermal chemical reactors of all themochemical cycles. Since the chemical reaction is endothermal, it removes heat from the primary system due to the conversion process and a disruption in the flow rate of either of the reactants will result in less heat being removed from the nuclear cycle. Consequently, the helium returning to the nuclear reactor has a significantly higher temperature than that of normal operating conditions and could result in a nuclear reactor scram. Regarding the HTTR, the reactor will scram if the temperature of the helium returning to the IHX exceeds the allowable limit. However, this will depend on the specific reactor under consideration and whether the incoming helium is used to condition the RPV, which will have limitations but which can be decreased or designed out by suitable measures. In this regard, safety measures are required to mitigate potential thermal disturbances as a result of transients in the chemical process such that continuous reactor operation without nuclear reactor scram is ensured. The safety requirement for this event in the HTTR/SMR project is to limit the secondary helium temperature variation within ± 15°C at the inlet of the

IHX to prevent a reactor scram (Verfondern & Nishihara, 2004a). Similar limitations will exist for all thermochemical, nuclear-assisted hydrogen production technologies, although the limitation will depend on the specific HTGR under consideration. With respect to the chemical process being a heat sink of the nuclear complex, under current regulations it is not allowed to be the ultimate heat sink of the nuclear plant since this function is limited to water and/or air and cannot be electricity or chemical energy as the result of a conversion process. Therefore, the chemical process is not designed to function as a safety system of the nuclear plant; these are exclusively left to the safety systems of the nuclear facility (Verfondern & Nishihara, 2004a & 2005; Verfondern, 2007).

5.5.4 RELEASE OF FLAMMABLE SUBSTANCES INTO THE NUCLEAR REACTOR BUILDING

In most of the thermochemical, nuclear-assisted hydrogen production technologies the intermediate heat exchanger is situated within the nuclear reactor building. Considering the SMR process, this allows for the possibility of flammable substances being released into the reactor building where it may lead to a deflagration or detonation hazard depending on the characteristics of combustion. However, the probability of this event to occur is extremely remote since it requires a rupture or leak in both the chemical reactor and the secondary heat transfer loop within the reactor building (see Figure 5-24). When this occurs, flammable feed and/or product gas escape into the secondary helium system, from where it is released into the reactor building through the leak or rupture in the secondary circuit located within the reactor building.

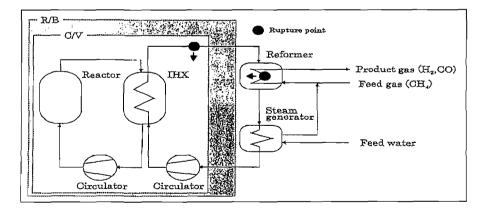


Figure 5-24: Ingress of flammable gases into the reactor containment (Verfondern, 2007)

The only conceivable event that could result in this hazard is that of an earthquake of significant strength and consequently these components should be designed for a high seismic safety level (Verfondern, 2007). Therefore, it is extremely unlikely that this hazard could occur but since the consequences of a fire or explosion within the reactor building are very severe, it should be considered during any safety analysis of a proposed nuclear/chemical complex that has the probability of flammable substances escaping into the secondary heat transfer loop. Considering the multistep thermochemical water splitting cycles (HyS, I-S), the reactants are non-flammable and the products are produced after several reaction "steps", after which they are removed from the recycling stream such that they cannot enter the secondary heat transfer loop at sufficient concentrations to pose a combustion risk if the hazard described above is to occur.

5.5.5 PHYSICAL SEPARATION REQUIREMENTS

Physical separation requirements include safety distances, which are usually based on quantity distance relationships, as well as other physical means of separation such as earthen mounds and (underground) placement of key facilities. These requirements are established by the applicable governing authorities and employed to protect the facility by mitigating the propagation of a hazard from one site to another. The principle separation requirement is that of safety distance, which is the distance required between the possible location of a hazard and the object to be protected. Considering a nuclear/chemical complex, this distance relates to the distance between the location of a flammable gas leakage and the nuclear plant, while taking into account the developing flammable atmosphere as well as the heat radiated and pressure waves generated by combustion of the flammable gas. The safety distance is usually determined as a function of the quantity of the flammable substance(s) relating to certain threshold values such as dose of thermal radiation and peak overpressure (BRHS, 2006). From a thermal-hydraulic perspective it would be beneficial if the two facilities were as near as possible to each other, whereas from a safety and regulatory perspective an increased distance between the facilities are preferred (Smith et al., 2005). To this extent, physical barriers such as earthen mounds and underground placement of critical facilities may be employed to reduce the separation distance required. The applicable US and German regulations regarding quantity distance relationships are illustrated in the following figure (Figure 5-25; BRHS, 2005).

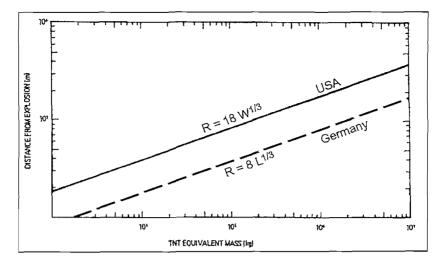


Figure 5-25: Quantity Distance relationships according to US and German regulations (BRHS, 2005)

According to Smith et al. (2005), the US Regulatory Guide 1.91 is related to:

"Structures, systems, and components important to safety and designed for high wind loads are also capable of withstanding pressure peaks of at least 7 kPa resulting from explosions".

Furthermore, no additional mitigating measures need to be taken if the following equation is met (Verfondern & Nishihara, 2004a; INEL, 1994):

$$R = 18W^{1/3}$$
 Equation 5-1

With:

- R Distance (m)
- W TNT equivalent of explosive substance (kg)

Due to the extreme distances obtained by Equation 5-1, this approach appears to be unrealistic for any thermochemical or hybrid thermochemical process employed at the nuclear/chemical complex. However, the regulation offers additional options such as risk analysis for further reduction of the safety distance. These options could include proving that the probability of the entire explosive inventory exploding is extremely remote, or that the explosive characteristics of hydrogen gas and TNT explosive is sufficiently different to warrant alteration, or that the attendant risk is sufficiently low (Verfondern, 2007; Verfondern & Nishihara, 2004a; Verfondern & Nishihara, 2005).

The German BMI regulation of 1976 regarding the "Protection of Nuclear Power Stations from Shock Waves Arising from Chemical Explosions" has the following quantity distance relationship associated with a peak overpressure of 30 kPa (Verfondern, 2007; BRHS, 2006):

$$R = 8L^{1/3}$$
 Equation 5-2

With:

- R Distance (m)
- *L* TNT equivalent of explosive substance (kg)

However, this legislation allows for reductions in the separation distance according to type of explosive material, but has to obey to a minimum distance of 100 m. Accordingly, as related to pressurized gaseous hydrogen the factor of 8 reduces to 6.3 (BRHS, 2006). As stated in Verfondern & Nishihara (2004a), the guideline is applicable to the currently operating fleet of nuclear power plants and it is explicitly mentioned that "*no statement can be given at present concerning its application to future nuclear process heat plants*". The authors elaborate that "*it is supposed to be a concomitant effort with the development of nuclear process heat plants to solve the problem of external vapour cloud explosions*" (Verfondern & Nishihara, 2004a). In order to access the implications of these regulations, the following table (Table 5-3) shows the separation distances obtained thereby.

Variable/Method	BMI	BMI (Reduced)	US RG 1.91
Hydrogen Production rate [kg/s]	2.00	2.00	2.00
Hydrogen Production rate [kg/day]	172800.00	172800.00	172800.00
On-site storage [kg]	172800.00	172800.00	172800.00
TNT Equivalent Factor [kg TNT/ kg H2]	26.50	26.50	26,50
Equivalent mass TNT stored onsite [kg]	4579200.00	4579200.00	4579200.00
Multiplication factor [m/kg^1/3]	8.00	6.30	18.00
R [m]	1328.47	1046.17	2989.07
R [km]	1.33	1.05	2.99

Table 5-3: Separation distances according to various regulations

From a thermal-hydraulic perspective, these distances are very large and a great amount of heat loss will be incurred during transport of heat to the chemical production facility. Fortunately, both the regulating authorities and industry acknowledge these distances to be of concern and it is expected that regulations specific to nuclear-hydrogen technologies are to be developed and implemented.

