2 NUCLEAR REACTOR- AND HYDROGEN PRODUCTION TECHNOLOGIES

2.1 INTRODUCTION

The successful implementation of a nuclear-assisted hydrogen production technology will depend significantly on the technological and economic feasibility of the combined complex, as well as on safety and regulatory aspects. These concerns are mostly interrelated and will have an influence on each other. Considering that the primary objective of the study is the evaluation of the safety and regulatory aspects associated with a combined complex, evaluation thereof will only be possible if comprehensive knowledge and understanding of the nuclear reactors and hydrogen production technologies are attained. To this extent, since previous studies regarding the safety of the combined complex are based on different nuclear reactors and hydrogen production technologies, these need to be investigated and assessed with regard to applicability to the HTGR and hydrogen production technologies to be investigated in this study. Moreover, the inherent and unique safety characteristics of Generation IV (Gen-IV) HTGR technologies form a vital driver for implementing nuclear process heat applications and require thorough consideration, especially if public acceptance of the technology is to be obtained. These characteristics of Gen-IV HTGRs are examined in this chapter, in addition to the leading candidate Gen-IV nuclear reactors and the different hydrogen production technologies proposed to be coupled with the nuclear reactor.

2.2 HTGRs

HTGRs are the latest development in gas-cooled reactors and differ significantly from the currently operated commercial fleet of light water (H_2O) and heavy water (D_2O) nuclear reactors. HTGRs operate at much higher temperatures, are gas-cooled (Helium) and graphite-moderated, and have very different fuel- and core designs and power conversion units (PCUs). Therefore, they are almost completely different from PWRs and BWRs.

The following table (Table 2-1) shows that experimental gas-cooled reactors operated without major incident from 1965 to 1989 and successfully investigated various configurations and operating conditions (Kugeler, 2005).

Table 2-1 Experimental gas-cooled reactors (Kugeler, 2005)

Parameter	Dimension	AVR	DRAGON	Peach Bottom	THTR-300	Fort St. Vrain
Thermal power	MW	46	20	115.5	750	842
Electrical power	MW	15	-	40	300	330
Net efficiency	%	~33	-	~ 34	40	39,2
Average core power density	MW/m^3	2,2	14	8,3	6	6,3
Core height	m	3	2.54	2,3	5,1	4,7
Core diameter	m	3	1,07	2,8	5,6	5,9
Type of fuel element	--	Pebbles	Rods in cluster	Rods	Pebbles	Blocks
Fuel + fertile material	--	UO_2, ThO_2	UO_2, ThO_2	UO_2, ThO_2	UO_2, ThO_2	ThC_2, UC_2
Dimensions of fuel elements	cm	6	~ 17	8,9	6	36 (hexagonal)
Fuel arrangement	--	Statistical pebble bed	7 hexagonal rods per cluster	Single positioning	Statistical pebble bed	247 columns with 6 blocks
Coolant pressure	Bar	11	20	24	40	49,2
Coolant heat up	°C	270->950	350->750	344->770	250->750	400->770
Direction of flow inside core	--	Upwards	Upwards	Upwards	Downwards	Downwards
Number of fuel elements in core	--	100.000	37 (7 rods each)	682	675.000	1482
Type of coated particles	--	BISO, TRISO	BISO	BISO	BISO	TRISO
Fuel loading	--	Continuous Loading, MEDUL	Loading during operation	Loading during operation	Continuous Loading, MEDUL	Batch
Heavy metal content in fuel element	gHM/fuel element	6...11	--	--	11 (10g ThO_2)	15.000
Average burn up	MWd/tHM	> 100.000	100.000	60.000	100.000	100.000
Fissile enrichment	%	< 11	93	93	U:93	U:93
Max. fuel temperature in operation	°C	< 1100	< 1350	< 1200	< 1200	< 1260
Max. fuel temperature in accident conditions	°C	< 1400	< 2000	< 2000	< 2200	< 2200
Steam pressure	Bar	73	16	100	180	173
Steam temperature	°C	505	200	540	530	538
Reheat conditions	Bar/°C	--	--	--	42/530	45/538

Both block-type and pebble-bed reactors under various operating conditions and configurations were successfully investigated. However, some innovations such as the use of helium as coolant and TRISO-coated fuel elements in conjunction with configurations that are self-reliant in power limitation and have self-acting decay heat removal may prove to be the most important.

Helium is a very favourable cooling medium: it is chemically inert, has a high heat capacity and does not influence the neutron economy at all. Combining helium as coolant with graphite as moderating- and structural material realizes much higher helium temperatures and therefore thermal efficiencies. However, the higher core temperatures necessitate that the core internals, specifically the fuel elements, be more temperature resilient (Kugeler, 2005). To this extent, optimizing of the fuel

design led to the successful development of block-type and spherical fuel elements that operate safely at extreme temperatures and differ significantly from the well-known pellet-fuel of the light water reactors (LWRs). Figure 2-1 shows the spherical fuel design known as a fuel "pebble" (PBMR, 2008).

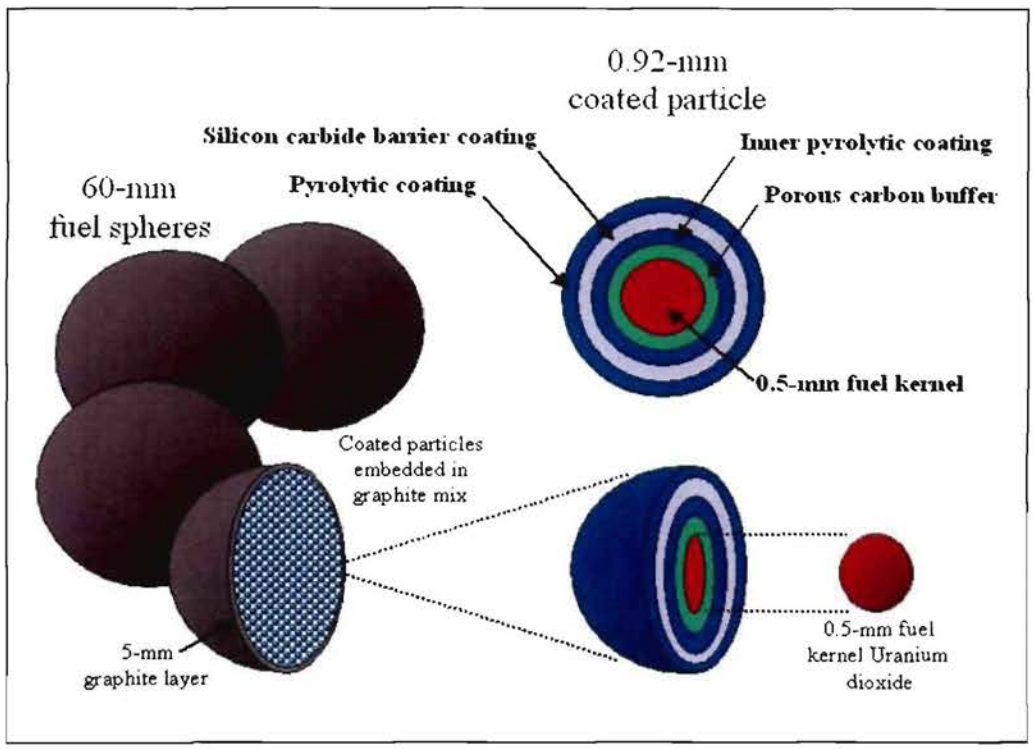

Figure 2-1: TRISO-coated fuel particle (PBMR, 2008)

The TRISO-coated particle consists of a fuel kernel surrounded by a porous carbon buffer layer, enclosed in three layers consisting of an inner pyrolytic carbon layer, a silicon carbide barrier and an outer pyrolytic carbon layer. These fuel particles are embedded in a graphite matrix to form a fuel sphere or "pebble". The porous carbon buffer layer allows the fission product gases to expand from the fuel kernel to reduce the build-up of pressure inside the coated particle, while the outer layers protect the fuel particles from damage as well as retention of the fission products within the coated particle. This design keeps its structural integrity and does not release unacceptable quantities of fission products during severe accident conditions (maximum of 1600^{0}C) and therefore allows for high operating temperatures (maximum of 1350^{0}C) to be realised safely (Kugeler, 2005).

2.2.1 GENERAL CHARACTERISTICS OF HTGRs

HTGRs differ significantly according to application, configuration and operating conditions but some general features characterize the technology. These include (Kugeler, 2005):

- High helium inlet temperatures (250 - 550°C) and outlet temperatures (700 - 950°C) with helium pressure in the range of 40 - 90 bar.
- Low core power densities in the range of 3 - 6 MW/m³ (in order to adhere to the principle of self-acting decay heat removal)
- Modular reactors with thermal power in the range of 200 – 600 MW_t
- Continuous loading of fresh fuel elements, recycling of partially spent fuel elements and removal of spent fuel elements in pebble-bed reactors (batch loading of fuel elements in block-type reactors).
- Control and shut down of pebble-bed reactors performed by absorber elements in borings in the side reflector (absorber rods introduced into borings inside the core for block-type reactors).
- Reflectors consist of graphite with a thickness of at least 1m to reduce the fast neutron flux in the reactor vessel.
- A thick-walled metallic core barrel surrounding the total graphite structure to ensure a stable arrangement of the total reactor internals.
- A power conversion unit (PCU) consisting of either a steam generator, a gas turbine process or a combined cycle that are more efficient than LWRs and therefore result in less specific amounts of high level radioactive waste production.
- Dedicated or co-generating options (electricity and heat) possible.

Even though these aspects characterize the technology, they differ considerably from one design to another and therefore in Section 2.2.4, the leading Generation IV (Gen-IV) HTGRs are examined in more detail. Furthermore, it is important to note at this point that the real appeal of the Gen-IV HTGRs is their safety characteristics, which subsequently form the next topic of discussion.

2.2.2 SAFETY CHARACTERISTICS OF HTGRs

The worldwide nuclear power plant boom of the sixties and seventies made the safety of nuclear power plants a major issue in the industry and topic of immense

public controversy. The Three Mile Island (TMI) and Chernobyl accidents "justified" these concerns to some extent (although much of the public's fear was almost irrational) and resulted in more stringent safety regulations. Even though these accidents were terrible and almost ruined the future of nuclear energy, some positives can be taken from them. The TMI accident may even be considered a very expensive test since an operator error that led to a core meltdown did not result in the release of any radioactive material to the environment, proving that the containment building was successful. Some commonalities also arise when considering these accidents and include operator error, water as cooling medium (possible phase change) and metal cladding around the fuel, while the RBMK-type reactor (Chernobyl) additionally had a positive void coefficient and insufficient containment. All Gen-IV reactors address these issues and make the probability of a core-meltdown accident extremely remote. Moreover, even if an accident is to occur, the containment will be such that it does not release any radioactive material to the environment. To this extent, the safety characteristics of Gen-IV HTGRs are (Kugeler, 2005):

- Full ceramic fuel elements and core structures that cannot melt, even during an extreme accident such as a total loss of coolant accident (LOCA).
- TRISO-coated fuel elements that retain the fission products very effectively inside the fuel. The coated particles are the main barriers and form approximately 10^9 small pressure vessels in a modular HTGR that retains the fission products within them). Additionally, a primary enclosure, an inner concrete cell and outer concrete building may be employed as barriers to the release of fission products. Figure 2-2 illustrates this defence-in-depth strategy to retain the fission products.
- Helium as cooling medium: Helium is chemically inert, does not influence the neutron economy, has a high heat capacity and allows for high operating temperatures and pressures without the possibility of phase change or danger of having a positive void coefficient.
- Reactor cores that have low power density and can tolerate the loss of coolant and all active decay heat removal. This principle of self-acting decay heat removal ensures that the temperature of the fuel elements stay within acceptable ranges. Therefore, a core meltdown event is extremely improbable.

