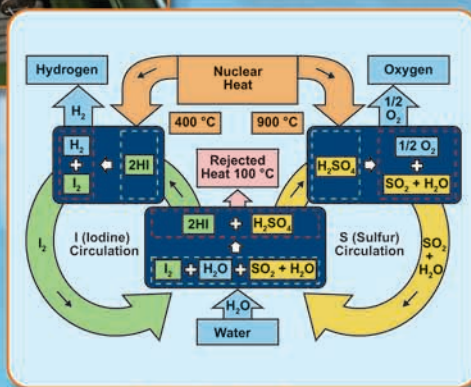
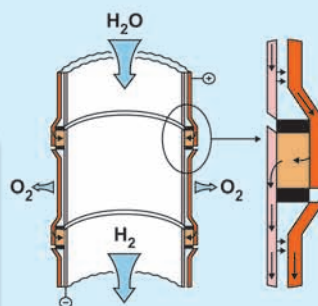


Nuclear Energy for Hydrogen Production

Karl Verfondern (Editor)



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Abstract

With the recent worldwide increased interest in hydrogen as a clean fuel of the future, Europe has also embarked on comprehensive research, development, and demonstration activities with the main objective of the transition from a fossil towards a CO₂ emission free energy structure as the ultimate goal. A major hydrogen economy exists already today and is expected to grow further. Largest near-term markets will be the petrochemical industries requiring massive amounts of H₂ for the conversion of heavy oils, tar sands, and other low-grade hydrocarbons, as well as the fertilizer and steel industries. In the near and medium term, fossil fuels are expected to remain the principal source for hydrogen. In the long term, H₂ production technologies will be strongly focusing on CO₂-neutral or CO₂-free methods.

Nuclear with its virtually no air-borne pollutants emissions appears to be an ideal option for large-scale centralized H₂ production. It will be driven by major factors such as production rates of fossil fuels, political decisions on greenhouse gas emissions, energy security and independence of foreign oil uncertainties, or the economics of large-scale hydrogen production and transmission. A nuclear reactor operated in the heat and power cogeneration mode must be located in close vicinity to the consumer's site, i.e., it must have a convincing safety concept of the combined nuclear/chemical production plant.

A near-term option of nuclear hydrogen production which is readily available is conventional low temperature electrolysis using cheap off-peak electricity from present nuclear power plants. This, however, is available only if the share of nuclear in power production is large. But as fossil fuel prices will increase, the use of nuclear outside base-load becomes more attractive. Nuclear steam reforming is another important near-term option for both the industrial and the transportation sector, since principal technologies were developed, with a saving potential of some 35 % of methane feedstock. Competitiveness will benefit from increasing cost level of natural gas. The HTGR heated steam reforming process which was simulated in pilot plants both in Germany and Japan, appears to be feasible for industrial application around 2015.

A CO₂ emission free option is high temperature electrolysis which reduces the electricity needs up to about 30 % and could make use of high temperature heat and steam from an HTGR. With respect to thermochemical water splitting cycles, the processes which are receiving presently most attention are the sulfur-iodine, the Westinghouse hybrid, and the calcium-bromine (UT-3) cycles. Efficiencies of the S-I process are in the range of 33-36 %, if operated at 950°C which is judged as a feasible upper temperature limit for the reactor and related heat transfer devices. Process optimization and material qualification still require considerable R&D efforts beyond 2015 with regard to the potential of higher efficiencies and more compact chemical reactors to be optimized for commercial use. Technical and economical feasibility, however, remains to be demonstrated; since production processes have not yet been tested beyond pilot plant scale.

A new, perhaps revolutionary nuclear reactor concept of the next generation will offer the chance to deliver besides the classical electricity also non-electrical products such as hydrogen or other fuels (e.g., methanol). In a future energy economy, hydrogen as a storable medium could adjust a variable demand for electricity by means of fuel cell power plants ("hydricity") and also serve as spinning reserve. Both together offer much more flexibility in optimizing energy structures. In China, France, Japan, Korea, and the USA, ambitious programs have been started within the GIF initiative with the main objective to bring nuclear hydrogen production to the energy market. In the European Union, a respective engagement by research, industry, and policy is mainly given by the participation in activities within the Framework Programmes (FP) of the EU.