If the hydrogen production facility is considered as a change to the currently operating fleet of nuclear power plants, the decision criteria falls under RG 1.174 to allow or disallow the changes. The US regulatory guide RG 1.174 is a risk-informed regulation that has the following general principles (Smith *et al.*, 2005):

- 1. The application meets current regulations unless it explicitly relates to a requested exemption or rule change.
- 2. The application is consistent with the general defense-in-depth philosophy.
- 3. The application maintains sufficient safety margins.
- 4. The application maintains small risk and is consistent with the intent of the NRC's Safety Goal Policy Statement.

Smith *et al.* (2005) consider defence-in-depth the most important principle regarding next-generation nuclear facilities and state that even if these facilities may be demonstrated to be safer than the current generation of nuclear power plants, the principle of defence-in-depth may still be required to account for uncertainties inherent in the safety of plant operations.

According to Smith *et al.* (2005), assessment of the change according to RG 1.174 requires that all safety impacts of the issue be evaluated in an integrated manner to improve the operational and engineering decisions. Since the quantification of risk is a fundamental part of this process, appropriate metrics such as CDF and large early release frequency should be used as bases for PSA. Moreover, the NRC has

"specifically requested that appropriate consideration of uncertainty be given in the analyses and that an interpretation of findings be performed as part of any analysis" (Smith et al., 2005).

To this extent, Figure 5-26 illustrates the decision criteria for risk-informed applications according to RG 1.174.

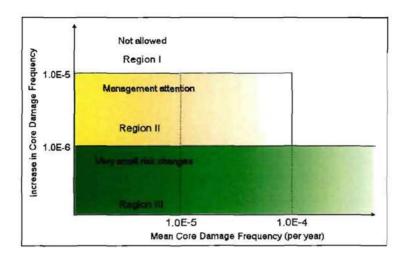


Figure 5-26: Decision criteria for risk-informed applications at the NRC (from RG 1.174 as illustrated in Smith et al., 2005)

From this figure, it is clear that if the hydrogen production facility results in an increase in CDF in excess of 10^{-6} /yr, the regulating authorities will significantly scrutinize it. Even though this regulation (RG 1.174) has only been applied to the current generation of nuclear plants, Smith *et al.* (2005) believe that the next-generation would be held to similar or even stricter limits.

Another regulation to consider is that of RG 1.78, which is "Evaluating the Habitability of a Nuclear Power Plant Control Room during a Postulated Hazardous Chemical Release". This guide discusses the protection of nuclear power plant control rooms and includes adequate protection of the control room from chemical dispersions events, primarily originating from storage tanks, cars, barges, and *etcetera*. However, the utilization of hazardous chemicals on site (as would be the case during nuclear assisted production of hydrogen) also falls under the umbrella of this regulatory guide (Smith *et al.*, 2005). This guide indicates additional criteria for control room habitability such that:

"Any hazardous chemical stored onsite within 0.3 miles [482 m] of the control room in a quantity greater than 100 pounds [45 kg] should be considered for control room habitability evaluation. Hazardous chemicals should not be stored within the close proximity (generally within 330 feet [91 m] or less) of a control room or its fresh air inlets, including ventilation system intakes and locations of possible infiltration such as penetrations. Small quantities for laboratory use, 20 pounds [9 kg] or less are exempt. The maximum allowable

inventory in a single container stored at specified distances beyond 330 feet [91 m] from the control room or its fresh air inlet varies according to the distance and the control room type" (NRC 2001a as quoted from Smith et al., 2005).

In light of these regulations, it seems that the safety distance as determined by quantity distance relationships will be the determining factor regarding the physical isolation of the plants, and possible even the ultimate success of the technology. Therefore, it is of utmost importance to investigate the options available for reduction of the safety distance, one of which is risk analysis through probabilistic safety assessments (PSA).

5.5.5.1 PSA REGARDING SEPARATION DISTANCES

Smith *et al.* (2005) performed a PSA regarding the safety distance required between a modular, prismatic HTGR and hydrogen production facility, which consist of either:

- 1. a sulphur-iodine process with sulphuric acid, I2, and HI,
- 2. a hybrid sulphur process with sulphuric acid but no iodine compounds, or
- 3. a high-temperature electrolysis process (no hazardous chemical inventory).

The PSA focused on a few key areas to determine an adequate separation distance between the nuclear and chemical facilities, which include overpressure events and dispersion events as is shown in the following figure (Figure 5-27).

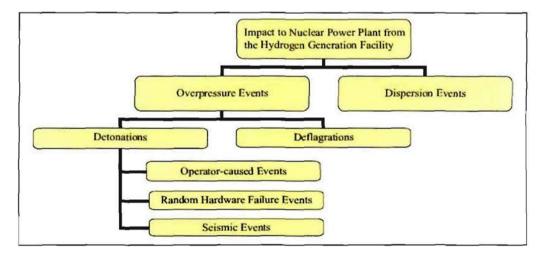


Figure 5-27: Master logic diagram for potential disruption scenarios (Smith et al., 2005)

Smith et al. (2005) did not perform a PSA on the HTGR itself or have access to a fullpower PSA for the specific HTGR under consideration, and therefore used a PSA performed in the mid-1980s by GA Technologies. This PSA evaluated a 558 MW(e) modularized prismatic HTGR, which is similar to the point design being evaluated for the hydrogen-producing very-high-temperature gas reactor (Everline, 1984 and MacDonald et al., 2003 as stated in Smith et al., 2005). The reactor building contains a reactor module embedded in the earth and consists of a concrete enclosure encasing the reactor internals. The reactor building also functions as a filtration system to capture particulates and halogens. However, since this system is not able to withstand accident-loading conditions, the study assumed that a loading in excess of 7 kPa will result in functional failure of portions of the aboveground portion of the reactor building (Smith et al., 2005). Additionally, the study did not consider internal events of the HTGR that may lead to core damage, nor did they analyze risk implications due to the secondary heat exchanger or the choice of the secondary working fluid (internal events). However, interactions between components in the chemical facility that may increase risk to the HTGR are included in the scope of the analysis (Smith et al., 2005).

Furthermore, the study assumed that the largest amount of hydrogen stored at the chemical facility and available to participate in a single detonation event is 100 kg, since it is not expected that extremely large quantities of hydrogen will be stored permanently on site (Smith *et al.*, 2005).

Smith et al. (2005) found that:

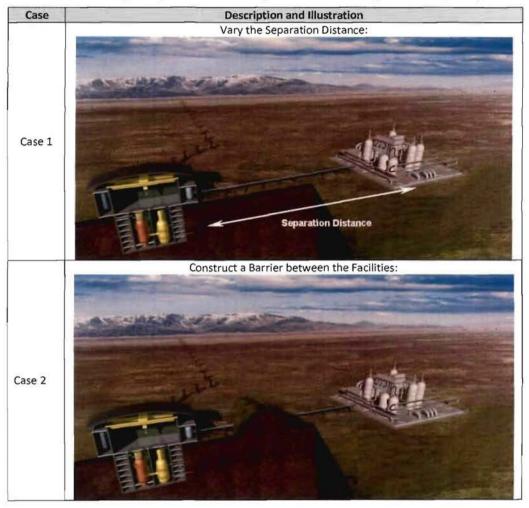
"while the nominal risk analysis results indicate that the CDF (7E-6/yr) is low at a separation distance of 60 m, these results are above the regulatory threshold (1E-6/yr) normally considered by the NRC in such guidance as RG 1.174".