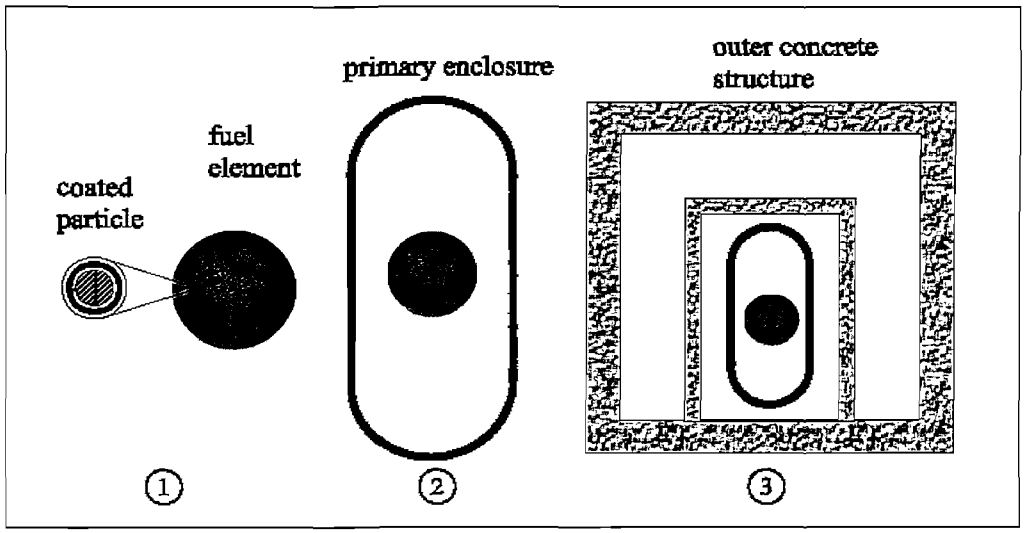

Figure 2-2: Barriers to the release of fission products in HTGR (Kugeler, 2005)

- Reactors have a self-reliant limitation of nuclear power, reactivity and therefore also temperature due to very strong negative temperature coefficients of the fuel elements (discussed in more detail in the next subsection).
- Helium circuits are relatively clean (due to the very efficient retention of fission products within the coated particles) and result in low contamination of the coolant gas, low release of radioactivity from the core and therefore small radiation doses to the operating personnel.

In light of these safety characteristics of HTGRs, it is clear that even when a catastrophic accident does occur it is completely contained within the nuclear power plant. In contrast to LWRs, no unacceptable release of radioactivity to the environment resulting in the loss of life, land contamination and subsequent large monetary damages are possible with HTGRs. Furthermore, HTGRs are considered as inherently safe since they adhere to the principles of inherently safe nuclear reactors.

2.2.3 PRINCIPLES OF INHERENTLY SAFE NUCLEAR REACTORS

The principles of inherently safe nuclear reactors are shown in Figure 2-3 and consist of the principles of nuclear, thermal, chemical and mechanical stability such that no reactor that adheres to these principles can be destroyed by any other accidents,

ever have a core meltdown or release radioactivity to the environment. Considering HTGRs and their use of coated particles, this effectively reduces to ensuring that the fuel elements never exceed the maximum fuel temperature or release unacceptable amounts of fission products in any hypothetical situation (Kugeler, 2005).

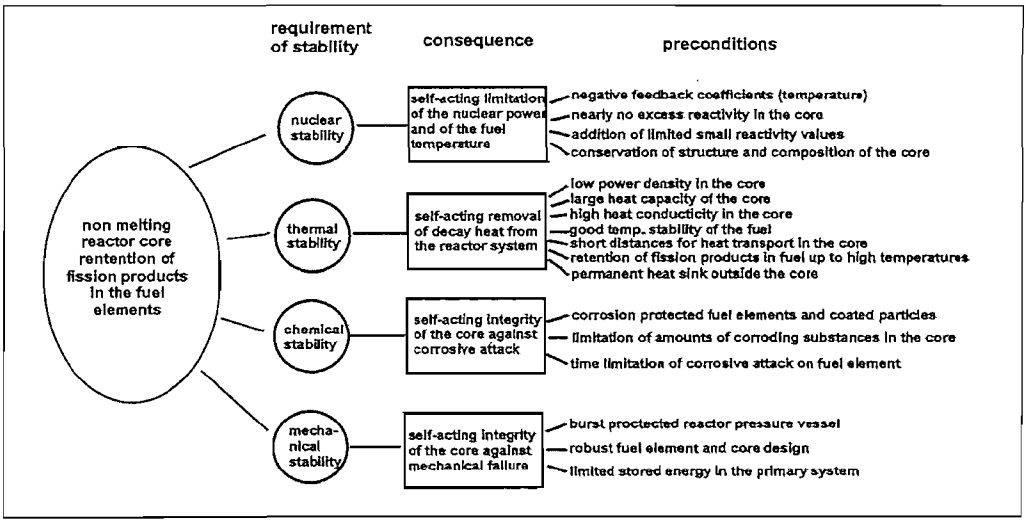

Figure 2-3: Principles of inherently safe nuclear reactors (Kugeler, 2005)

2.2.3.1 NUCLEAR STABILITY

Nuclear stability refers to a nuclear reactor that has a self-reliant limitation on nuclear power and therefore fuel element temperatures and reactivity such that the core can never experience a meltdown in any imaginable situation. This is achieved by having a significantly strong negative temperature coefficient of reactivity (to effectively limit the fuel element temperature) and the Nuclear Doppler Effect. The temperature coefficient of reactivity (α_T) is the response of a reactor to a change in temperature and is defined by the following relation (Lamarsh & Baratta, 2001):

$$\alpha_T = \frac{dp*}{dT} \qquad \textbf{Equation 2-1}$$

With:

α_T Temperature coefficient of reactivity [K^{-1}]
$p*$ Reactivity [%, $, c]
T Temperature [K]

If α_T is negative, an increase in temperature leads to a decrease in power, resulting in a decrease in temperature until the reactor returns to its original state. Similarly, a decrease in temperature leads to an increase in power, resulting in an increase in temperature such that the reactor once again returns to its original state. This system is inherently stable to changes in temperature and is illustrated in the following figure (Figure 2-4; Kugeler, 2005; Lamarsh & Baratta, 2001).

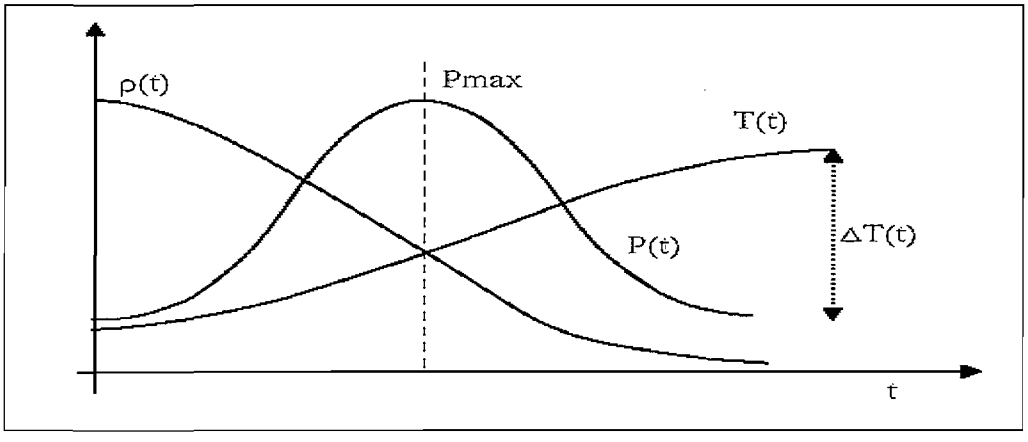

Figure 2-4: Negative temperature coefficient of reactivity (Kugeler, 2005)

Conversely, if α_T is positive, an increase in temperature leads to ever-increasing temperatures and power until the reactor is shut down by outside intervention or it melts down. Furthermore, a decrease in temperature leads to a decrease in power resulting in another decrease in temperature until the reactor shuts down. This system is inherently unstable to changes in temperature (Lamarsh & Baratta, 2001).

The prompt (immediate) temperature coefficient of most reactors is negative due to the phenomenon known as the nuclear Doppler Effect (also called Doppler broadening) that is illustrated in Figure 2-5. In this figure, the y-axis represents the microscopic capture cross section (σ_c), which is a quantity that characterizes the probability that a (fast) neutron is captured within the nucleus and effectively "lost" for fissioning purposes. Figure 2-5 illustrates the nuclear Doppler broadening of U-238 as the temperature is increased from 0^0K (a theoretical value) to 20^0C and 1000^0C (Lamarsh & Baratta, 2001).

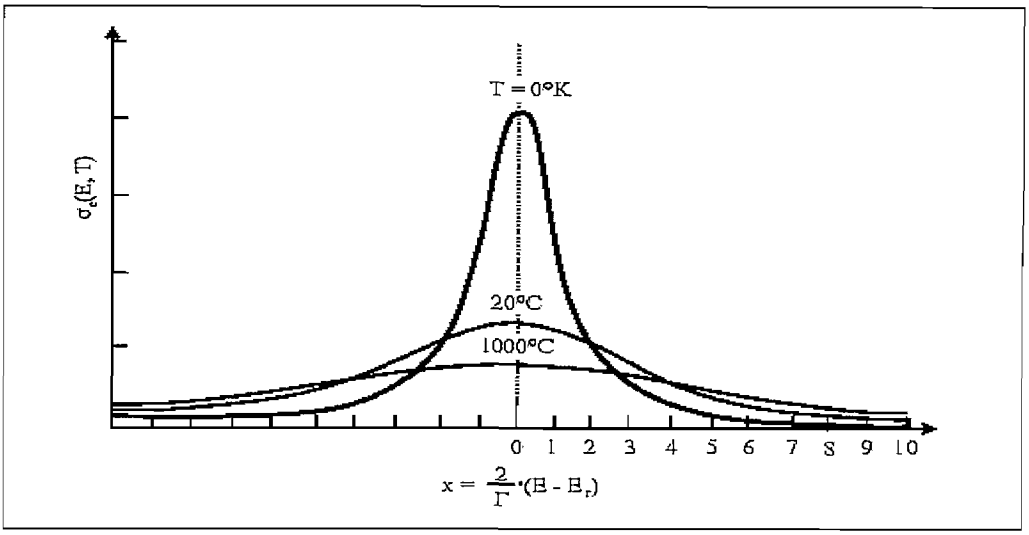

Figure 2-5: Nuclear Doppler Effect (Lamarsh & Baratta, 2001)

The increase in fuel temperature increases the vibration of the atoms of the fuel, which relative to the direction of vibration, increases (away) or decreases (towards) the energy required for the neutron to be captured within the nucleus. This broadening of the energy range and corresponding probability of neutron capturing becomes more pronounced at higher temperatures due to the increased velocity of the vibrating atoms. Therefore, as the fuel temperature increases, the probability of neutrons to be captured increases and the reactivity of the reactor decreases.