Kurzfassung

Das weltweit gestiegene Interesse an Wasserstoff als sauberen Energieträger der Zukunft hat auch die Europäische Union veranlasst, ein umfangreiches Forschungs-, Entwicklungs- und Demonstrationsprogramm zu starten mit dem primären Ziel, den Übergang von einer überwiegend auf fossilen Brennstoffen basierenden Energiestruktur auf eine neue mit reduzierten CO₂-Emissionen zu gestalten. Eine Wasserstoffwirtschaft in größerem Maßstab existiert bereits heute und sie wächst weiterhin stark an. Die größten Marktchancen bieten sich derzeit in der petrochemischen Industrie, die enorme Mengen an Wasserstoff für die Raffinierung schwerer Öle, Ölsände oder anderer Kohlenwasserstoffe niedriger Qualität benötigt, ebenso wie in der Dünger- oder Stahlindustrie. Kurz- und mittelfristig werden die fossilen Rohstoffe die Hauptquelle für Wasserstoff bleiben. Langfristig allerdings wird die Produktion auf CO₂-neutralen bzw. -freien Methoden (regenerativ, nuklear) basieren müssen.

Die Nuklearenergie ist aus mehreren Gründen für eine Wasserstoffproduktion im großen Maßstab geeignet. Dies sind zum einen die geringer werdenden Förderquoten fossiler Rohstoffe, klimapolitische Entscheidungen, die gesicherte Energiebereitstellung bei starker Abhängigkeit von Ölimporten, zum andern aber auch die Sicherheit und Wirtschaftlichkeit bei Produktion und Transport großer Mengen an Wasserstoff.

Als kurzfristig verfügbare Option für eine nukleare Wasserstoffproduktion gilt die heute im Industriemaßstab betriebene Niedertemperaturelektrolyse, falls billige Elektrizität aus Nuklearreaktoren verwendet werden könnte. Bei weiter ansteigenden Preisen für fossile Energieträger würde Nuklearenergie auch außerhalb der Grundlast zunehmend attraktiver. Die Dampfreformierung von Erdgas auf der Basis nuklearer Primärenergie stellt eine weitere, mit heutigen Verfahren kompatible Alternative dar. Die Verwendung von Prozesswärme aus einem HTR verspricht dabei ein etwa 35 %iges Einsparpotential beim Rohstoff Erdgas. Sie könnte mittelfristig industriell machbar sein, da bereits alle wesentlichen Komponenten entwickelt und in Pilotanlagen in Deutschland und Japan unter nuklearen Bedingungen erfolgreich demonstriert wurden.

Ganz frei von CO₂-Emissionen ist die Wasserstofferzeugung durch die Spaltung von Wasser. Bei der Hochtemperaturelektrolyse verringert sich der Bedarf an Elektrizität um rund 30 % gegenüber der konventionellen Elektrolyse, wobei die benötigte Prozesswärme von einem HTR bereitgestellt werden könnte. Letzteres trifft ebenfalls auf die thermochemischen Kreisprozesse zu, unter denen der Schwefel-Jod (S-I) Prozess, der sog. Westinghouse Hybridprozess (HyS) sowie der Calcium-Brom (UT-3) Prozess als besonders viel versprechend gelten. Bei Temperaturen von 950°C werden Wirkungsgrade von mehr als 35 % erwartet. Jedoch sind noch umfangreiche F&E Arbeiten zur Prozessoptimierung und Materialqualifikation notwendig, um die technische und ökonomische Machbarkeit zu demonstrieren, so dass die kommerzielle Anwendung erst weit nach 2015 gegeben sein dürfte.