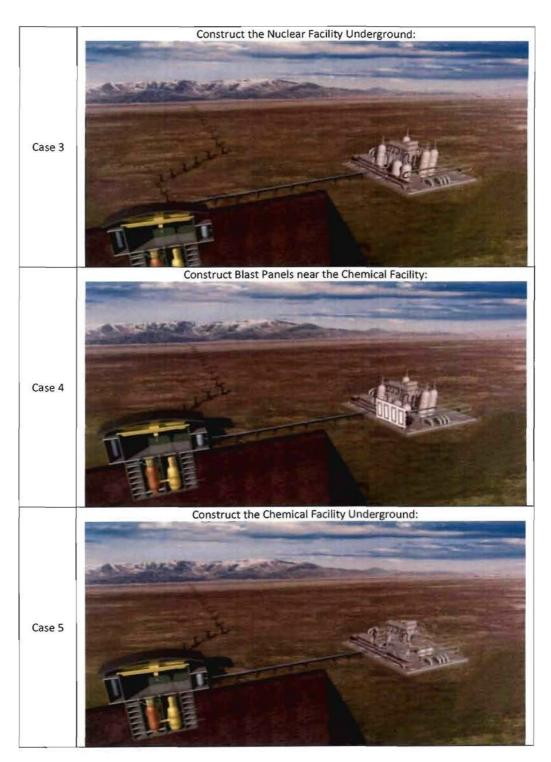
To this extent, the study undertook several sensitivity analyses to help mitigate or in some cases remove risk drivers. The sensitivity analysis evaluated six different situations, which are listed beneath and graphically represented in the subsequent table (Table 5-4; Smith *et al.*, 2005):

1. Varying the separation distance between the two facilities

- 2. Constructing an earthen barrier between the nuclear and chemical facilities
- 3. Constructing the nuclear facility primarily underground
- 4. Constructing blast panels near the chemical facility
- 5. Constructing the chemical facility primarily underground
- 6. Moving the nuclear plant control room offsite.

Table 5-4: Sensitivity Analyses related to separation distances (Smith et al., 2005)







CASE 1: VARY THE SEPARATION DISTANCE

The first sensitivity analysis considers only the variation in the separation distance between the HTGR and chemical facility. The mean frequency of core damage (per year) as a result of the separation distance being varied from 20 to 140 m (in 20 m increments) is shown in the following figure (Figure 5-28). According to Smith *et al.* (2005), hydrogen detonation events dominate at separation distances below 100 m, while other types of core damage scenarios become more likely at distances greater than 100 m (such as a hydrogen detonation leading to a plant upset condition that result in core damage).

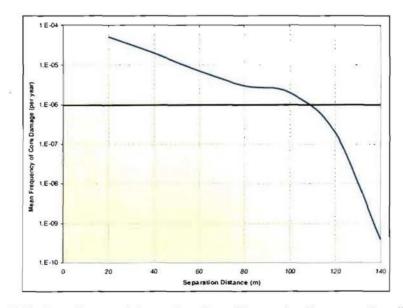


Figure 5-28: Core damage risk as a function of increasing the separation distance between the hydrogen production facility and the nuclear plant (Smith et al., 2005)

CHAPTER 5

CASE 2: CONSTRUCT A BARRIER BETWEEN THE FACILITIES

In Case 2, an earthen barrier is constructed between the chemical and nuclear facilities to mitigate potential detonation events from propagating from the hydrogen facility to affect the nuclear plant. The barrier is at a height approximately equal to that of the nuclear facility and allows the probability of structural damage from hydrogen detonation scenarios to be assumed negligible. However, the height of the barrier is not such that dispersion events can be neglected. Refer to Table 5-4 for a summary of the results (Smith *et al., 2005*).

CASE 3: CONSTRUCT THE NUCLEAR FACILITY UNDERGROUND

In Case 3, the entire reactor confinement is constructed underground to protect it from potential explosion events. However, the conditional core damage probability was not set to zero since damage to the aboveground structures may affect the underground structures or systems. Even though the study did not assume that this damage would lead directly to core damage, there was a probability of seeing core damage because of the hydrogen detonation and subsequent structural damage. To model the event, the probability of structural damage due to explosion events is set to zero, while dispersion events occur without mitigation since the intakes of the control room is still aboveground (Smith *et al., 2005).*

CASE 4: CONSTRUCT BLAST PANELS NEAR THE CHEMICAL FACILITY

In Case 4, passive safety systems, in this case blast panels, are constructed near the chemical facility to mitigate the hazards associated with hydrogen detonation and deflagration events. Again, the probability of structural damage due to explosion events is set to zero, while dispersion events occur without mitigation since the panels do not prevent upsets in the chemical facility. It is also assumed that, either the panels do not become missiles due to the explosion events or that they do not have enough energy to cause structural damage to the nuclear plant when becoming missiles (Smith *et al., 2005*).

CASE 5: CONSTRUCT THE CHEMICAL FACILITY UNDERGROUND

In Case 5, critical portions of the chemical facility is constructed underground to dampen the effects of both explosion and dispersion events as related to the nuclear facility. To model this event, the probability of having a hydrogen explosion is set to zero, which results in the probability of structural damage due to explosion events also being zero. Moreover, since the probability of a hydrogen explosion is zero, the probability of chemicals being released is also zero (Smith *et al., 2005*).

CASE 6: MOVE THE CONTROL ROOM OFFSITE

In Case 6, the control room of the nuclear plant is moved offsite to mitigate the hazards associated with dispersion events. Offsite refers to the control room being situated at a significant distance from the facilities (500 m). This scenario does not mitigate core damage, but protects the control room from dispersion events in order to conform to RG 1.78 regarding habitability (Smith *et al., 2005*).

Case	Desentpillon	Mean ODF per year	Mean Chemical dispersion
Nominal	No mitigation features at a separation distance of 60m	7.00E-06	3.00E-03
Case 1	Vary the separation distance	Refer to Figure 5-28	3.00E-03
Case 2	Construct an earthen mound between facilities	4.00E-10	3.00E-03
Case 3	Construct nuclear facility underground	4.00E-10	3.00E-03
Case 4	Construct blast panels near chemical facility	4.00E-10	3.00E-03
Case 5	Construct chemical facility underground	8.00E-11	8.00E-04
Case 6	Move nuclear plant control room offsite	7.00E-06	No Relevance

It is clear that physical barriers such as the earthen mound, blast panels, and underground placement of critical systems significantly reduce the probability of having core damage due to explosion events. However, these barriers do not reduce the risk related to dispersion events. The only design modification considered in this study that significantly reduces the hazard of dispersion events is that of moving the control room offsite, but this modification in turn does not influence the probability of having core damage. Therefore, a combination of design modifications will most probably be implemented in the next-generation of nuclear plants if hydrogen is to be produced nearby. When focussing only on the separation distance between the two facilities, facilities situated within 100 m of each other will probably face licensing issues as related to RG 1.174 (CDF > 10⁻⁶ per year). In conclusion, Smith et al. (2005) propose that the hydrogen production facility should utilize the electrolysis option since it eliminates the dispersion hazard and corresponding need to relocate the control room offsite. In conclusion, this study considered the safety of the combined complex as related to separation distance requirements; however, it was based on several assumptions and therefore a complete PSA regarding the specific combined complex under consideration is required. A factor of safety evaluations is

the identification of hazards that could affect the health of the operating personnel or public, as well as the frequencies with which they occur to affect health. Since this evaluation will also be site, technological and configuration specific, a PSA regarding the HTTR/SMR system is discussed in the following section as an example.

5.5.6 PSA STUDY ON THE HTTR/SMR COMPLEX

Nelson et al. (2007) performed a PSA study on the HTTR/SMR complex to determine the frequency of an accident at this complex that could affect the population. Based on existing studies, a simplified HAZOP study was performed to identify three main initiating events. The first initiating event is that of a total break in the methane piping in the place that would cause the most damage and with the least possibility of preventing the explosion. A nominal flow of 1400 kg/h of methane leaks from the pipe and it is assumed that the only way to stop the flow in this location is by stopping the methane supply pump. The second initiating event is a worst-case helium duct rupture resulting in a depressurization accident. The HTTR designers consider this accident as the worst event since it would result in the helium not cooling the HTTR core and the only way to remove the heat is by the vessel cooling system (VCS). The third main initiating event considers a break in a heat exchanger pipe inside the primary pressurized water cooler (PPWC) and results in water ingress into the HTTR core. This event could lead to the oxidation of the internal graphite structures and possibly a hydrogen explosion due to hydrogen being produced in the core (Nelson et al., 2007). Table 5-6 summarizes the most significant end states of the initiating events considered for the HTTR/SMR complex that could affect human health.