2.2.3.2 THERMAL STABILITY

Thermal stability of a HTGR refers to the principle of self-acting decay heat removal from the core in any conceivable situation and without the use of any active cooling systems. Therefore, the reactor should be able to remove the decay heat only by the self-acting processes of conduction and radiation of heat and natural convection of air such that the fuel temperature never exceeds the allowable limits and thereby ensuring that the fission products stay within the coated particles. Note that decay heat is the energy generated by the β- and γ- decay of fission products after the reactor was shut down and may be as much as 7% of the total thermal output of the reactor. Figure 2-6 illustrates the principle of self-acting decay heat removal in HTGRs and that of LWR, which does not have self-reliant decay heat removal systems (Kugeler, 2005).

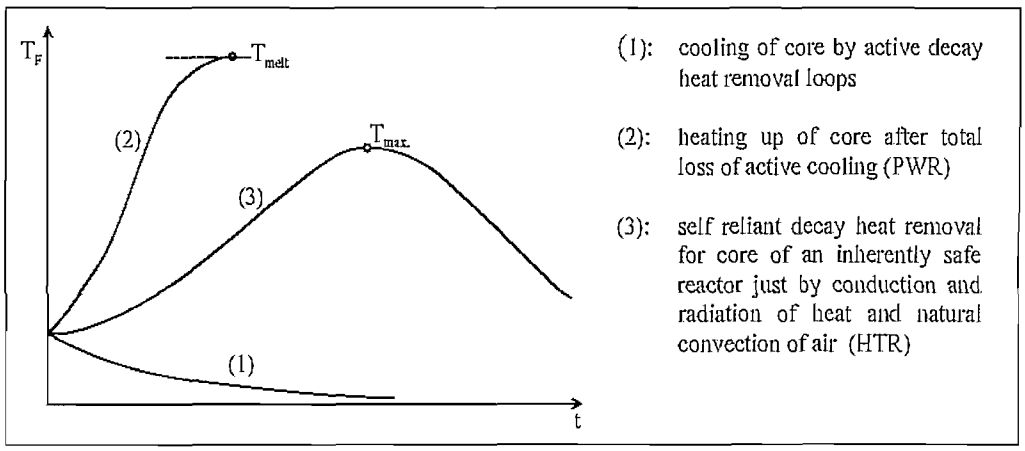

Figure 2-6: Self-acting decay heat removal in HTGR (Kugeler, 2005)

From this figure, it is clear that both LWR and HTGR quickly remove the decay heat when active cooling systems are available. However, when the active cooling systems are not available (due to a total loss of coolant accident) the temperature of the fuel in a LWR rapidly increases above the melting temperature and result in a core meltdown accident, while the HTGR successfully removes the decay heat by self-acting processes. Therefore, core meltdown and the subsequent release of the fission products from the coated particles due to decay heat are not possible for HTGRs if they adhere to the principle of self-acting decay heat removal (Kugeler, 2005). However, the applicability of this principle on the reactor being considered depends on all factors that influence the thermal-hydraulic and neutron behaviour of the reactor, which may be more attainable by modular HTRs due to their design characteristics such as size, core power density and thermal power ratings.

2.2.3.3 CHEMICAL STABILITY

The use of helium as cooling medium forms the foundation of chemical stability of the reactor system due to it being chemically inert and unable to react with the reactor materials even during extreme accident conditions. However, if the boundary of the primary system is compromised, foreign media may enter the reactor system and result in very hazardous situations. Therefore, additional school of thoughts to promote chemical stability is to design the fuel elements and core internals to be able to withstand any corrosion by the ingress of "foreign" media (such as H_2O and air) into the primary circuit, or to design the primary circuit to limit the amount of material that could ingress into the primary circuit. Figure 2-7 shows a concept considering

the latter that consists of an inner concrete cell, a non-retum flap and grain hopper leading to a filter and storage unit (Kugeler, 2005).

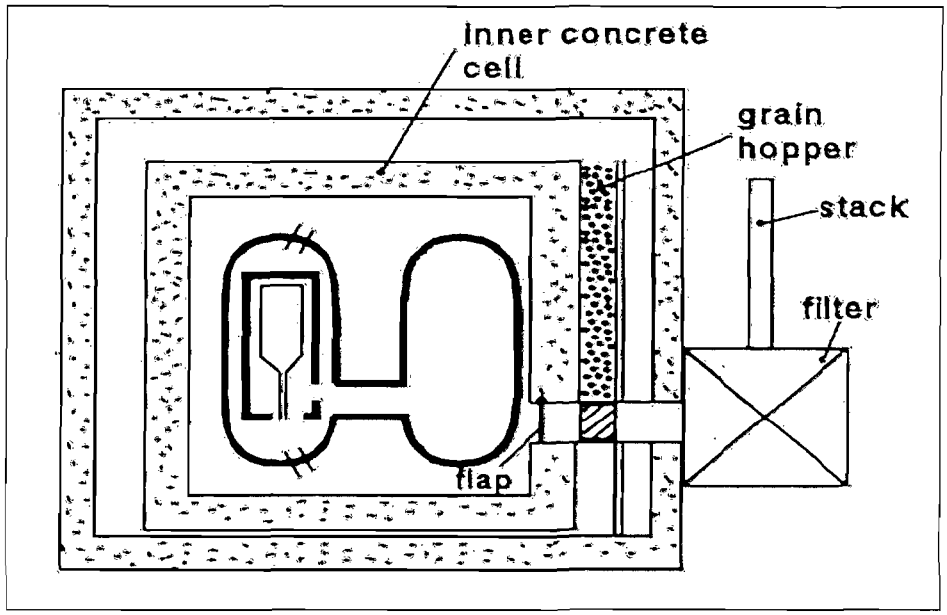

Figure 2-7: Concept of limiting the ingress of foreign media into the primary circuit of HTGR (Kugeler, 2005)

Even though the core intemals are very robust, they are susceptible to corrosion and therefore the main approach to reduce corrosion of the core internals is to limit the amount of foreign media that can ingress into the primary circuit. However, since none of the HTGR designs currently being investigated is contemplating such a solution, the feasibility of this concept (Figure 2-7) is obviously of concern.

2.2.3.4 MECHANICAL STABILITY

Mechanical stability demands that no inner- or predictable outer originating mechanical impacts could alter the consistence or dimensions of the reactor core such that the fuel elements release the fissile material or fission products, even when under extreme accident conditions. In order to adhere to this principle for inner originating mechanical impacts, the reactor core internals are designed to be very robust. While a thick-walled metallic core barrel surrounds the already robust total graphite structure of the reactor to ensure a stable arrangement of the reactor internals, appropriate mechanical fuel design of the fuel elements (Table 2-2) ensures their integrity (Kugeler, 2005).

Table 2-2: Mechanical characteristics of HTGR fuel pebbles (Kugeler, 2005)

Parameter	Dimension	Value	Conditions
Crashing strength	kN	26,3	parallel equator
Crashing strength	kN	23,7	vertical equator
Free fall index	-	638	number of drops before damage
Corrosion rate	mg/cm^2 h	0,08	at 900 °C
Corrosion rate	mg/cm^2 h	0,70	at 1000 °C
Abrasion	µm	500	500
Fraction of defect particles	-	$1,3 \cdot 10^{-5}$	results of irradiation
Coating layer sequence		Buffer/PyC/SiC/PyC	Buffer/PyC/SiC/PyC

However, the robustness of the reactor core internals and fuel elements cannot ensure their integrity when all predictable external events are considered. To this extent, a thick walled outer concrete structure is employed to protect the reactor internals from external events such as those shown in Figure 2-8 and considered during the licensing process (Kugeler, 2005).

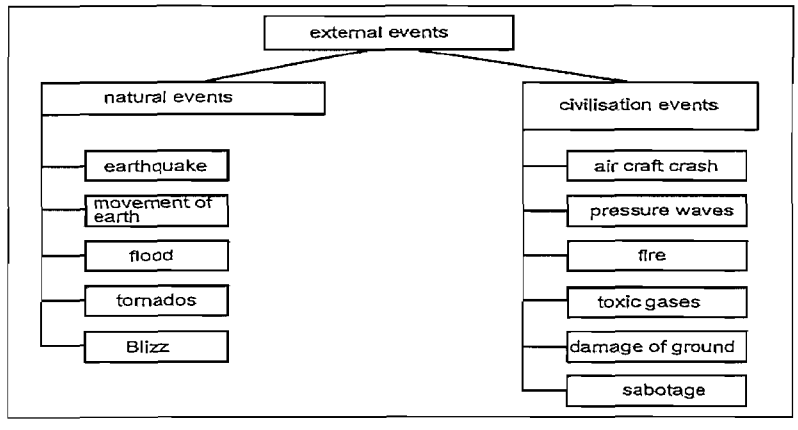

Figure 2-8: External events considered for licensing of HTGR (Kugeler, 2005)

External events not considered during the licensing process include terrorist attacks, sabotage, war impacts, extreme earthquakes, meteorites and atomic bombs, some of which can be avoided by locating the power plant underground or by covering the facility underneath suitably thick solid soil layers (Kugeler, 2005).

The success of the experimental HTGRs, the inherent safety characteristics of the Gen-IV HTGRs and the resurgence of interest in nuclear power plants have led to the investigation of several innovative HTGRs. The following table (Table 2-3) is a summary of the leading candidate HTGRs and their design parameters, which correspondingly form the next topic of discussion (Kugeler, 2005).