Neue Konzepte von Nuklearanlagen der nächsten Generation werden in der Lage sein, neben Elektrizität auch Prozesswärme (Kraft-/Wärmekopplung) und andere Produkte zu liefern wie z.B. die Energieträger Wasserstoff oder zur Erzeugung von Wasserstoff (oder anderer, synthetischer Kraftstoffe) Methanol. In einer künftigen Energiewirtschaft könnte Wasserstoff als Speichermedium dienen und – in Kombination mit Brennstoffzellen-Kraftwerken – die variable Nachfrage nach Elektrizität regeln, um so flexible Energiestrukturen zu schaffen. Ehrgeizige Forschungs- und Entwicklungsprogramme sind in den Ländern China, Frankreich, Japan, Südkorea, oder den USA bereits angelaufen mit dem Ziel, nuklearen Wasserstoff auf den Energiemarkt zu bringen. Auch in der EU gibt es ein entsprechendes Engagement seitens der Forschung, Industrie und Politik, überwiegend im Rahmen einer Beteiligung an nationalen oder europäischen Forschungsprogrammen.

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LIST OF ACRONYMS

ADAM	Anlage mit drei adiabaten Methanisierungsanlagen (Facility with Three Adiabatic Methanation Reactors), Germany
AFCR	Annualized Fixed Charge Rate
AHTR	Advanced High Temperature Reactor, USA
ANL	Argonne National Laboratory, Argonne, USA
APR	Advanced Power Reactor, Korea
ATR	Autothermal Reforming
CCGT	Combined Cycle Gas Turbine
CEA	Commissariat à l'Energie Atomique, France
CHP	Combined Heat and Power
CH ₂ P	Cogeneration of Hydrogen and Power
CLC	Chemical Looping Combustion
CNG	Compressed Natural Gas
DME	Dimethylether
EC	European Commission
EHFCP	European Hydrogen and Fuel Cell Technology Platform
EPR	European Pressurized Reactor
EPRI	Electric Power Research Institute, USA
EU25	European Union of 25 Member States (since May 1 st , 2004)
EVA	Einzelspaltrohr-Versuchsanlage (Single Splitting Tube Test Facility), Germany
EVO	Helium Turbine Power Plant built by Energieversorgung Oberhausen (EVO), Germany
FBR	Fast Breeder Reactor
FC	Fuel Cell
FCV	Fuel Cell Vehicle
FLiBe	Molten salt containing Fluor, Lithium, Beryllium
FP	Framework Programme, EU
FR-MR	Fast Reactor – Membrane Reformer, Japan
FZJ	Forschungszentrum Juelich (Research Center Juelich)
GA	General Atomics, San Diego, USA
GFR	Gas Cooled Fast Reactor
GHG	Greenhouse Gases
GIF	Generation IV International Forum

GRI	Gas Research Institute, USA
GT	Gas Turbine
HCNG	High Compressed Natural Gas
HHLT	Hybrid Hydrogen Process in the Lower Temperature Range
HHV	Higher Heating Value
HHV	Hochtemperatur Helium Versuchsanlage (High Temperature Helium Test Facility), Germany
HIx	Mixture of HI, I ₂ , H ₂ O
HTE	High Temperature Electrolysis
HTGR	High Temperature Gas Cooled Reactor
HTTR	High Temperature Engineering Test Reactor, Japan
HyS	Hybrid Sulfur (Westinghouse)
IAEA	International Atomic Energy Agency
ICE	Internal Combustion Engine
IDC	Inflation and interest rates during construction of an industrial plant
IEA	International Energy Agency
IGCC	Integrated Gasification Combined Cycle
IHX	Intermediate Heat Exchanger
INL	Idaho National Laboratory, Idaho Falls, USA
INPRO	Innovative Nuclear Fuel Reactor and Fuel Cycles
IPSR	Integral Primary System Reactor
ITM	Ion Transport Membrane
JAEA	Japan Atomic Energy Agency, existing since Oct. 1 st , 2005, by merging JAERI and JNC
JAERI, now JAEA	Japan Atomic Energy Research Institute
JNC, now JAEA	Japan Nuclear Cycle Development Institute
JRC	Joint Research Center, European Union
KLAK	Kleine Absorberkugeln (Small Absorber Balls shutdown system)
KVK	Komponenten-Versuchskreislauf (Components Test Circuit)
lbm	Pound (of mass), 1 lbm = 0.45359237 kg
LH ₂	Liquid Hydrogen
LHV	Lower Heating Value
LNG	Liquefied Natural Gas
LOCA	Loss of Coolant Accident
LPG	Liquid Petroleum Gas