Zenci State	Frequency
Methane Explosion	6.533E-08
Fission Product (FP) Release	5.996E-09
FP release due to possible structural damage	6.108E-11
Hydrogen Explosion	1.127E-11
FP release due to structural damage	7.100E-13
Hydrogen Explosion resulting in FP release	3.186E-15
Methane Explosion resulting in possible structural damage	1.617E-15
Methane Explosion resulting in thermal damage to chemical plant	8.748E-17
Total	7.140E-08

Table 5-6: PSA results conducted on the HTTR/SMR complex (Nelson et al., 2007)

Nelson *et al.* (2007) found that a methane explosion is the most dominant hazardous event (90%) that could affect the health of the personnel or public, and that the addition of a cut-off valve in the methane line can reduce the explosion frequency by two orders of magnitude. The study also shows that an accident at one of the plants has little effect on the other due to the design base distance between the plants (not stated), the fact that the reactor is underground, as well as other safety characteristics of the nuclear power plant (Nelson *et al.*, 2007).

5.5.7 PLANT PHENOMENA IDENTIFICATION & RANKING TABLES

Forsberg *et al.* (2007) performed a process heat and hydrogen production (PHHP) plant phenomena identification and ranking table (PIRT) to address the safety issues regarding the nuclear reactor due to coupling the nuclear reactor to a chemical plant. According to Forsberg *et al.* (2007), the objectives of the PHHP PIRT are to:

- 1. "identify the safety-relevant phenomena that are introduced by coupling process heat or hydrogen production systems to the NGNP,
- 2. rank the importance of these phenomena in the assessment of the overall safety of the NGNP, and
- 3. assess the knowledge base"

It is important to note that the PHHP PIRT is a nuclear plant PIRT and not a chemical plant PIRT, which means that the chemical and nuclear plant incidents and accidents are evaluated according to their impact on nuclear power plant safety (reactor or operating personnel). Due to differences in the safety philosophies of the plants, the coupled hydrogen or chemical process plant is not treated as an extension of the nuclear plant, but rather as an external facility that can influence nuclear reactor operation. Supplementary to the nuclear plant and chemical facility, the intermediate heat transfer system was also investigated with regard to plant incidents and accidents that could affect the safety of the nuclear plant. The study considered helium as working fluid of the intermediate heat transfer system, although molten salt loops are also under consideration for the NGNP. The design parameters of the VHTR nuclear reactor of the NGNP, as well as the nuclear hydrogen production options considered in the PHHP PIRT are given in the following two tables (Tables 5-7 and 5-8; Forsberg et al., 2007). Please note that since the study was performed on a VHTR of this particular design, some of the results may not be applicable to all Gen-IV HTGR concepts.

Parameter	Value/description		
Reactor Type	Pebble bed or prismatic block		
Reactor Power	500-600 MW(t)		
Primary coolant	Helium		
Inlet temperature	350-490 °C		
Outlet temperature	850-950 ⁰ C		
Coolant pressure	7-9 MPa		
Active core height	10-11 m		
Inner core/reflector diameter	2 m		
Outer core diameter	4 m		
Side reflector outer diameter	6 m		
RPV outer diameter	7 m		
RPV length	30 m		

Table 5-8: Nuclear hydrogen production options (Forsberg et al., 2007)

e Process	Primeny nuclear plant sec. moutes: 23	k Chemical≻ plant : inputs		Chemistry (chemicals) ininventory)
Steam reforming of natural gas	Heat	Natural gas and H₂O	H ₂ , CO ₂	$CH_4+H_2O \rightarrow CO+3H_2$
Electrolysis	Electricity	H2O	H2. O2	$2H_2O$ (water) $\rightarrow 2H_2+O_2$
High- temperature Electrolysis	Heat, steam and electricity	Steam	H ₂ . O ₂	$2H_2O$ (steam) $\rightarrow 2H_2+O_2$
Hybrid-sulphur cycle	Heat and electricity	H₂O	H ₂ . O ₂	$2H_2SO_4 \rightarrow 2SO_2+2H_2O+O_2 \text{ (heat)}$ $2H_2O+SO_2 \rightarrow H_2+H_2SO_4 \text{ (electricity)}$
Sulphur-iodine	Heat	H₂O	H ₂ . O ₂	$\begin{array}{l} 2H_2SO_4 \rightarrow 2SO_2+2H_2O+O_2 \ (heat) \\ 2HI \rightarrow I_2+H_2 \ (heat) \\ I_2+SO_2+2H_2O \rightarrow 2HI+H_2SO_4 \ (heat) \end{array}$

The study identified four main classes of accident scenarios, each consisting of several subcategory scenarios, which are (Forsberg *et al.*, 2007):

- 1. Chemical plant releases: Steady-state and accidental releases of hydrogen, oxygen, flammables, corrosives, toxic gases, and asphyxiates from the chemical plant.
- 2. Process thermal events: Heat transfer transients due to variation in the heat demand from the chemical plant.
- 3. Heat transport system failures: Accident scenarios due to failures in the intermediate heat transfer loop.

4. VHTR upsets: Accidents initiated in the nuclear reactor that progress to the chemical plant with subsequent feedback events to the reactor creating the potential for a radiological release.

As quoted from Forsberg *et al.* (2007), the PHHP PIRT was developed using the following strategy:

- "The types of accident events were identified and the qualitative result or direct consequence of that event was estimated. The question being addressed was what kind of challenge to the NGNP could this event cause?
- The next step was to examine the phenomena that controlled the severity of the potential insult or impact on NGNP. The characteristics of released materials, conditions associated with the release, magnitude of the thermal event, and potential timing all were considered in defining the magnitude of the potential threat to NGNP.
- The final step was to evaluate the potential impact on the NGNP resulting from that event. The impacts on the NGNP were generally categorized as effects on safety related plant equipment or systems, or the impact on people, workers, workers with safety related functions, or the public".

The importance of the phenomena and the status of the knowledge base were based on the criteria and considerations listed in the following two tables (Tables 5-9 and 5-10; Forsberg *et al.*, 2007).

Importance	Criteria
High (H)	Material release or thermal event could potentially affect the likelihood or severity of core damage
Medium (M)	Event impacts operations or contribute to other safety-related events, but not strongly impact the severity of an accident
Low (L)	Event primarily impacts operations, but have limited effects on the reactor or workers

Table 5-9: Ranking criteria	a of importance of any phenomena	(Forsberg <i>et al.</i> , 2007)
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Table 5-10: Ra	ting criteria of kno	wledge base (F	orsberg e <i>t al.</i> , 2007)

Rating	Criteria
High (H)	Tools and data base are considered adequate and available now
Medium (M)	Either the tools or the database are considered incomplete in some area
Low (L)	Tools and/or data base are missing and require significant R&D

Each of the accidents is discussed according to possible impact to the safety-related reactor plant SSCs (systems, structures and components) and/or operator injury or impairment.

5.5.7.1 CHEMICAL RELEASES

Chemical releases occur during normal and accident conditions at a chemical plant and could impact the nuclear plant by shock waves and heat radiation during fires and explosions, as well as by migration of hazardous components to the nuclear plant. The latter has historically had the most catastrophic off-site consequences since heavier-than-air clouds can travel significant distances. The potential of chemicals to be transported beyond the perimeter of the chemical facility depend on the particular chemical, the inventory and temperature and pressure before the accident. The general observations regarding chemical releases from the chemical facility are (Forsberg *et al.*, 2007):

- Plant layout: Separation distances between process units, storage tanks located away from the process units and employment of physical barriers around storage tanks. The layout of the combined complex is considered the most important single safety system to assure chemical plant releases do not impact nuclear plant safety.
- Inventory: Consequences or extents of hazards depend on the total chemical inventories that can participate in the accident.
- Inherent safety: Employing appropriate operating conditions to minimize the occurrence of accidents or reduce their consequences.

Chemical releases include releases of hydrogen, oxygen, flammables, corrosives, toxic gases, and asphyxiates gases with regard to possible consequences to the nuclear plant.

HYDROGEN RELEASES

Hydrogen releases could affect the nuclear plant by damage, wear or impairment of the safety-related SSCs due to shock waves and heat radiation, as well as operator impairment or injury due to burns. The PHHP PIRT panel found that shock waves are of intermediate importance while heat radiation and operator burns are of low importance. All panel members ranked the knowledge base as high due to the ,

widespread use and production of hydrogen in the chemical industry (Forsberg *et al.*, 2007). Refer to Table 5-11 for the PIRT evaluation.