Table 2-3: Leading candidate HTGR technologies (Kugeler, 2005)

Parameter	Dimension	HTR Module 200	HTR-100	HTTR-30	HTR-10	PBMR	GAC 600
thermal power	MW	200	250	30	10	400	600
electrical power	MW	80	100	-	2,5	165	286
electrical efficiency	%	40	40	-	25	41	~47,5
average core power density	MW/m³	3	4.4	2.5	2	4	6.6
core shape	-	cylindrical	cylindrical	cylindrical	cylindrical	annular	annular
core height	m	9.43	8	2.9	2	8	8
type of fuel element	-	pebbles	pebbles	block type (pin in graphite blocks)	pebbles	pebbles	block types
fuel + fertile material	-	UO_2	UO_2	UO_2	UO_2	UO_2	UO_2, PuO_2(90%)
dimensions of fuel elements	cm	6	6	36	6	6	36
fuel arrangement	-	statistical pebble bed	statistical pebble bed	block type	statistical pebble bed	statistical pebble bed	block type
coolant pressure	bar	60	70	40	30	70	70
coolant heat up	°C	250 →700	255→ 700	395→ 850 (950)	250→ 700 (250→ 950)	560→ 900	490→ 850
direction of flow inside the core	-	downwards	upwards	downwards	downwards	downwards	downwards
number of fuel elements in the core	-	350 000	317 000	270	27 000	545 000	1020
type of coated particles	-	TRISO	TRISO	TRISO	TRISO	TRISO	TRISO
fuel loading		contin. loading MEDUL	contin. loading MEDUL	batch	contin. loading MEDUL	contin. loading MEDUL	2 x batch
heavy metal content in fuel elements	g HM/fuel element	7	14.6		5	8	~ 700 g Pu
average burn up	MWd/tHM	70 000	100 000	22 000	80 000	80 000	121 000
fissile enrichment	%	8	9	6	17	8	~ 15.5
max.fuel temp. in operation	°C	950	1050	1400	900	1100	< 1200
steam pressure	bar	180	190	IHX: 41, (He)	40	-	-
steam temperature	°C	530	530	IHX: 880/ 144 (He)	440	-	-
max. fuel temp. in accident conditions	°C	< 1500	< 1700	< 1600	< 1000	< 1600	< 1600

2.2.4 LEADING CANDIDATE GEN-IV HTGRs

The leading candidate Gen-IV HTGR technologies discussed in this section are the HTTR-30 (Japan), PBMR (South Africa), HTR-10 (China) and GT-MHR (USA/GUS).

2.2.4.1 HTTR-30 (JAPAN)

The HTTR-30 was developed by JAERI in Japan and has been in operation since 1999 with a thermal power output of 30 MW and an average core power density of 2.5 MW/m^3. Figure 2-9 shows the core structure, which is cylindrical and has an active core height of 2.9 m and diameter of 2.3 m. Helium coolant gas enters at the top of the core at 40 bar and is heated from 395 ^{0}C to 850 ^{0}C (recently 950^{0}C) before exiting at the bottom of the core (Kugeler, 2005).

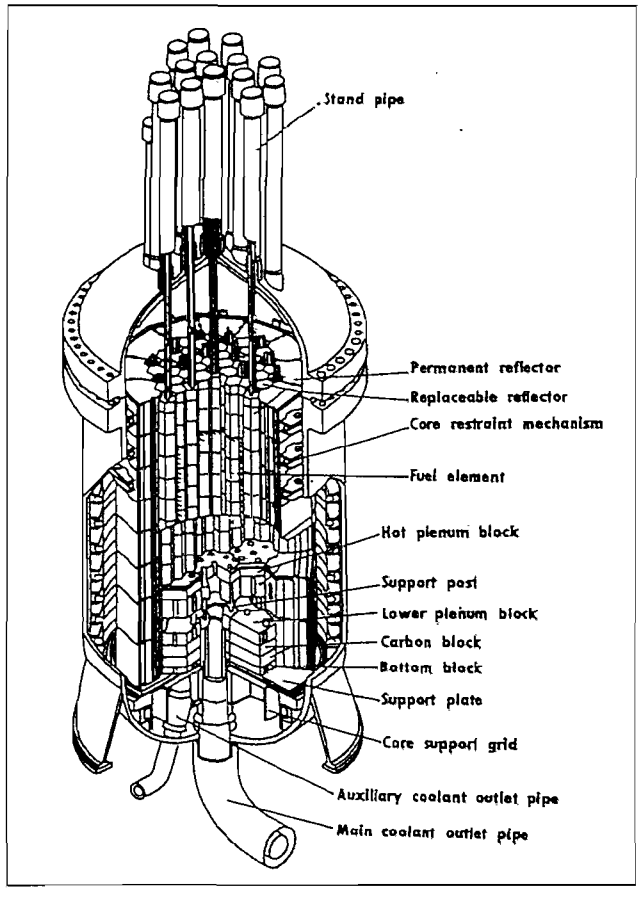

Figure 2-9: HTTR-30 core (Kugeler, 2005)

The core consists of 30 fuel columns and 7 control rod columns that are batch-loaded with 270 block-type fuel elements, which have a maximum temperature of 1400^{0}C during normal operation and 1600^{0}C in accident conditions. Figure 2-10 shows the fuel elements, which are TRISO-coated UO_2 pellets with 6% enrichment. The coolant helium gas flows through the gap between the vertical hole and the fuel rod surface to remove the heat produced by fission and gamma heating (Kugeler, 2005).

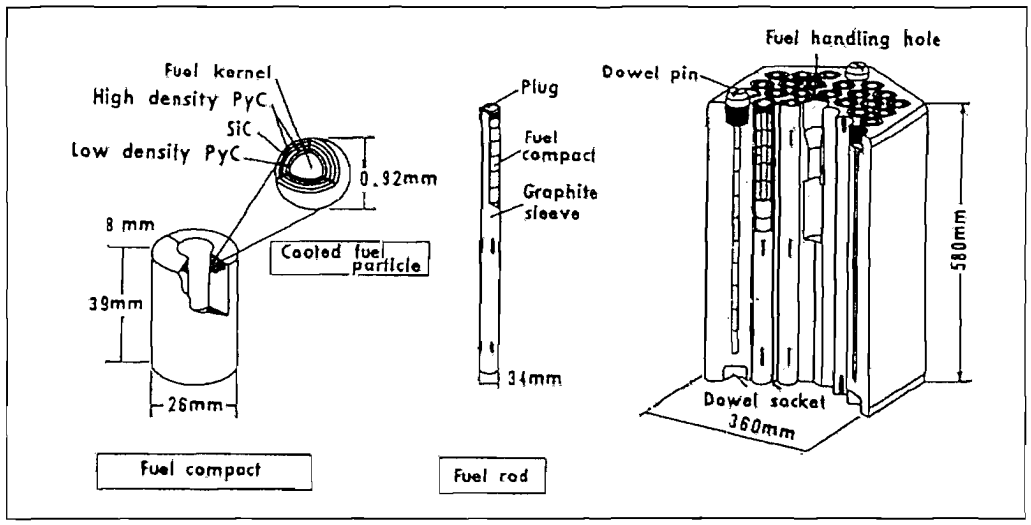

Figure 2-10: HTTR-30 fuel elements (Kugeler, 2005)

The hot coolant gas leaves the reactor via a concentric hot gas duct system and is cooled in a vertical, helically coiled He/He intermediate heat exchanger (IHX) in one line, or by a pressurised water cooler (PWC) in the other line. The PWC is a vertical U-tube heat exchanger with helium coolant flowing on the outside of the heat transfer tubes and pressurised water flowing inside the tubes (Kugeler, 2005).

Reactivity is controlled by inserting control rods into channels in the active core and replaceable reflector regions, while reactor shutdown is achieved by inserting additional control rods into the core. A back-up shutdown system is provided and consists of inserting boron carbide/graphite pellets into holes in the control rod blocks (Kugeler, 2005).

The reactor cooling system consists of a main cooling system (MCS), an auxiliary cooling system (ACS) and two reactor vessel cooling systems (VCS). The ACS is a back-up cooling system in case of failure of the MCS, while the VCS operates continuously to cool the biological shield around the reactor vessel and serve to cool the reactor vessel and core if the MCS fails (Kugeler, 2005).

2.2.4.2 PBMR (SOUTH AFRICA)

The PBMR is a pebble-bed modular reactor combined with a Brayton cycle and a three-shaft-turbo-machine. The components and arrangement of the primary system of the PBMR concept are illustrated in the following figure (Figure 2-11; PBMR, 2008; Kugeler, 2005).

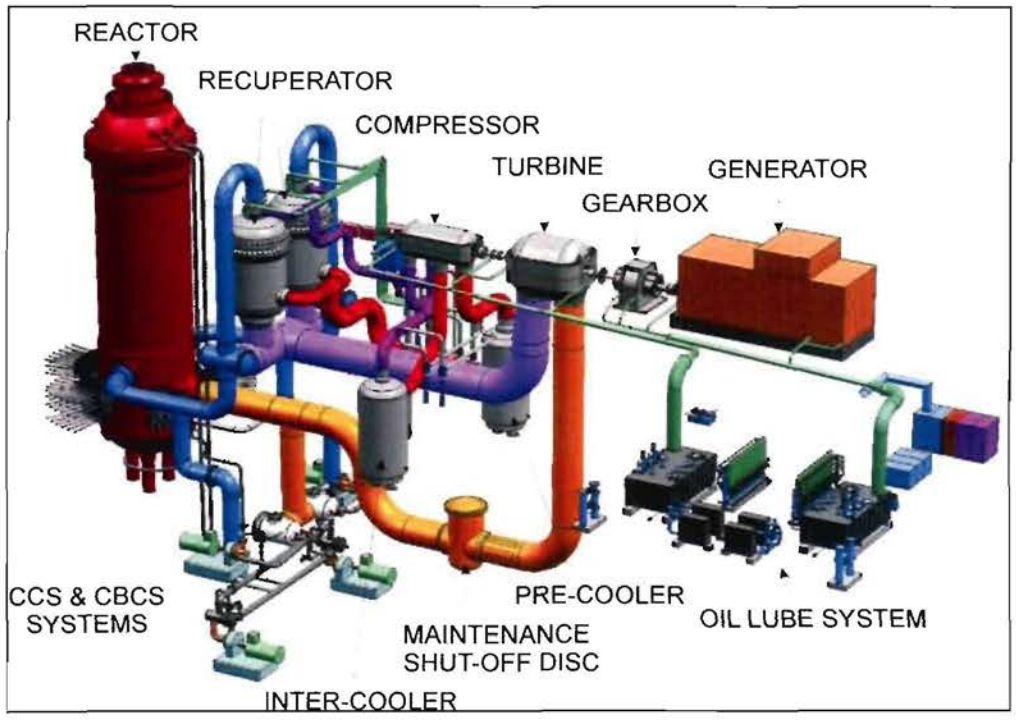

Figure 2-11: Arrangement of primary system of the PBMR (PBMR, 2008)

The PBMR has an annular core with a randomly packed pebble bed fuel arrangement of TRISO coated fuel pebbles in the ring (annular) section and a central graphite reflector in the middle of the core (see Figure 2-12). This arrangement realises a high thermal power rating in the core, while not exceeding the limitation of maximum fuel temperature below 1600 ^{0}C even during severe accidents. The fuel core has an average fuel core height of 11 m, an inner and outer diameter of 2 m and 3.7 m respectively, giving it a fuel core volume of 83.73 m^3. This volume is filled with approximately 450 000 fuel spheres at an average packing fraction of 61% and having an average residence time in the core of 758 days (Van Antwerpen, 2007).