LWR	Light Water Reactor
MACRS	Modified Accelerated Cost Recovery System
MCFC	Molten Carbonate Fuel Cell
MED	Multiple Effect Distillation
MHR	Gas Turbine Modular Helium Reactor, USA
MMBTU	Million British Thermal Unit, 1 BTU = 1054.4 J = 2.9288*10 ⁻⁴ kWh
MMI	Methane-Methanol-Iodomethane thermochemical cycle
MTBE	Methyl tertiary-butyl ether
MTOE	Million Ton of Oil Equivalent, 1 TOE = 41,868 MJ
NERI	Nuclear Energy Research Initiative, USA
NFE	Projekt Nukleare Fernenergie (Project Nuclear Long Distance Energy), Germany
NG	Natural Gas
NGNP	Next Generation Nuclear Plant
NHDD	Nuclear Hydrogen Development and Demonstration project, Korea
NHI	Nuclear Hydrogen Initiative, USA
NPP	Nuclear Power Plant
OECD/NEA	Organization for Economic Cooperation and Development / Nuclear Energy Agency
OEM	Original Equipment Manufacturer
O&M	Operation and Maintenance
OTTO	Once Through Then Out
PBMR	Pebble Bed Modular Reactor, South Africa
PBR	Pebble Bed Reactor
PCHE	Printed Circuit Heat Exchanger
PEM	Proton Exchange Membrane or Polymer Electrolyte Membrane
PFHE	Plate Fin Heat Exchanger
PH	Process Heat
PMHE	Plate Machined Heat Exchanger
PMR	Prismatic Modular Reactor
PNP	Prototype Plant Nuclear Process Heat Project, Germany
POX	Partial Oxidation
PSA	Pressure Swing Adsorption
PSI	Paul Scherrer Institute, Switzerland
RCCS	Reactor Cavity Cooling System

SCF	Standard Cubic Feet, 1 scf = 28.317 liters
SCWR	Super Critical Water Reactor
SG	Steam Generator
S-I	Sulfur-Iodine thermochemical cycle
SMR	Steam Methane Reforming
SNL	Savannah River National Laboratory, Aiken, USA
SOFC	Solid Oxide Fuel Cell
SOWE	Solid Oxide Water Electrolysis
SPEWE	Solid Polymer Electrolyte Water Electrolysis
SR	Steam Reformer
STAR-LM / -H ₂	Secure Transportable Autonomous Reactor-Liquid Metal / -Hydrogen
TRU	Transuranium
UPS	Uninterruptible Power Supply
UT	University of Tokyo, Japan
VAT	Value Added Tax
VHTR	Very High Temperature Reactor
WEU	Western Europe

1. INTRODUCTION

The European Union is an association of 25 countries (EU25) with a high degree of industrialization and with extended urban agglomerations. It therefore needs to rely on a secure and economic supply with energy. In addition, there is the interest to increase the supply security by diversification of the primary energy carriers and, at the same time, limit the effects of energy consumption on the environment. The situation in the European Union as predicted for the next 30 years is characterized by a growing demand for energy and, at the same time (after 2010), a decreasing domestic energy production as can be seen from Fig. 1-1 [Martin Bermejo 2004]. While in 2000, 48 % of the energy demand had to be covered by imports, this share will increase to 70 % in 2030, if no additional measures are taken. In addition, this development will push CO₂ emissions in the EU countries to a plus of 14 % compared to the 1990 level, far off the Kyoto commitment of an 8 % reduction.