	Evenť	Evaluation criterion	Issue (phenomena, process, etc.)	Importance ¹	Knowleds base ¹
Chemical releases	H ₂ release	Damage of SSCs	Blasé effects Hear flux	M L	H M
		Operator impairment	Burn and hear flux to people (VHTR. operators)	L	м
	O ₂ release	Damage of SSCs	Plume behavior Allowable	н н	H M
			concentrations Spontaneous combustion	н	м
		Operator impairment	Burn to VHIR. operators	м	м
	Flammable release	Damage of SSCs	Plume behavior Heat flux	M M	ਸ ਸ
		L	Blast effects	<u>M</u>	H
	Corrosive release	Damage of SSCs	Burns to people Plume behavior	M	H H
`	Corrosive release	Damage of SSUS	Allowable concentrations	M	M
		Operator impairment	Burns to people	M	M
	Toxic gas release	Operator impairment	Plume behavior Toxic concentrations and effects	M M	M M
	Suffocation gas release	Damage of SSCs	Plume behavior Backup power/O ₂	M M	H H
		Operator impairment	concentrations Concentration for	M	н
Process thermal	Loss of heat load	Damage of SSCs	people Loss of hear sink to reactor	M	м
events	Temperature transient	Damage of SSCs	Cyclic loading Earmonics	M L	M M
Hear transport system faihures	IEX failure (intermediate heat exchanger)	Damage of SSCs	Blowdown effects, large mass transfer, pressurization of either secondary or primary side	н	М
	PHX failure (process hear exchanger)	Damage of SSCs	Fuel and primary system corrosion	н	М
	Mass addition to reactor (He)	Damage of SSCs	Turbomachinery response; potential for No He mixture	м	м
	Mass addition to reactor	Damage of SSCs	Reactivity spike due to neutron thermalization.	н	м
	(hydrogeneous material, e.g., steam)		Chemical attack of TRISO layers and graphite	н	м
	Loss of intermediate fluid	Damage of SSCs	Loss of heat sink; cooling, then no heat sink; HX hydrodynamic loading	H	м
VHIR events that impact chemical planr	Anticipated operations: unitum	Dose to VHIR plant workers.	Diffusion of ³ H	L	н
	term safety)	Dose to process 335 users: industrial and consumers.	Diffusion of ³ H	L	н
	Radiologic release pathways through HX loops and plant	Dose to public	Accident radionachde release	M	м
	Generic power or hermal transients initiated in VHTR.	SSC, suess on IHX or other component in contact with BOP	Stress causes stress on other SSC component	L	м

OXYGEN RELEASES

Since there is no regulatory standard for the release of oxygen it may be continuously released to the atmosphere, therefore both accident releases and normal releases must be considered. Oxygen releases affect the nuclear plant by damage, wear or impairment of the safety-related SSCs due to oxygen causing a fire or oxygen degrading equipment over time, as well as operator impairment or injury due to bums. The PHHP PIRT panel found that oxygen releases are of high importance due to the following reasons (Forsberg *et al.*, 2007):

- the possibility of plume formation if oxygen is released from a high-pressure containment
- the possibility of "spontaneous combustion" of many materials in the presence of pure oxygen becomes more likely
- materials that are usually considered as non-combustible can burn in an oxygen atmosphere if ignited
- oxygen can be considered a pollutant if the concentrations are significantly higher than normal atmospheric levels
- oxygen is able to permeate into many materials resulting in a change of some of their properties as well as characteristics during a fire
- oxygen accelerates the degradation of most materials, which is a specific concern regarding long-term oxygen exposure and exposure to ultrahigh oxygen concentrations during an accident

The panel found that the importance of operator impairment or injury due to burns is of medium concern, while the knowledge base of oxygen plume formation, allowable and ultrahigh concentrations are high, medium and medium respectively (Forsberg *et al.*, 2007). Refer to Table 5-11 for the PIRT evaluation.

FLAMMABLE RELEAES

Several flammable substances may be present at the chemical facility if the SMR process is employed or if the nuclear reactor nuclear reactor supplies heat to oil refineries, shale oil and tar sands production facilities, or coal gasification and liquefaction processes. Similar to hydrogen releases, the potential risks to the nuclear facility include damage, wear or impairment of the safety-related SSCs due to shock waves and heat radiation, as well as operator impairment or injury due to burns. In contrast to hydrogen, many flammable releases may form heavier-than-air

plumes that can travel considerable distances to impact the nuclear facility. Moreover, the heat radiated from a hydrocarbon fire is also significantly higher than that of hydrogen fires. Therefore, the inherent risk of flammables releases is considered more pronounced that that of hydrogen (Forsberg et al., 2007). Refer to Table 5-11 for the PIRT evaluation.

CORROSIVE RELEASES

Thermochemical water splitting cycles involve the use of several corrosive substances to produce hydrogen. Where the hybrid sulphur only has sulphuric acid as corrosive substance, the iodine sulphur cycle has sulphuric acid, iodine, hydrogen iodide and hydriodic acid, and the calcium bromide cycle has bromine and hydrogen bromide as corrosive substances. Therefore, the release of corrosive substances is significantly possibly when these cycles are employed to produce hydrogen. Corrosive releases can impact the nuclear facility by damage, wear or impairment of the safety-related SSCs due to material corrosion, as well as operator impairment or injury due to burns. Accident phenomena associated with corrosive releases such as sulphuric acid and hydrogen iodide) and corrosion of material over time. However, it was found that since the material corrosion rate is very low, appropriate action could be taken to avoid serious consequences (Forsberg *et al.*, 2007). Refer to Table 5-11 for the PIRT evaluation.

TOXIC GAS RELEASES

Many corrosive substances are toxic, but usually the corrosive hazard dominates the toxic hazard. Where corrosive releases affect both equipment and personnel, toxic releases are only hazardous to the personnel. Therefore, as related to the criteria of affecting the safety of the nuclear plant, only operator impairment by poisoning (inhalation or sorption through the skin) is considered for toxic gas releases. Some of the toxic gases present in the thermochemical cycles are iodine, hydrogen iodide, bromine and hydrogen bromide. The accident phenomena associated with toxic gas releases are plume behaviour and allowable concentrations of toxic gases. Similar to many of the previous releases, toxic gas releases can form heavier-than-air plumes that can travel considerable distances to affect the nuclear plant. However, due to the operators being able to identify the risk at an early stage, they can take action to reduce the consequences of this hazard (Forsberg *et al.*, 2007). Refer to Table 5-11 for the PIRT evaluation.

ASPHYXIATE GAS RELEASES

Thermochemical production of hydrogen by nuclear assisted technologies may contain asphyxiate gases such as helium, nitrogen and carbon dioxide, which can result in impairment of operators (asphyxiation) and certain equipment (diesel generators) by displacing the oxygen in air. Therefore, as related to the criteria of affecting the safety of the nuclear plant, asphyxiate gas releases can impact the nuclear facility by damage, wear or impairment of the safety-related SSCs due to oxygen displacement, as well as operator impairment or injury due to asphyxiation. Accident phenomena associated with asphyxiate gas releases are plume behaviour, oxygen concentrations needed to operate backup power systems and asphyxiation of the operators (Forsberg *et al.*, 2007). Refer to Table 5-11 for the PIRT evaluation.

5.5.7.2 PROCESS THERMAL EVENTS

Process thermal events are based on the endothermal nature of the chemical reactors, which remove heat from the heat transfer system due to the conversion process. Therefore, transients and failures in the chemical systems result in thermal turbulences due to inconsistencies in the heat removal rate, which in turn result in increased heat being "recycled" back to the nuclear reactor. However, the nuclear reactor thermal transient is mitigated by the thermal mass of the intermediate coolant cycles, the piping and materials of the heat exchangers, as well as the core internal structures. In HTGRs, the graphite internal structures are a very significant heat storage medium that dampens transients drastically. Additionally, molten salt loops can be employed to mitigate this hazard due to their high heat capacities and low operating pressures. To this extent, the heat capacities of candidate molten salts proposed for the NGNP nuclear-hydrogen initiative (NHI) are given in the following table (Table 5-12).