The helium coolant with a mass rate of 160 kg/s enters the reactor at 350 ^{0}C and is heated to 950 ^{0}C while flowing downward through the core and incurring a pressure drop 158 kPa over the length of the core (Van Antwerpen, 2007).

Reactivity control and shutdown of the reactor is achieved by the Reactivity Control System consisting of 12 control rods and 12 shutdown rods and the Reserve Shutdown System that inserts Small Absorber Spheres into 8 borings inside the central reflector (Van Antwerpen, 2007).

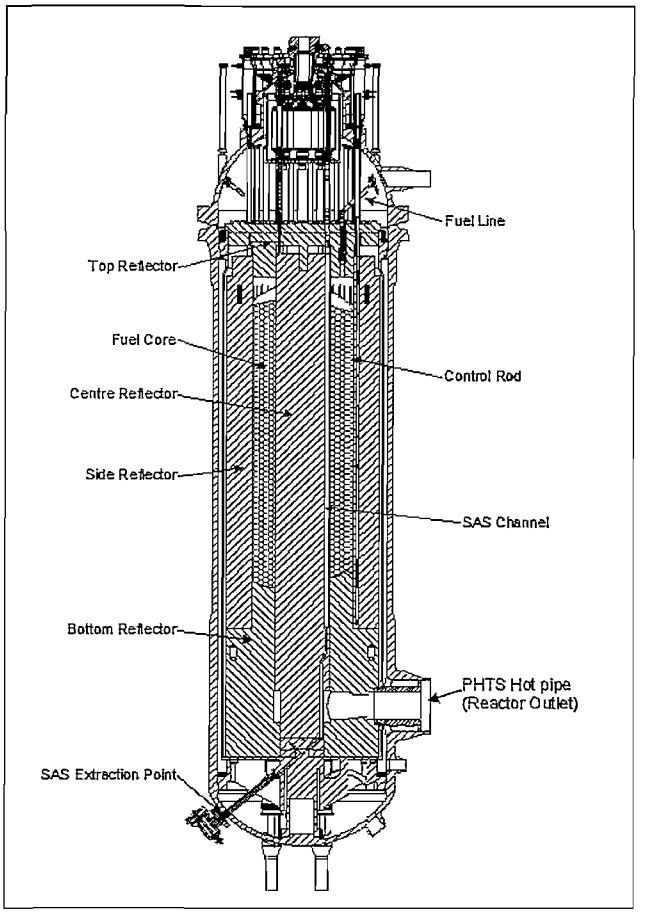

Figure 2-12: Vertical schematic section through PBMR (Van Antwerpen, 2007)

2.2.4.3 HTR-10 (CHINA)

The HTR-10 is an experimental nuclear reactor and started operation in 2002 with the intention of improving HTGR technologies in China and verification of the inherent safety features of the modular HTRs. In the first phase of experiments, the

nuclear reactor is coupled with a steam generator to generate electricity and supply district heat. However, in a later phase of experiments, the nuclear reactor will be coupled to a gas turbine or co-generation plant. The flow sheet and a vertical view of the HTR-10 coupled with a steam generator are shown in Figures 2-13 and 2-14 respectively (Kugeler, 2005).

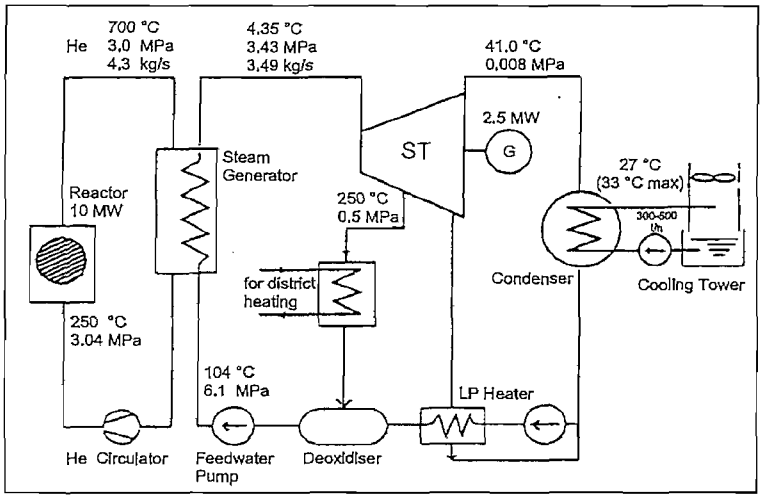

Figure 2-13: Flow sheet of the HTR-10 coupled with a steam generator (Kugeler, 2005)

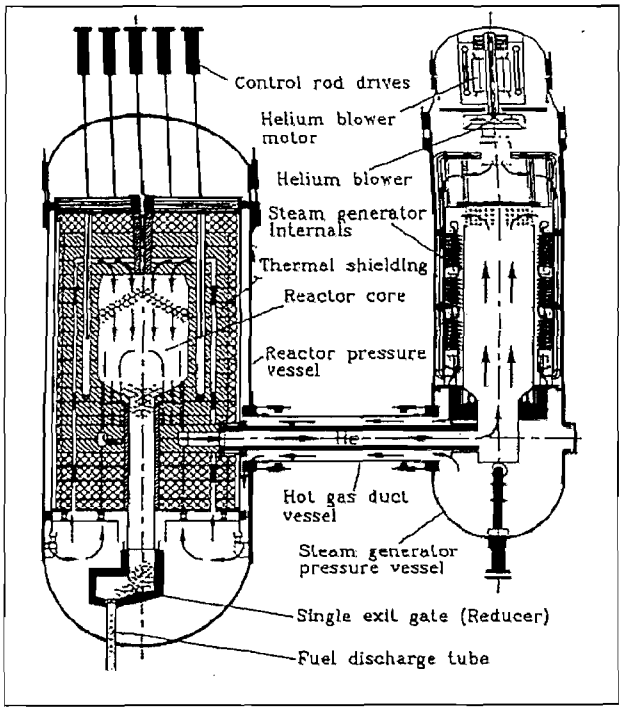

Figure 2-14: Vertical section of the HTR-10 with steam generator (Kugeler, 2005)

The HTR-10 is a statistically packed pebble bed reactor with cylindrical core and a thermal power of 10 MW. The helium coolant enters the top of the core at 250 ^{0}C and is heated to 700 ^{0}C while flowing downwards through the pebble bed core. The HTR-10 has a continuously loaded (MEDUL) fuel cycle and contains approximately 27 000 TRISO-coated fuel pebbles in the cylindrical core. It operates at a helium coolant pressure of 30 bar, has an electrical efficiency of 25 %, a maximum fuel temperature of 900 ^{0}C during normal operation and an average core power density of 2 MW/m^{3} (Kugeler, 2005). These values are comparable low due to the objectives of the reactor being of experimental nature.

2.2.4.4 GT-MHR (USA/GUS)

The GT-MHR concept is being investigated in the USA and is a 600 MW_t VHTR with a helium gas turbine. The following figure (Figure 2-15) illustrates the GT-MHR reactor-building cutaway and shows the arrangement of the reactor and power conversion systems (Hayner *et al.*, 2006).

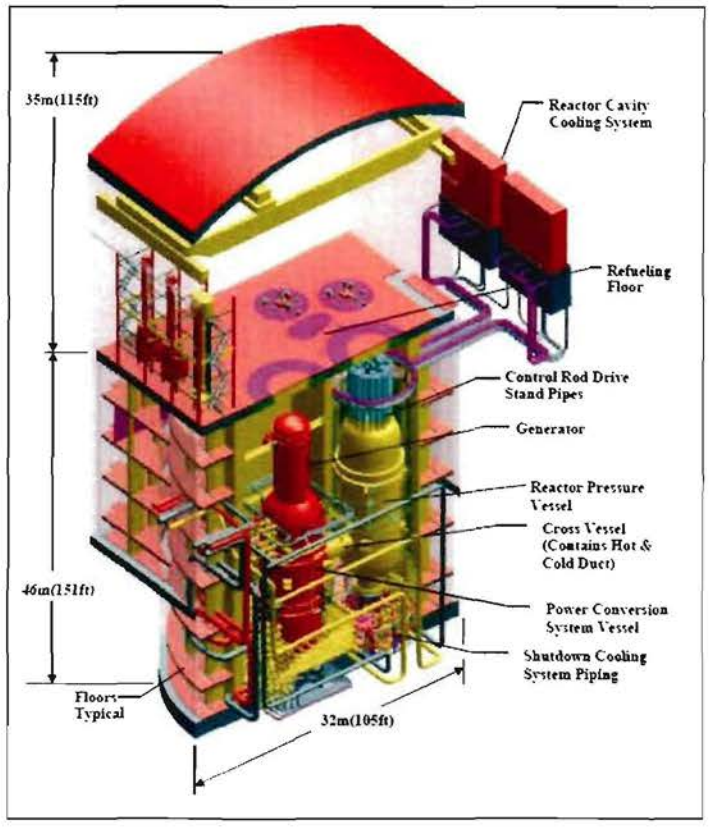

Figure 2-15: GT-MHR reactor building (Hayner *et al.*, 2006)

Figure 2-16 shows the GT-MHR reactor, which has a non-fuelled graphite reflector as core centre and an annular core of TRISO-coated fuel particles embedded in graphite compacts and placed in graphite prismatic blocks. It operates at a helium coolant pressure of 71.2 bar, has a maximum fuel temperature of 1250 ^{0}C during normal operation and an average core power density of 6.5 MW/m^3 (Hayner *et al.*, 2006).