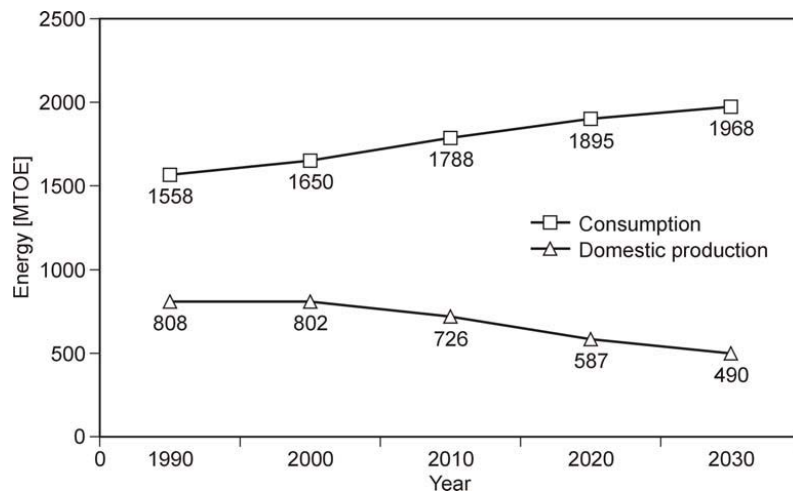


Fig. 1-1: Energy consumption and domestic production [MTOE] in the 25 EU member states
1 TOE (= ton of oil equivalent) = 41, 868 MJ = 11,630 kWh

The principal policy objectives of the European Union are:

- meeting EU Kyoto commitments of an 8 % CO₂ reduction by 2008-2012 compared to 1990 level anticipating the need for a much stronger reduction required by 2015-2025;
- maintaining security of energy supply;
- reducing the dependence on fossil energy imports;
- promoting industrial competitiveness.

Although aware of the major risks of a rapid increase in energy demand worldwide, particularly in emerging economies, and of the uncertainty about how long oil and gas reserves will last, the discussion in the EU on the structure of its future energy economy has not yet reached the top of the political agenda. Main reasons are the diversity in national energy policies and the tendency among the EU member states to consider their energy strategies as a matter of national security. However, a closer collaboration among the EU countries and harmonization of European activities is underway; policy and legislation will be increasingly decided at EU level.

Another important point is the fact that a competition of the different energy carriers is not really given. Market is distorted by the existing questionable automatism of a coupling in the price fluctuations for all energy carriers. There is no rational reason of increasing prices for abundantly available primary energies such as coal or gas, when the oil price is escalating due to resource shortage. A decoupling, however, is necessary for future energy alternatives to keep them affordable.

Nuclear power is one of the major energy sources in Western Europe. From the 25 EU member states, 14 are operating a total of 148 NPP with a capacity of 130.6 GW (as of July 2005) covering more than a third of the total electricity demand. The nuclear share, however, varies significantly between countries: while some countries completely refrain from nuclear, in other countries the nuclear shares range between ~3 % for the Netherlands to ~78 % for France. On the European level, there exists long-term intensive cooperation among the nuclear vendors, utilities and research organizations, not only aiming at an evolutionary development of existing nuclear technology, but also searching for innovative concepts of power plants and components with different and improved safety characteristics. It was the main incentive for the foundation of the MICHELANGELO Network to move away from the fragmentation and isolation of national research efforts and elaborate a common European position on the priorities of future R&D for a sustainable use of nuclear energy within the worldwide activities in this area.

The consideration of nuclear energy is mainly to ensure the security of energy supply and to alleviate the dependency on imports of fossil fuels. The development of indigenous energy sources remains one of the major objectives of energy policy in most countries. Non-electric products from nuclear energy have the potential to improve the countries' energy independence.

Different from electricity consumption which does not imply transport of a medium, non-electricity applications of nuclear energy will not only require a very high safety level with regard to the exclusion of radioactivity release, but also has to show an exclusion of contamination of products and heat transfer media transported to the consumer. Additionally, the first-of-a-kind feature of a new nuclear system in a new application area will raise considerable impediments for a market introduction and will need international support and consortia. It should be noted that any use of nuclear process heat directly reduces significantly CO₂ emissions.