Salt constituents	Malan assumption	Heat capacity (cal/g-°C)		
San constituents	Molar composition	Measured "	Predicted	
LiF-NaF-KF	(46.5-11.5-42)	0.48	0.387	
NaF-ZrF₄	(59.5-40.5)	0.28	0.275	
KF-ZrF4	(58-42)		0.251	
LiF-NaF-ZrF4	(26-37-37)		0.296	
LiC1-KCl	(59-41)	0.287	0.289	
LiCl-RbCl	(58-42)	0.213	0.212	
NaCl-MgCl ₂	(58-42)	0,258	0.262	
KCl-MgCl ₂	(67-33)	0.276	0.229	
NaF-NaBF4	(8-92)	0.36	0.435	
KF-KBF,	(25-75)	0.312	0.367	
RbF-RbBF4	(31-69)	0.218	0.258	

Table 5-12: Heat capacities of candidate molten salts (Williams, 2006)

Helium has an even higher specific heat capacity (≈ 1.24 cal/g.K at 70 bar, 900 °C) but requires high operating pressures, which result in guick transfer of the thermal turbulences to the nuclear reactor (high operating pressure with associated high velocities). The high operating pressure of helium is due to its low density ($\approx 2.854 \text{E}^{-3}$ g/cm³ at 70 bar, 900 ^oC), therefore a more relevant heat transfer property in this case would be the volumetric heat capacity (ρ^*Cp), which is 3.542 E⁻³ cal/cm³.K for helium (70 bar, 900 °C) and significantly less than that of the molten salts. The higher helium operating pressures (velocities) and larger helium volumes in the heat transfer loops result in large heat transfer loops with high pumping requirements. However, the molten salts are extremely corrosive and require robust heat transfer materials. Moreover, the lower operating pressures of the molten salt coolant loops allows for a large pressure drop over the IHX (from primary to secondary loop) and increases concerns regarding the material requirements and integrity of the IHX, especially its durability considering the extended operating lifetimes, high operating conditions and corrosive environments. The following table (Table 5-13) gives a summary of the properties of candidate molten salt coolants for the NGNP/NHI heat-transfer loop (Williams, 2006).

				Heat-transfer properties at 700°C				
Salt "	Formula weight (g/mol)	Melting point (°C)	900°C vapor pressure (mm Hg)	ρ, density (g/cm ³)	<i>p*Cp</i> , volumetric heat capacity (cal/cm³-°C)	μ, viscosity (cP)	k, thermal conductivity (W/m-K)	
LiF-NaF-KF	41.3	454	~ 0.7	2.02	0.91	2,9	0.92	
NaF-ZrF4	92.7	500	5	3.14	0.88	5.1	0.49	
KF-ZrF4	103.9	390	1.2	2,80	0.70	< 5,1	0.45	
LiF-NaF-ZrF4	84.2	436	~ 5	2.92	0.86	6.9	0,53	
LiCl-KCl	55.5	355	5.8	1.52	0_435	1.15	0.42	
LiCl-RbCl	75.4	313		1.88	0.40	1.30	0.36	
NaCl-MgCl ₂	73.7	445	< 2.5	1.68	0.44	1.36	0.50	
KCl-MgCl2	81.4	426	< 2.0	1,66	0.46	1.40	0.40	
NaF-NaBF4	104.4	385	9500	1.75	0.63	0.90	0.40	
KF-KBF4	109.0	460	100	1.70	0_53	0.90	0,38	
RbF-RbF₄	151.3	442	< 100	2.21	0.48	0.90	0.28	

	Table 5-13: Summar	y of the p	properties of	candidate molten	salts	(Williams,	2006)
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Forsberg et al. (2007) evaluated two broad classes of process thermal events; these are loss of heat load and temperature transients.

LOSS OF HEAT LOAD

This class of events includes the particular failures and transients in the chemical plant that reduce heat consumption resulting in diminished heat removal from the intermediate loop. Since the chemical reactions are endothermal, if the chemical feeds to these reactors decrease, the chemical reactors use less heat and return the intermediate heat transfer fluid at higher temperatures (Forsberg *et al.*, 2007). The worst case scenario associated with this event is that of a guillotine break in one of the chemical feeds resulting in no heat removal from the intermediate circuit. However, with regard to impacting the safety of the nuclear plant, the importance of this event was judged to be moderate due to the thermal and physical separation of the plants, as well as that this event would undoubtedly be one of the reactor design criteria (Forsberg *et al.*, 2007). Refer to Table 5-11 for the PIRT evaluation.

TEMPERATURE TRANSIENTS

This event is based on the relative complexity of the thermochemical hydrogen production processes, which contain multiple reactors and other process units than can initiate or amplify thermal transients. Moreover, maladjustment of the process control software in the process heat system can also induce or amplify thermal transients. Therefore, as related to the impact on nuclear plant safety, temperature transients have the potential for damage, wear, or impairment of the reactor SSCs. Due to the relatively small magnitude of this event, the damage to SSCs may take place over a long term and may not be immediately evident. The importance of this event was considered to be moderate due to the hydrogen production plant utilizing only a small portion (10% for the NGNP) of the heat supplied to the cogeneration plant, the heat load being well separated from the nuclear reactor and the relative small damage to reactor SSCs this event could be responsible for (Forsberg *et al.*, 2007). Refer to Table 5-11 for the PIRT evaluation.

5.5.7.3 HEAT TRANSFER SYSTEM FAILURE

Failures in the heat transfer system are considered of high importance by the PHHP PIRT panel since it may lead to thermal upsets and mass additions or losses between the nuclear and chemical plants. The heat transfer system includes the IHX that transfers heat from the primary to the intermediate heat loop, and the process heat exchanger (PHX) that transfers heat from the intermediate heat loop to the chemical process. Since it is a nuclear plant PIRT, the events considered as heat transfer system failures are IHX failure, PHX failure, mass addition to the nuclear reactor (helium or hydrogenous material) and loss of intermediate fluid (Forsberg *et al.*, 2007).

IHX FAILURE

The IHX is considered a critical component of process heat and cogeneration nuclear technologies and therefore necessitates thorough investigation. The IHX forms the boundary between the nuclear plant and the intermediate heat transfer loops such that any failure of the IHX will affect the integrity of the primary system. Implications to the IHX due to pressure or temperature gradients may affect both near-term and long-term safety of the nuclear plant. Consequences as a result of IHX failure will depend on the construction, configuration and operating conditions of the specific system under consideration. Potential consequences of IHX failure include (Forsberg *et al.*, 2007):

- Rapid blow-down of the primary system (and subsequent loss of active cooling ability of the reactor core)
- Pressurization of the primary or secondary heat transfer loop
- Pressurization of the nuclear reactor containment building (IHX failure inside the reactor building)
- Blow-down of the secondary heat transfer loop (resulting in thermal turbulence)
- Slower thermal or pressure events
- Radionuclide transport into the reactor building, intermediate heat transfer loop or the environment depending on the blow-down direction and the amount and location(s) of failures

The importance of these events was rated as high by the PIRT panel since they may directly affect the core cooling ability, the integrity of the primary system or radionuclide transport during an accident (Forsberg *et al.*, 2007). Refer to Table 5-11 for PIRT results.

PHX FAILURE

The PHX is the interface between the intermediate heat transfer loop and the chemical process, but could also be the interface between the primary system and the chemical or electrochemical process in a direct-cycle design. Potential consequences of PHX failure include (Forsberg *et al*, 2007):

- Failures in the process plant resulting in loss of process fluids and therefore thermal turbulences (and possibly the release of hazardous material at the chemical facility)
- Transport of process gases to the intermediate heat transfer loop (primary system in a direct-cycle design)
- Transport of intermediate (or primary) heat transfer fluid (in this case helium) to the chemical process
- Loss of intermediate loop working fluid (resulting in no heat removal from the primary system and no heat transport to the endothermal chemical reactors with the corresponding loss of conversion)
- Increased rate of corrosion of the IHX components if corrosive substances from the chemical process were to enter the intermediate loop
- Other mass transfer, thermal and pressure events

The importance of these events was rated as high by the PIRT panel due to the potential impacts to the integrity of the primary system and the IHX, as well as rapid thermal and pressure transients (Forsberg *et al.*, 2007). Refer to Table 5-11 for PIRT results.