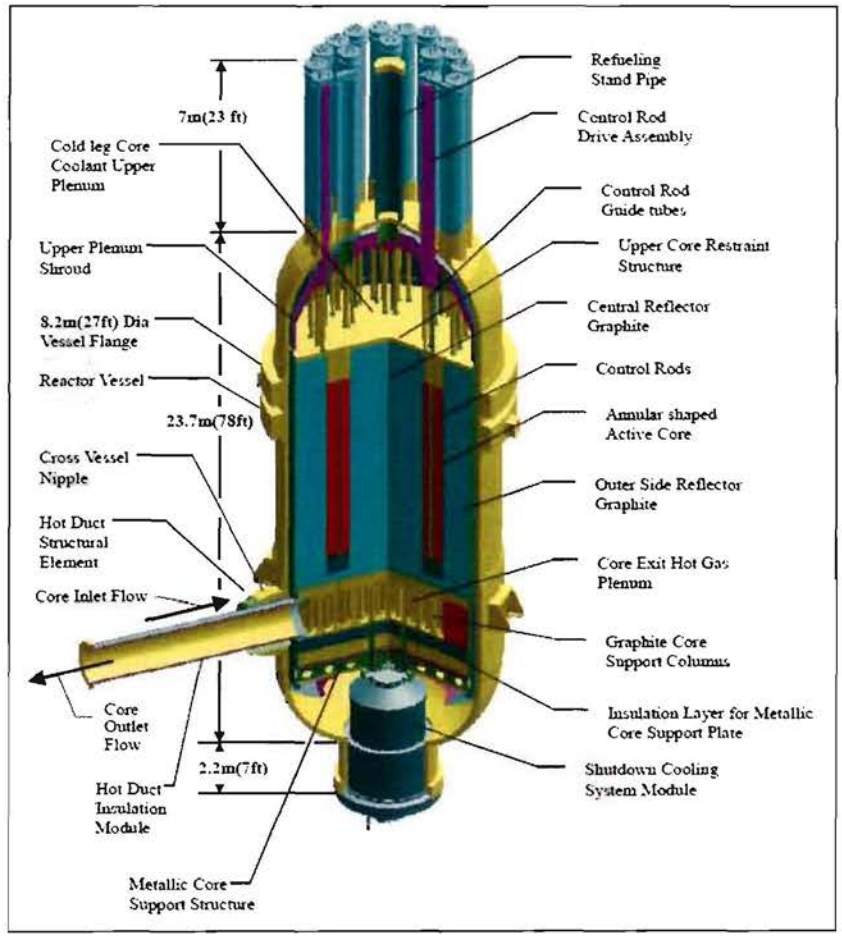

Figure 2-16: GT-MHR reactor system cutaway (Hayner *et al.*, 2006)

The helium coolant enters the top of the core at 488 ^{0}C and is heated to 850 ^{0}C while flowing downwards through the core (Hayner *et al.*, 2006). However, some GT-MHR concepts propose to have helium outlet temperatures in the range of 950 – 1000 ^{0}C, which will pose severe materials requirements considering the operating lifetime of 60 years. The comparably high core power density of 6.5 MW/m^3 will also test the capability of the decay-heat removal systems.

2.3 NUCLEAR-HYDROGEN PRODUCTION TECHNOLOGIES

The energy generated by a HTGR can be utilized to supply electricity and process heat to a hydrogen production facility. The following options exist:

1. Supplying electricity for electrochemical processes (electrolysis)
2. Supplying high-temperature heat for purely thermochemical processes.
3. Supplying both electricity and high-temperature heat for hybrid-thermochemical processes and high-temperature steam electrolysis.

Therefore, the energy generated by a HTGR can be used for various hydrogen production technologies. In this section, the currently available technologies of low-temperature water electrolysis (LTWR) and steam methane reforming (SMR), as well as the most promising medium- to long-term technologies of high-temperature steam electrolysis (HTSE), partial oxidation (POX) of methane, plasma-arc reforming of methane, iodine-sulphur cycle (I-S) and hybrid-sulphur cycle (HyS) are examined.

2.3.1 ELECTROLYSIS PROCESSES

Electrolysis processes include low-temperature electrolysis of water and high-temperature electrolysis of steam where the water or steam decomposes directly into H_2 and O_2. Both processes require large quantities of electrical energy that render them (currently) very expensive. Where the low-temperature process can be coupled with LWRs, the high-temperature process requires a HTGR or other high-temperature nuclear reactor (HTR). The benefit of using the low temperature electrochemical process to produce hydrogen is that the nuclear plant and hydrogen production facility can be separated by relatively large distances (Verfondern, 2007).

2.3.1.1 LOW-TEMPERATURE WATER ELECTROLYSIS (LTWE)

Pure hydrogen can be produced by the electrolysis of water by either using liquid electrolyte cells (most commonly potassium hydroxide) or solid polymer proton exchange membranes (PEM). In both techniques the water is dissociated by applying an electrical current over the "system" (Verfondern, 2007).

In an alkaline electrolysis cell, which normally contains an aqueous caustic solution of 20-40% KOH or NaOH, the electrical energy is applied to the nickel or chromium-

nickel steel electrodes and decomposes the water at the cathode to H_2 and OH^-. The OH^- travels through the electrolyte and a separating diaphragm to the anode where the O_2 is liberated. This process is schematically represented in Figure 2-17 and consists of the following three electrolytic reactions:

Electrolyte:	$4H_2O \Leftrightarrow 4H^+ + 4OH^-$	**Reaction 2-1**
Anode:	$4OH^- \Leftrightarrow O_2 + 2H_2O + 4e^-$	**Reaction 2-2**
Cathode:	$4H^+ + 4e^- \Leftrightarrow 2H_2$	**Reaction 2-3**

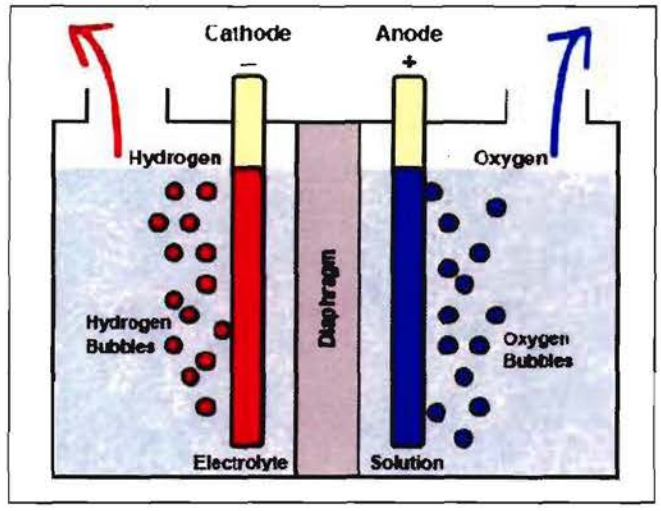

Figure 2-17: Schematic representation of an electrolytic cell (Verfondern, 2007)

The second technique uses an acidic proton exchange membrane (PEM) as a diaphragm and makes the use of an additional electrolyte obsolete because the PEM allows the H^+ to diffuse through the membrane and recombine with electrons to form H_2, but retains the oxygen in the water solution (Verfondern, 2007).

2.3.1.2 HIGH-TEMPERATURE STEAM ELECTROLYSIS (HTSE)

During high-temperature steam electrolysis, the HTR supplies both steam and electricity to the electrolytic cell as is illustrated in Figure 2-18. This is achieved by splitting the high-temperature stream exiting the HTR into two and using them to produce electricity in the PCU and superheated steam in the steam generator respectively (Verfondern, 2007). Therefore, high-temperature steam electrolysis is a

hybrid-thermochemical process, but it is discussed in this subsection due to its relation to low-temperature water electrolysis.

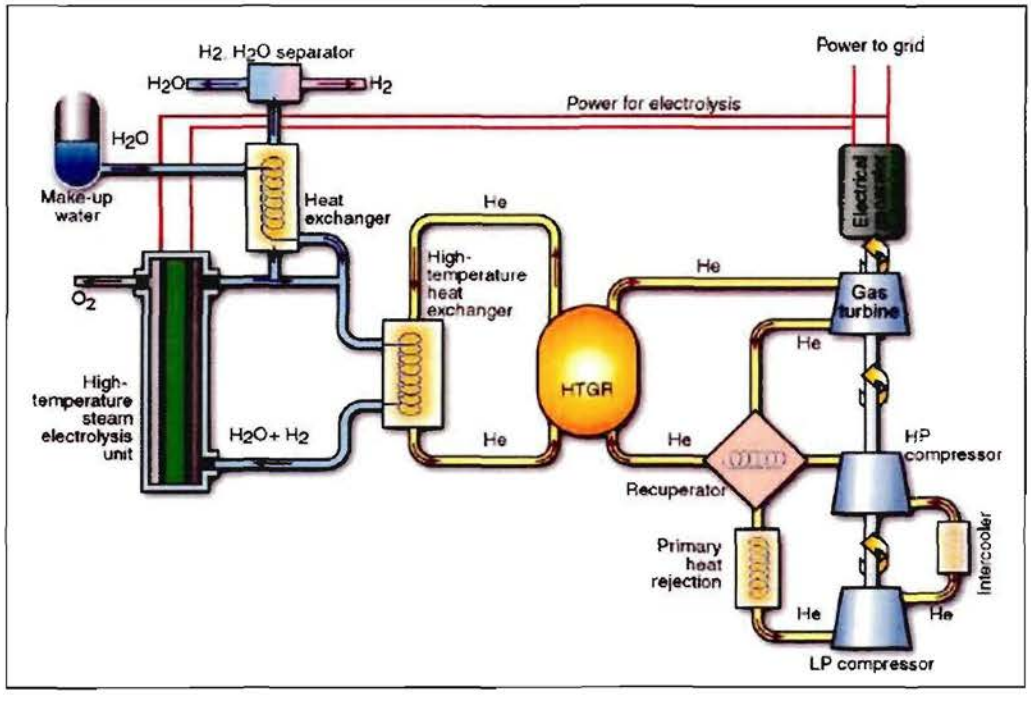

Figure 2-18: HTSE coupled with a HTGR (Verfondern, 2007)

HTSE utilizes solid-oxide electrolytic cells in which the water vapour molecules dissociate at the porous cathode to produce an enriched H_2O/H_2 mixture, while the oxygen ions are transported through a non-porous, ion-conducting solid electrolyte to recombine at a porous anode. In this way, the ion-conducting solid electrolyte membrane automatically separates the product gases (H_2 and O_2). However, a disadvantage of both LTWE and HTSE is that they require a high cell voltage (Verfondern, 2007).

2.3.2 THERMOCHEMICAL CYCLES

Thermochemical cycles release hydrogen from hydrocarbons or water through thermally assisted chemical reactions. In these cycles, the HTGR provides high-temperature process heat that is used to drive the chemical reactions through the use of appropriate heat exchangers (reactors). Some processes use hydrocarbons as feedstock whereas others split water into H_2 and O_2 through a series of thermally

driven chemical reactions. The thermochemical cycles discussed in this section are nuclear steam methane reforming, the sulphur-iodine cycle and partial oxidation of methane or natural gas.

2.3.2.1 NUCLEAR STEAM METHANE REFORMING

The SMR process involves the catalytic decomposition of methane by reacting (reforming) with steam in the presence of an iron or nickel catalyst to produce a hydrogen rich gas mixture. These endothermal reforming reactions typically occur at 850^{0}C and pressures of 2.5 - 5 MPa according to (Verfondern, 2007):

$$CH_4 + H_2O \Leftrightarrow 3H_2 + CO \qquad (-206 \text{ kJ/mol}) \qquad \textbf{Reaction 2-4}$$

$$CH_4 + 2H_2O \Leftrightarrow 4H_2 + CO_2 \qquad (-165 \text{ kJ/mol}) \qquad \textbf{Reaction 2-5}$$

Furthermore, in order to increase the production of hydrogen and reduce carbon deposition on the catalyst surfaces by the Boudouard reaction, the carbon monoxide produced during the first reforming reaction are catalytically converted by the exothermic water-gas-shift reaction with steam according to (Verfondern, 2007):

$$CO + H_2O \Leftrightarrow H_2 + CO_2 \qquad (+41 \text{ kJ/mol}) \qquad \textbf{Reaction 2-6}$$

The composition of the reformer gas, and therefore the conversion of methane, depends significantly on the fuel characteristics, the steam-to-carbon ratio, and the reformer outlet temperature and pressure. To this extent, high steam-to-carbon ratios (>2 to avoid carbon deposition), high reformer temperatures and low pressures allow for higher methane conversions to be obtained. Furthermore, if excess steam is injected the equilibrium in the water-gas-shift reaction shifts toward the production of H_2 and CO_2, thereby increasing the output of H_2 and reducing the amount of CO that can result in carbon deposition by the Boudouard reaction. Since the stream exiting the water-gas-shift reactor contains CO, CO_2, residual steam and unreformed CH_4, these components need to be removed by pressure swing absorption or membrane separation units to realize a H_2 product with purity > 99% (Verfondern, 2007).