Data from the EU High Level Group show that the projection of hydrogen demands stays on a rather high level for refineries, petrochemicals, ammonia, steel etc. even beyond the scope of the forecast whereas the niches and additional markets for hydrogen are slowly developing with an uncertain slope not before some decades from now. The indicated tensions due to oil price increase might not only favor hydrogen technologies but also other technologies to prolong oil resources by tertiary oil recovery, oil shale processing or coal gasification. This trend was already experienced during the past oil crises showing that these technologies are near to competitiveness if the oil price stabilizes on a slightly higher level. In addition, any progress in CO₂ sequestration will at least prolong the "fossil era" for several decades.

As part of the so-called European Research Area, the "Hydrogen and Fuel Cells Technology Platform" was founded in 2003 to provide guidance on the R&D needs and development issues for both hydrogen and fuel cells. In its draft "Strategic Research Agenda" issued in 2004, the Platform recognizes with respect to H₂ production that efficiency improvement and cost reduction are key areas. Future R&D efforts have been recommended to include the "development of commonly understood reforming and gasification techniques, also high-temperature primary energy systems, such as Generation IV nuclear reactors, and solar-thermal concentrating systems" [EHFCP 2005a].

In this report the major items of a roadmap for the implementation of an industrial production of hydrogen using nuclear energy are given in further detail. The report first provides in chapter 2 a description of the markets for hydrogen, its present conventional production methods (fossil fuel conversion, steam reforming, electrolysis, etc.) Also the scenarios for future use of hydrogen in the EU will be illustrated. This chapter includes also the short term options (next 5-15 years) for

producing hydrogen by electrolysis as well as the long term option of the co-existence of electricity (electrons) and hydrogen (protons) to form complementary, inter-changeable and synergetic options for transferring and storing energy for different end-uses.

Chapter 3 focuses on the nuclear production technologies for hydrogen. In the widely applied methods of steam reforming of natural gas or of other crude oil residua, the necessary process heat which is in the conventional process supplied by fossil fuel, can be replaced by nuclear process heat from an HTGR. This chapter includes a technical description of the coupling schemes defining the technical characteristics of the required components and include the past and ongoing R&D in this field. It is followed by sections dedicated to H₂ production methods which are still at R&D level. Candidate processes for thermal water splitting covering thermochemical (hybrid) cycles and high temperature electrolysis are being described and assessed with regard to feasibility and thermal efficiency issues. On-going R&D efforts in these areas are compiled to identify complementary issues for a global R&D approach.

The coupling of a nuclear reactor to a hydrogen production plant located in a chemical complex requires special attention with regard to safety, regulatory aspects, and licensing to be provided by the EU and national or local authorities. Chapter 4 will contribute to have a common approach of safety issues related with hydrogen such as explosions and fires, confinement and limits of contaminants (e.g., tritium), and reliable isolation of both nuclear and chemical plants. In a similar way, consideration will be given to the modifications of current safety requirements, presently oriented to LWR nuclear plants, to new reactor concepts prone for hydrogen production.

Following the analysis on present and future hydrogen demands, chapter 5 identifies medium & long-term strategies for introducing nuclear hydrogen production into the market. An economic comparison will be made to quantify the benefits of nuclear process heat utilization.

2. CURRENT STATUS AND PROSPECTIVE MARKETS FOR HYDROGEN

2.1. Current and Future Demand of Hydrogen and Transport Fuel

The hydrogen applications are generally divided into stationary applications and transport applications. Note that some references propose a closed synthetic hydrocarbon-CO₂ cycle instead of the hydrogen-water cycle to eliminate difficulties related to the production, transport and use of hydrogen [Bossel 2003]. We use the same definitions as those used by the European Platform for Fuel Cell and Hydrogen Technologies in the “Strategic Research Agenda” [EHFCP 2005a]:

- Stationary applications refer to decentralized power generation, including residential 1-10 kW and community (5-50 kW), public and commercial buildings and industrial (50-500 kW), and large-scale (1 MW and above) applications. They also include niche and power premium applications such as Uninterruptible Power Supply (UPS) and other back-up systems.
- Transport refers to vehicles and different systems for propulsion applications as well as auxiliary power units for vehicles, defence, marine and aeronautic applications.
- Portable applications refer to portable power generators (500 W – 5 kW), light traction (wheelchairs, maintenance robots etc.: 100 W – 5 kW) and device-integrated supply.