MASS ADDITION TO REACTOR (HELIUM)

The mass addition of helium (the intermediate loop working fluid) to the reactor is possible if the IHX is designed such that the pressure of the intermediate loop is higher than that of the primary loop, where a failure in the IHX leads to depressurization of the intermediate loop into the primary loop. The inventories of both loops and their respective operating conditions (pressure, temperature) will determine the impact of this hazard on the safety of the nuclear plant. Possible consequences of this event are pressurization of the primary system resulting in temperature transients and pressure effects, as well as response of turbo machinery associated with circulators or electrical generation. Furthermore, if the intermediate loop working fluid differences in heat transfer coefficients need to be considered. In general, mass additions to the nuclear reactor could lead to rapid thermal and pressure transients depending on the particular substance and quantity thereof added to the reactor. The importance of helium mass addition events was rated as moderate by the PIRT panel due to helium's small reactivity potential and limited

threat to the primary system (Forsberg *et al.*, 2007). Refer to Table 5-11 for PIRT results.

MASS ADDITION TO REACTOR (HYDROGENOUS MATERIAL)

The mass addition of hydrogenous material into the nuclear reactor is possible if the steam generator (or steam reformer) is located within the primary system (no intermediate heat transfer loop) and it fails such that steam (or reformer gases) is able to enters the primary system. However, most concepts employ an intermediate loop where the consequences of steam generator failure are less significant with regard to the safety of the nuclear plant. The consequences of hydrogenous mass addition to the primary system are reactivity events, chemical attack on graphite and fuel materials and the possibility of forming dangerous hydrogen concentrations within the primary system. The magnitude of these consequences will depend on the amount of hydrogenous material entering the primary system and the specific reactor and steam generator operating conditions under consideration. The importance of hydrogenous mass addition events are considered as high due to the potential increase in reactivity in the core, with the subsequent power increase that could lead to a more serious thermal event. The corrosive effects were considered as long-term events if small amounts of steam leak into the primary system, but could potentially be very significant under accident conditions when large additions at elevated temperatures occur (Forsberg et al., 2007). Refer to Table 5-11 for PIRT results.

LOSS OF INTERMEDIATE FLUID

This event considers a failure of the piping or structures of the intermediate heat transfer loop resulting in the loss of intermediate working fluid. The intermediate heat transfer loop forms the connection between the nuclear plant and chemical facility and may be of considerable size depending on the distance between the facilities. According to Forsberg et al. (2007), the intermediate loop is one of the most exposed components in the entire nuclear/chemical complex and its probability to fail is high considering the operating lifetime of the plant. Consequences of failures in the heat transport system include the following (Forsberg *et al.*, 2007):

- mass additions, blow-downs or pressurization of systems, containment or confinement depending on the location of the failure and the propagation direction of the failure
- loss of heat sink or thermal transients depending on the failure mode and magnitude

- potentially severe pressure and temperature transients in the IHX that could result in its failure, which in turn could pose a threat to the primary system (see IHX failure for other possible consequences)
- pressure and temperature transients in the PHX that could result in its failure and resultant in the release of hazardous substances or the other events described in PHX failure

The importance of the loss of intermediate fluid events is considered as high due to the potential for affecting the IHX and primary system by pressure and thermal transients (Forsberg *et al.*, 2007). Refer to Table 5-11 for PIRT results.

5.5.7.4 VHTR UPSETS

The nuclear reactor, in this case a VHTR, can initiate several events that could influence the safety and operability of the chemical process to which it is coupled to. Considering that this is a nuclear plant PIRT, the pertinent parameter of these events is whether feedback from the chemical plant (due to these events) occurs to affect the safety of the nuclear plant. The events considered are tritium, fission products and (nuclear) reactor temperature transients (Forsberg *et al.*, 2007).

TRITIUM

Tritium production, transport and contamination of the products of chemical process have already been discussed in this chapter, however, feedback from the chemical facility due to tritium contamination has not. To this extent, the PIRT panel rated the importance of tritium regarding the safety of the nuclear plant, nuclear plant personnel and public as low. However, they found that the issues related to tritium are of high importance in terms of public acceptance, which makes it a sensitive political issue but not a safety issue (Forsberg *et al.*, 2007).

Regarding tritium regulations and the probability of nuclear hydrogen technologies adhering to these regulations, Forsberg *et al.* (2007) states that:

"There are no established regulations for the tritium content of nuclearproduced hydrogen, but the Japanese have adopted a target value of 11.8 $Bq/g H_2$, resulting in a hydrogen user dose of 0.1 mSv/year" and that "This limit can be met if the permeation rate from the reactor to the hydrogen product is <0.06%, which is well within present fuel and heat exchanger technology".

The dose limit of 0.1 mSv/year is approximately the same as that obtained by cooking on a natural gas stove. However, the sources of tritium are restricted to that released from the damaged fuel particles (currently a less than 10⁻⁵ occurrence at end of life) and due to the neutron bombardment of the helium coolant. Therefore, it does not include tritium produced by activation of lithium and beryllium impurities in the fuel outer layers or core graphite components. However, these sources contribute only a fraction of the total tritium produced in the core and could be lowéred by several techniques (Forsberg *et al.*, 2007). Refer to Table 5-11 for PIRT results.

FISSION PRODUCTS

Similar to tritium, other fission products can end up in the primary system by diffusing through a "hot spot" in the fuel elements or due to damage to the coated fuel elements. Silver is of particular concern for the diffusion event. When in the primary system, the fission products are transported by the coolant throughout the primary cycle where they tend to plate out on cooler parts of the primary coolant system. However, the noble gases remain mixed in the helium coolant system. According to Forsberg *et al.* (2007), with the exception of tritium, the other fission products are unlikely to be transported to the chemical process since their diffusion through metal heat exchangers is essentially nonexistent. The impact of fission products are released to the environment. Accordingly, the importance of fission products are vith respect to the chemical systems is considered moderate since multiple barriers that must fail before fission products can migrate into the intermediate loop and to the chemical plant (Forsberg *et al.*, 2007). Refer to Table 5-11 for PIRT results.

REACTOR TEMPERATURE TRANSIENTS

Reactor temperature transients are induced by unplanned movement of control rods, which influences the reactivity and power generation in the core, or by changes in the coolant flow rate. These events result in thermal turbulence in the heat transferred to the chemical process, which can increase thermal stresses on the materials of the heat transfer surfaces and present challenges to the control of the chemical process. Since the heat exchangers are barriers to the transport of tritium and other fission products to the chemical process, failures thereof may result in radioactive

contamination of the chemical products. In this regard, nuclear reactor temperature transients could result in repeated thermal stresses that will lead to fatigue cracking of the materials subjected to the transients. Moreover, well-designed control algorithms in both plants could mitigate the effects of temperature transients. The PIRT panel considers the impact of reactor temperature transients of low importance even though it presents some challenges (Forsberg *et al.*, 2007). Refer to Table 5-11 for PIRT results.

5.5.7.5 CONCLUSIONS

Forsberg et al. (2007) concluded that:

- The accidental release of hydrogen does not pose a significant hazard to the nuclear plant if a safety distance is employed
- The accidental release of heavier-than-air components such as oxygen, corrosive gases and toxic gases pose a more significant hazard to the nuclear plant due to "heavy" plumes being able to travel considerable distances
- The release of oxygen is of special concern due to its unique capabilities to generate fires
- Heat exchanger failures are of major concern since it may result in many hazardous scenarios that could affect the nuclear plant including:
 - Blow-down of the intermediate heat transfer loop
 - Leaks into the reactor primary system
 - Chemical additions to the reactor core
 - Hot fluids escaping into the reactor containment building

Refer to the preceding sections for thorough descriptions and evaluations of these hazards, specifically with regard to the safety of the nuclear plant. Even though this was a nuclear plant PIRT, it serves as an identification of hazards that are possible in the nuclear/chemical complex. However, this is not a complete identification of hazards since many additional hazards may arise that are dependent on the particular chemical process and layout of the combined complex under consideration.