Figure 2-19 illustrates a conceptual flow sheet of a SMR facility coupled to a HTGR plant and shows that the nuclear power plant (NPP) provides the heat energy

required by the reforming reactions and produces the superheated steam. After driving the steam turbines, the steam is mixed with the feed and recycling CH_4, is preheated twice and then enters the helium-heated steam reformer (Kugeler, 2005). By utilizing the heat generated by a HTGR for pre-heating of the process streams, approximately 30 % less CO_2 is produced compared to a conventional SMR plants that utilize fossil fuels for this purpose (Verfondern, 2007).

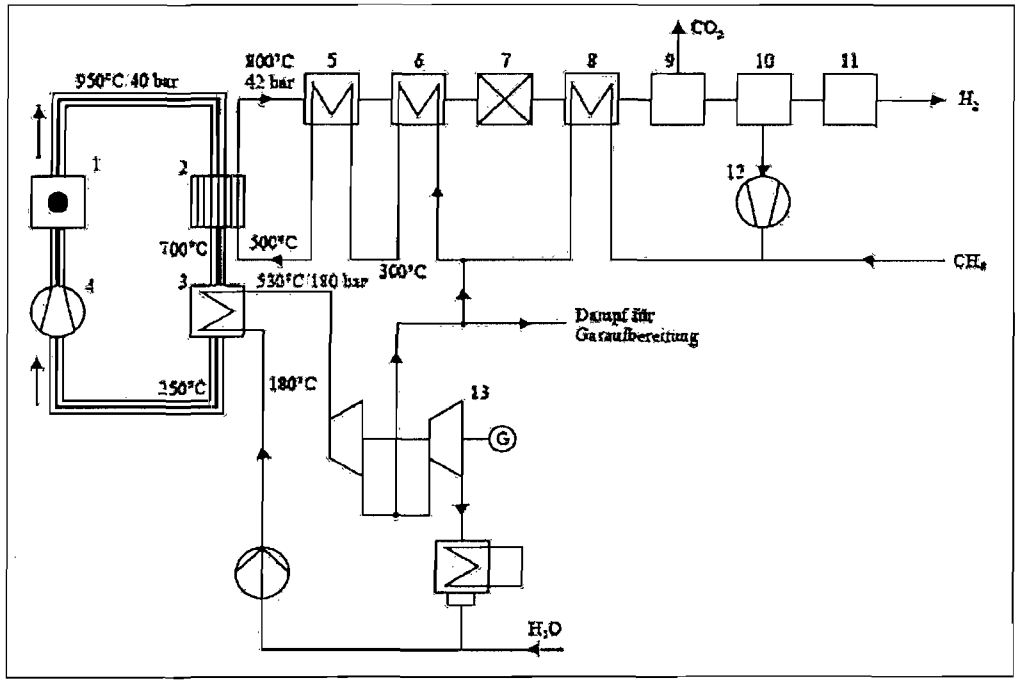

Figure 2-19: Flowsheet of nuclear SMR (Kugeler, 2005)

In this figure, the following is applicable: (1) - HTGR, (2) – helium-heated steam reformer, (3) - steam generator, (4) - helium blower, (5) and (6) – preheating of steam and CH_4 mixture, (7) – water-gas-shift conversion, (8) – preheating of CH_4, (9) - CO_2 remover (PSA or membrane separation), (10) - H_2/CH_4 separation unit, (11) – methanation unit, (12) - CH_4 compressor, and (13) - steam turbine plant.

2.3.2.2 IODINE-SUPHUR CYCLE (I-S)

The Iodine-Sulphur (or Sulphur-Iodine) cycle was originally developed by the American company GA in the mid-1970's and has since received a great deal of attention worldwide. Several modifications have been made to the original process,

however, the base reactions responsible for the dissociation of water remain the same and are (Verfondern, 2007):

$$I_2 + SO_2 + 2H_2O \Leftrightarrow 2HI + H_2SO_4 \quad (120^0C) \quad \textbf{Reaction 2-7}$$

$$H_2SO_4 \Leftrightarrow SO_2 + H_2O + {}^1/_2\, O_2 \quad (830\text{-}900^0C) \quad \textbf{Reaction 2-8}$$

$$2HI \Leftrightarrow I_2 + H_2 \quad (300\text{-}450^0C) \quad \textbf{Reaction 2-9}$$

In this process, all reactions involve fluid interaction and all reagents are recycled, therefore the process does not have any effluents. Moreover, the net process takes in water and uses the high-temperature heat supplied by the HTGR to drive the necessary endothermal reactions, and produces only hydrogen and oxygen as a by-product. It is important to note that some technical issues and uncertainties exist that influence the performance and economics of the process. These include (Verfondern, 2007):

- Materials durability at high temperatures and highly corrosive environments
- Recovery of the HI inventory in the system
- Separation of the reactants and products in certain solutions
- Design of the SO_3 decomposition reactor

Both the decomposition of HI and H_2SO_4 are associated with severely corrosive environments and require materials that can accommodate corrosion and ensure durability. Significant materials testing indicate that tantalum-based materials are the most corrosion resistant in these environments. However, tantalum is also very expensive. Therefore, not only do the materials used in this process influence the efficiency of the process (heat transfer effectiveness), they will also significantly affect the capital cost of the I-S plant. Moreover, the endothermal decomposition of HI into H_2 and I_2 occurs only partially and has an azeotrope in the ternary mixture (Verfondern, 2007; Duigou, *et al.*, 2007; Yildiz & Kazimi, 2006). Therefore, the successful decomposition of HI and separation of H_2 from the remaining components are very difficult and of critical importance to the efficiency of the process since they determine the production of H_2. These issues need to be resolved before the process can successfully be implemented on an industrial scale. Figure 2-20 shows a flow sheet of the Iodine-Sulphur cycle coupled with the HTTR and indicates the proposed operating conditions.

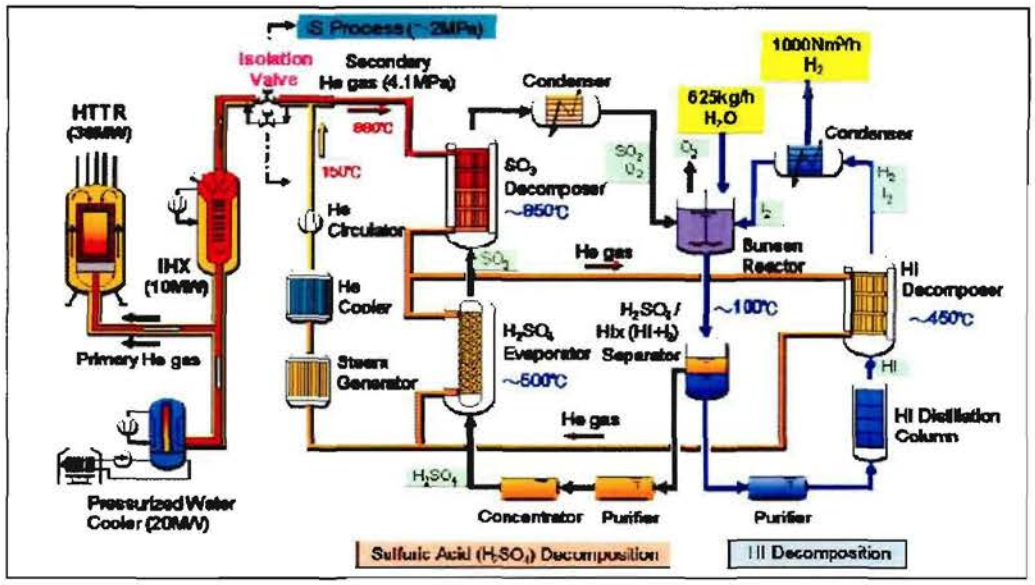

Figure 2-20: Iodine-Sulphur Cycle combined with the HTTR (Verfondern, 2007)

2.3.2.3 PARTIAL OXIDATION (POX) OF METHANE

A natural gas feedstock consisting mainly of methane can be converted to hydrogen in a POX reactor according to the following reaction mechanism:

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O \quad \text{[-802.86 kJ/mol]} \quad \textbf{Reaction 2-10}$$

$$3CH_4 + CO_2 + 2H_2O \rightarrow 4CO + 8H_2 \quad \text{[+659.46 kJ/mol]} \quad \textbf{Reaction 2-11}$$

With the net reaction either of the following two reactions:

$$4CH_4 + 2O_2 \rightarrow 4CO + 8H_2 \quad \text{[-142.64 kJ/mol]} \quad \textbf{Reaction 2-12}$$

$$CH_4 + {}^1/_2\, O_2 \rightarrow CO + 2H_2 \quad \text{[-35.64 kJ/mol]} \quad \textbf{Reaction 2-13}$$

A proposed concept that combines a POX system with a HTGR and hybrid sulphur cycle is illustrated in Figure 2-21. The helium heated by the PBMR exits the reactor at a temperature of 900°C to provide heat to the secondary loop, after which it re-enters the reactor at 350°C. Water entering the HyS is separated into hydrogen and oxygen using the high temperature heat and electricity provided by the PBMR (discussed in Section 2.3.3.1). The hydrogen product is send to storage while the oxygen by-product is routed to a POX process. In the POX process, methane reacts

with oxygen in a flame reactor to produce synthesis gas. Synthesis gas mainly consists of hydrogen, carbon monoxide, carbon dioxide, water and some residual methane.In the case depicted in Figure 2-21, the synthesis gas reacts with steam in a so-called water-gas-shift reactor (WGS) and converts the carbon monoxide into additional hydrogen and carbon dioxide. The subsequent process is a pressure swing adsorption (PSA) unit, which is a very efficient option for the separation of hydrogen from impurities. The PSA is operated batch-wise, but with a sufficient number of units continuity of production can be achieved. The hydrogen product from the PSA is of very good quality at 99.99% purity, while the waste stream of the PSA unit contains the carbon dioxide, carbon monoxide, residual methane and some hydrogen (Conradie, 2008).