The biggest consumers that currently employ almost 100% fossil resources and that could in principle use nuclear hydrogen or synthetic hydrocarbons instead were identified in Table 2-1. All these sectors are linked to both population increase and gross national product.

Table 2-1: Major areas of energy consumption

Application	Current annual consumption [10^{17} J/yr]	Reference
Total hydrogen production (world)	70	[IEA 2003]
Ammonia production for fertilizers from natural gas (world)	29.8	[NOU 2004]
Hydrogen for refining crude oils from natural gas (world)	22	[NOU 2004]
Passenger transport (EU25)	60.5	[IRF]
Small-scale combustion installations (domestic heating, hot water etc.) (EU25)	120	[Pye 2004]
Air transport (USA)	30	[Locke 2005]
Air transport (Germany)	3	[Locke 2005]
Air transport (UK)	5.1	[Locke 2005]
Air transport (Canada)	2.4	[Locke 2005]

2.1.1. Stationary

2.1.1.1. Large-Scale Industrial Applications

Current global hydrogen production is estimated by the International Energy Agency at around $7 \cdot 10^{18}$ Joule per year corresponding to ~ 50 million t/yr (HHV) or ~ 550 billion Nm^3/yr , which is about 2 % of the world's total energy consumption [IEA 2003]. Almost all hydrogen on earth is found in compounds, mainly in combination with oxygen as water or in combination with carbon as organic substances. Contemporary hydrogen production is primarily based on the extraction from fossil resources. It is in first place the reforming of natural gas (48 % of the world's production), but also processes like partial oxidation of oil (30 %) or the gasification of coal (18 %) [Scholz 1992]¹. Another major source is the chlor-alkaline electrolysis (2 %), where hydrogen is generated as a side product, or in the chemical industries the off-gas from refinery processes. Still at a minor scale and in the demonstration phase is biomass gasification to produce a hydrogen and/or methane rich fuel gas.

As is shown in Fig. 2-1, about half of the total hydrogen is used for producing ammonia (fertilizer), 37 % is used in petroleum refining processes (tendency increasing due to heavier crude oils), 8 % is used in methanol production, 1 % is used as a fuel in space programs while the residual 4 % is used for other purposes [NOU 2004]. Most important processes in refineries are steam reforming of natural gas (59 %) followed by off-gas purification (35 %) [Ranke 2004]. The market for hydrogen, captive (consumed at production site) which covers 95 % of all hydrogen, and merchant, is expected to grow at a rate of about 4 %/yr. The (small) merchant H_2 market alone is currently growing at an even faster rate of around 10 %/yr [TIG 2003].

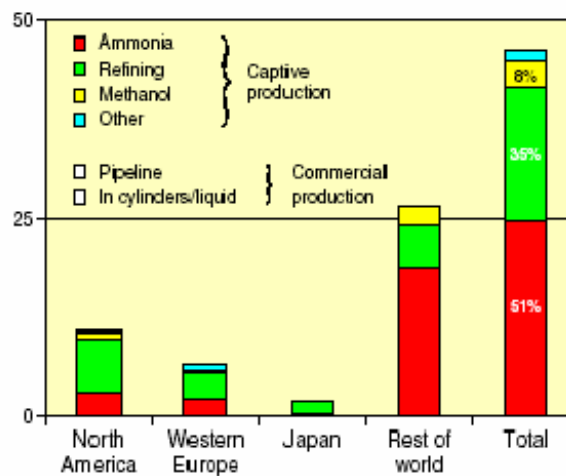


Fig. 2-1: Worldwide hydrogen production (in million tons)

Only 1 % of current hydrogen production is used for energy purposes although in principle, hydrogen could be used for energy purposes in industry in the future, for example as feed for industrial boilers and process heaters [NA 2004].

¹ Another set of figures was found in [NOU 2004], according to which from the total hydrogen production, 85 % is based on natural gas steam reforming while 7 % is derived from oil, 4 % from coal, and 4 % from electrolysis.