5.5.8 MATERIAL CONSIDERATIONS

Development of materials that could be used in the nuclear-hydrogen technologies is of great concern since the proposed operating conditions are at the limit of current engineering technologies (Forsberg, 2003; Forsberg *et al.*, 2007). Considering the long operational lifetime of the components in the combined complex and the additional thermal turbulences induced by coupling the facilities, it may well be beyond the current engineering limits (GAO, 2006; GAO, 2007). Two of the most important issues regarding the materials to be used at the complex are probably duration of the materials and the hydrogen permeation rate through the material. Figure 5-29 shows the time required to rupture at different temperatures and stresses (Verfondern, 2007).

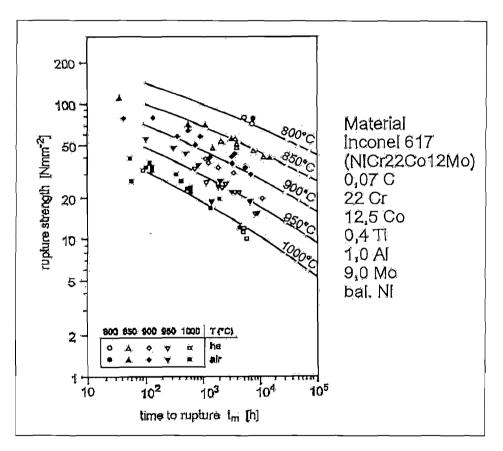


Figure 5-29: Material diagram (FZJ as illustrated in Verfondern, 2007)

The next aspect to consider as that of the hydrogen permeation rate through the material, which is shown in the following figure (Figure 5-30) for transport through Hastelloy XR.

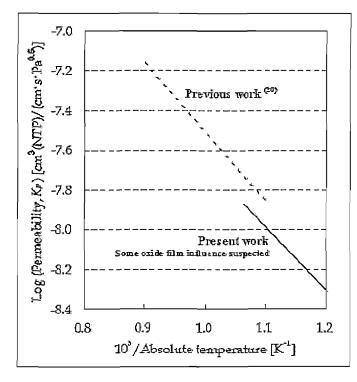


Figure 5-30: Comparison of present and previous work of hydrogen permeation in Hastelloy XR (Sakaba, 2005 and Takeda, 1999 as illustrated in Verfondern, 2007)

The discrepancy in the permeability of hydrogen through Hastelloy XR as related to the "present work" and "previous work" indicated in the figure is presumably due to formation of an oxide layer on the IHX tube surfaces (Sakaba, 2005 as cited in Verfondern, 2007). According to Forsberg *et al.* (2007), the current fuel and heat exchanger designs sufficiently retain tritium within the fuel or primary system. However, if more severe limits regarding tritium concentration in the products are enforced by the regulating authorities, materials with lower hydrogen permeation rates may need to be developed.

5.6 REMARKS REGARDING THE LAYOUT OF THE COMBINED COMPLEX

It will not be meaningful to give specifics regarding the layout of the combined nuclear/chemical complex since the precise design parameters regarding the nuclear plant and chemical production facility are unknown (for the purposes of the study). However, certain aspects regarding the layout of the complex have come to light during evaluation of the safety aspects of the complex. Regarding the HTGR nuclear

plant, it is highly probable that the nuclear reactor and containment building will be situated primarily underground. Even though the outer concrete structure (2 m thickness) is supposed to be able to withstand the impacts of external events, it is highly improbable that the design of this structure considered external events of the magnitude arising from a dedicated process heat application facility, especially considering that the safety distance between the two critical facilities are likely to be reduced. The current regulations regarding separation distances are extreme and need to be augmented if technically (high-temperature heat) and economically feasible operation of the process heat plant is to be achieved. To this extent, several design alterations that are able to reduce the risks of core damage and chemical dispersion accidents such that safe operation of the nuclear plant is possible, are examined in Section 5.5.5.1. Even though the study indicated that a separation distance of greater than 110 m is sufficient to reduce the consequences of chemical plant events, the applicability of that separation distance to any other proposed nuclear/chemical complex is suspect due to different nuclear and chemical plant design parameters, process operating conditions and scale of operations. A multiple barrier system is therefore proposed consisting of an underground nuclear facility, employment of an earthen mound between the nuclear plant and chemical facility in addition to an appropriate separation distance. The precise value of the separation distance will depend on the specific nuclear/chemical complex under consideration, but by employing a multiple barrier system the separation distance should not differ significantly from 110 m. Additionally, the use of an IHX to isolate the nuclear and chemical systems from another essentially is mandatory and any concept that does not include it will be subject to severe scrutinizing by both the regulatory authorities and public, especially if the product is to be used by the general public (consumers of hydrogen in the so-called hydrogen economy). In contrast to some concepts, the IHX should be housed within the outer concrete structure of the nuclear plant to protect it from the impacts external events and to locate it as close as possible to the nuclear reactor.

The choice of process heat application, in this case hydrogen production, will obviously affect the safe and optimal layout of the combined complex since each option has different hazards depending on the operating conditions and chemical inventories of the heat application facility. The hydrogen production facility is specifically referred to as a process heat application facility because electrochemical technologies are not required to be in close proximity to the nuclear plant and will in all likelihood be significantly separated from the nuclear island. Moreover, the main hydrogen production options considered in this investigation are SMR, POX and HyS, which are thermochemical and hybrid thermochemical technologies that require high-temperature process heat and need to be as close as possible to the nuclear plant to reduce thermal losses incurred during transport of heat to the chemical plant. The SMR and POX processes have the added disadvantages of using a flammable. heavier-than-air raw material (methane or natural gas), which is additionally required to be stored onsite in significant quantities if plant upsets are to be reduced. While the PHHP PIRT study identified all heavier-than-air substances as significant hazards to the nuclear plant, the PSA study on the HTTR/SMR project considers a methane explosion as the hazard with the greatest probability to result in deaths of humans or affect human health. Additionally, due to methane and natural gas being flammable and detonable, they are required to adhere to regulations regarding separation distance to the nuclear plant. In this regard, the presence of carbon monoxide in the process streams (or as constituent of the product synthesis gas) of the SMR and POX technologies is also subject to separation distance regulations due to its flammable and detonable characteristics. In addition, the presence of oxygen as input to the POX process or as by-product of the HyS cycle is of serious concern due to oxygen's innate ability to induce and promote fires and detonations, as well as increasing the consequences thereof. In this context, the POX process does not specifically require the onsite storage of oxygen (it can be separated from air as part of the process), while the oxygen produced by the HyS cycle is not required to be stored onsite (it is released into the environment if not specifically produced as commercial product). However, both technologies have oxygen present at some stages of production, which is of concern even if the quantities thereof are significantly less than when it is stored onsite as by-product or input material. Therefore, it can be concluded that the storage of oxygen and flammable substances should be at the outer perimeters of the plant such that they are as far away from critical systems (such as the nuclear reactor and important chemical process stages) as practically achievable. In addition, oxygen should not be stored close to that of the flammable components such as hydrogen, methane, natural gas or synthesis gas in order to reduce the flammable and detonation hazards associated with their storage.

The use of other hazardous chemical components (H_2SO_4 , SO_2 , SO_3 , I_2 , HI and hydriodic acid) in the HyS and I-S cycles are of concern considering their heavier-air-air nature resulting in risks to the nuclear reactor SSCs, operating personnel and visitors to the site. Fortunately, their quantities stored onsite will be minimal and should not significantly influence the layout of the plant. Moreover, their quantities

utilized in the production cycles will be according to the economical and technical specifications of the process, which will most probably be as low as practically achievable. However, the presence of these components will be an issue when the habitability of the nuclear plant control room is under consideration.

5.7 CONCLUDING REMARKS

It is clear that significant research and development is still required for successful implementation of the combined nuclear/chemical complex. Especially, materials development and the design and construction of the IHX and PHX are of significant concern. However, none of the issues regarding the safety of the combined complex or the technological constraints discussed in this chapter is such that appropriate research and development will be unable to address them, but the time required therefore may be of concern. Regardless of the safety, technological and economical feasibility of the combined complex, licensing of the technology need to be obtained before it can be implemented. Similarly, construction and operation of the combined complex authorities. Therefore, the next topic of discussion is the regulatory aspects associated with the combined nuclear/chemical complex.