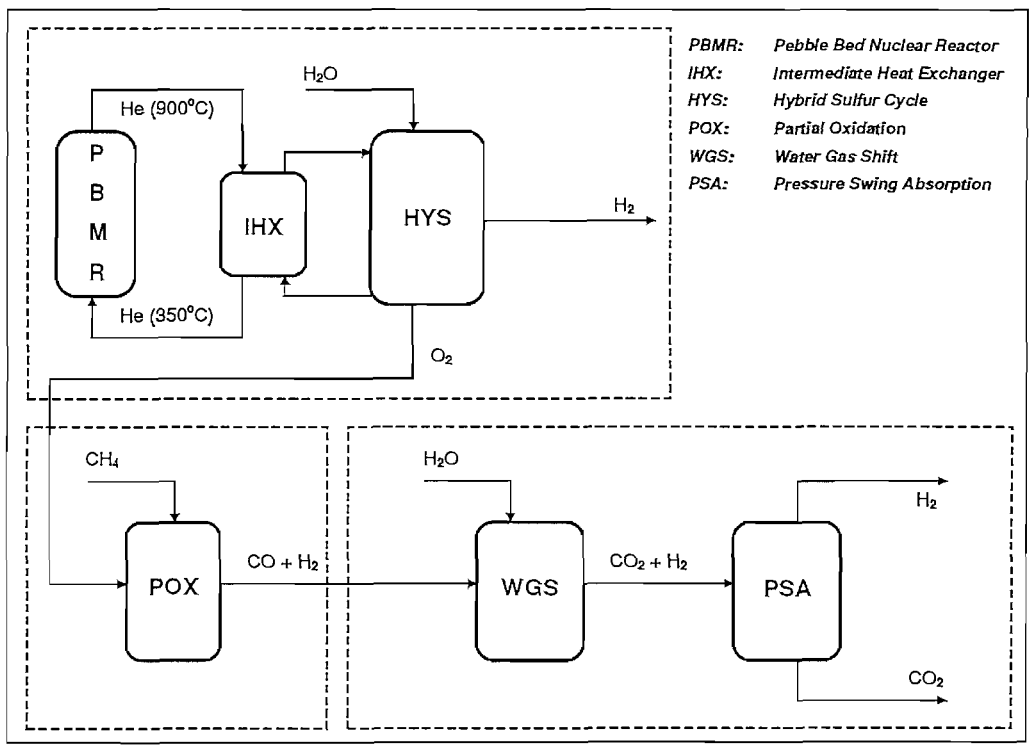

Figure 2-21: Proposed POX system combined with a HTGR and HyS process (Conradie, 2008)

2.3.3 HYBRID THERMOCHEMICAL CYCLES

Hybrid thermochemical cycles combine thermochemical and electrochemical cycles to split water into hydrogen and oxygen. Therefore, in hybrid cycles the nuclear process heat thermally assists the high-temperature endothermal reactions, while the

low-temperature reactions can use electricity produced by the PCU of the nuclear reactor. The hybrid-thermochemical options include high-temperature steam electrolysis, the HyS cycle and plasma-arc reforming of natural gas or methane.

2.3.3.1 HYBRID-SULPHUR CYCLE

The Hybrid-Sulphur cycle (also known as the Westinghouse-Sulphur process - WSP) was developed by Westinghouse during 1973-1983 and is a two-step, sulphuric acid hybrid cycle. The two reaction steps involved in the HyS are the thermal decomposition of H_2SO_4 and the electrolysis of water with SO_2 according to the following reactions (Verfondern, 2007; Jeong *et al.*, 2005):

$$H_2SO_4 \Leftrightarrow H_2O + SO_3 \Leftrightarrow SO_2 + {}^1/_2\, O_2 + H_2O \qquad (871\ ^0C) \qquad \textbf{Reaction 2-14}$$

$$2H_2O + SO_2 \Leftrightarrow H_2SO_4 + H_2 \qquad (120\ ^0C) \qquad \textbf{Reaction 2-15}$$

The first reaction of the HyS is the same chemical decomposition reaction that takes place in the I-S cycle and is extremely endothermal. Therefore, the nuclear process heat is used to drive this reaction as well as to preheat the feed to the reactor in an acid vaporizer as shown in Figure 2-22. This figure is a flow sheet of the HyS cycle and gives the proposed operating conditions of the process. As mentioned above, the second reaction of the HyS is the electrolysis of water with SO_2 to produce sulphuric acid and hydrogen. This reaction takes place without the assistance of nuclear process heat at 20-110°C, E = 0.17 V and 0.2-1 MPa. However, the electrical energy required for this process is supplied by the PCU of the HTGR, hence the term hybrid. The presence of SO_2 in the electrolyser reduces the electrode potential required to well below that of pure water electrolysis (theoretical voltage of 0.17 V vs. 1.23 – 2 V) and therefore reduces the total energy required by the electrolyser. The main difference between the HyS and I-S cycle is the replacement of the HI-decomposition step with the SO_2 electrolyser. Subsequently, the HI-related materials requirements and inventory difficulties are removed from the process. The HyS is simpler in design, requires less reactions to produce hydrogen and since the low-temperature hydrogen production reaction is electrically driven, it can be situated away from the nuclear reactor in order to adhere to regulatory requirements. (The first reaction requires process heat and therefore needs to be close to the reactor to reduce heat losses, but does not involve any combustible components).

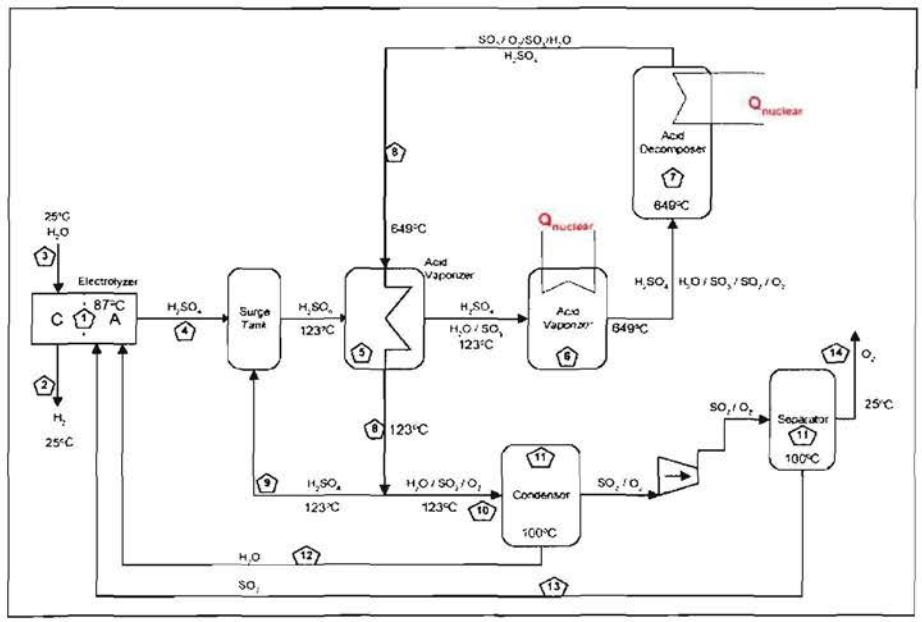

Figure 2-22: HyS cycle or Westinghouse Sulphur process (Jeong et al., 2005)

2.3.3.2 PLASMA-ARC REFORMING OF METHANE WITH CO2

The plasma-arc reforming of methane with CO_2 as oxidizing agent is discussed as a hybrid cycle since both the electricity and the process heat can be applied to assist the reactions. However, the options exist to operate the process as either a thermochemical or electrochemical process depending on the economics and proposed hydrogen production rate. In this section, both the nuclear process heat and the electricity are used to drive the plasma-arc units (19 x 8 MW arc units for a 500 MW_t PBMR) according to the following reaction:

$$CH_4 + CO_2 \rightarrow 2CO + 2H_2 \qquad \textbf{Reaction 2-16}$$

The product gas exiting the plasma-arc unit is cooled and compressed by a series of compressors and coolers, where after it is mixed with superheated steam such that it is at the appropriate conditions to undergo high-temperature and low-temperature water-gas-shift reactions (Reaction 2-6). The product exiting the low-temperature shift reactor is flashed and sent to a PSA unit to obtain a product with > 99% hydrogen purity. Figure 2-23 illustrates the plasma-arc reforming process of methane with CO_2 as oxidizing agent. The unique advantage of this process is that it allows

CO_2 to be extracted from the atmosphere and thereby play a role in damping the Greenhouse effect if employed on a large scale. This aspect may allow policy makers to favour it as hydrogen production technology.

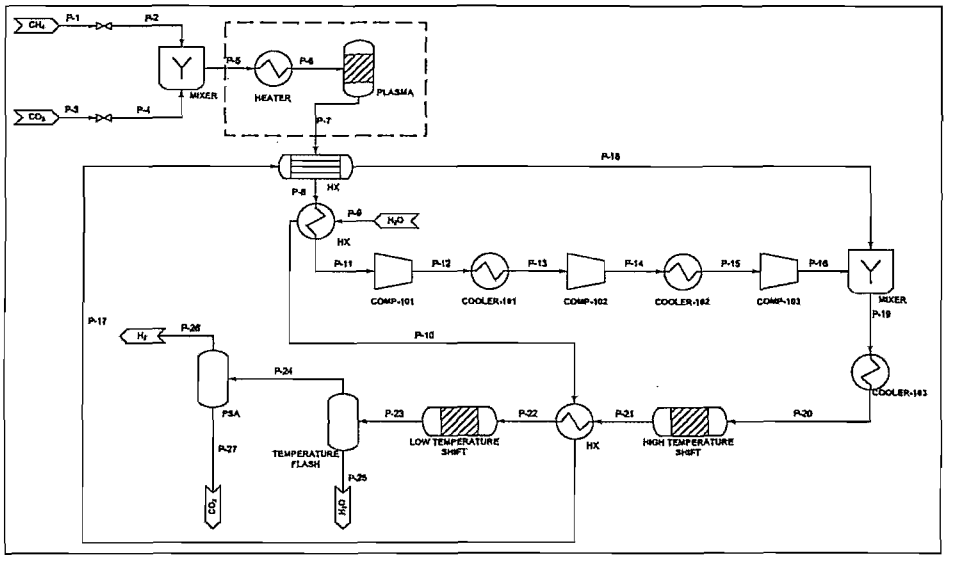

Figure 2-23: Hybrid plasma-arc reforming of methane with CO_2 (Basson, 2008)

2.4 CONCLUDING REMARKS

In this chapter, the general and safety characteristics of Gen-IV HTGR technologies, the leading candidates thereof and the hydrogen production technologies considered to be coupled with the nuclear reactor were examined. In light of this, the characteristics of Gen-IV HTGRs make them uniquely applicable to hydrogen production technologies, especially those that require high-temperature process heat to drive the endothermal reactions. However, the presence of the chemical facility in close proximity to the nuclear reactor results in additional aspects to consider, especially if licensing of the combined complex is to be obtained. Each of the production options have safety concerns due to flammable, corrosive, toxic or asphyxiate components being present in the production streams. An important result of discussing each production option is that the hazardous components associated with each option are indentified, which consequently form the next topic of discussion.