The combined hydrogen consumption of the fertilizer and petrochemical industry currently produced through steam reforming by natural gas corresponds to $7 \cdot 10^{18}$ J/yr $\times 0.48 \times (0.5+0.37)$, equivalent to $9.27 \cdot 10^{10}$ W.

To illustrate this with orders of magnitude for potential nuclear power plant demand, we assume that this hydrogen is supplied by nuclear power. Optimistically and simplifying we assume for a High Temperature Gas Cooled Reactor (HTGR) a thermal power of 600 MWt, a thermal to hydrogen conversion efficiency of 50 % and no other losses, and for a European Pressurized Reactor (EPR) an electric power of 1600 MWe, an electric to hydrogen conversion efficiency of 75 % and no losses.

To produce the hydrogen for fertilizers and crude oil refining, 309 HTGRs or 77 EPR would be required. Both sectors are growing, the fertilizer industry due to demand from emerging countries and the petrochemical industry due to heavier crude oil feeds. These numbers illustrate clearly the enormous existing market potential for nuclear hydrogen including the savings in corresponding CO₂ emission and other pollutants. As hydrogen from natural gas is the main competitor in this area, it is understood that nuclear has to compete with natural gas.

2.1.1.2. Small-Scale Combustion Installations

According to [Pye 2004], the energy used within EU25 for small-scale combustion installations amounts to $12 \cdot 10^{18}$ J/yr. This heat is used mainly for the heating of buildings (residential and corporate), hot water and boilers for small industrial applications including agriculture. A significant amount of this heat could be replaced by hydrogen or electricity whatever is more economic. Both can be provided by nuclear power (1268 HTGRs or 317 EPRs for hydrogen).

Different types of fuel cell systems are currently being demonstrated and introduced commercially in sizes from a few kilowatts and up to megawatt sizes often combining the local production of electricity with the use of waste heat for producing hot water and residential heating.

For large generators today's fuel cell systems do not offer higher electrical efficiencies than other technologies that are already on the market such as gas turbines. However, in the future, fuel cell systems are expected to bring about higher electrical efficiencies. At the utility scale, hybrid systems integrating fuel cell systems and gas turbines could bring systems using natural gas with electric efficiencies greater than 65 % and such systems are expected to become cost competitive with competing generating technologies within the next decades [NA 2008]. By comparison, current state-of-the-art power plants using combined-cycle technology combining gas turbines and steam turbines have electrical efficiencies of around 55-60 % and are currently cheaper (400-500 €/kW) than fuel cell systems (2500-8000 €/kW). Technologies available for distributed power generation, with electrical capacities of less than 60 MW, include gas turbines, reciprocating engines, micro-turbines, wind turbines, biomass-based generators, solar photovoltaic systems and fuel cells. Some studies foresee that fuel cells might initially emerge as distributed generators in applications where users are willing to pay an extra margin for reliable energy generators. In the USA, 10.7 million distributed generators are in place, of which 99 % are small emergency/standby reciprocating engines that are not interconnected with the grid. The market for distributed generation is typically in the commercial sector in applications where reliable energy is needed or in remote locations where grid power is not available.

Another area of interest for fuel cells is small-scale distributed CHP units to be installed in residential buildings. For example, 1.2 kW PEM fuel cells are currently being introduced in Japan in limited volume by Ballard Power Systems [NA 2004], and the company Vaillant is currently installing a number of PEM fuel cells in Europe [NOU 2004]. SOFCs can also be made available for small-scale CHP in the future [NOU 2004].

2.1.2. Transport

As shown in Table 2-1, transportation is another big consumer of energy that is currently provided almost exclusively by fossil fuels. Just an example to get an idea of the magnitude of the problem for hydrogen to penetrate the market of conventional transportation fuels: The USA consume presently per day [Wilson 2003]:

2,610,000 t of crude oil
1,030,000 t of gasoline
497,500 t of diesel / heating oil
230,000 t of jet fuel

Only considering the approximate 1 million tons of gasoline per day, this amount corresponds to 339,500 t/d of H_2 on an equivalent energy basis. Assuming commercial electrolysis for its production, it translates into a daily demand of 3 million t of clean water and consuming about 845 GW of electric power which is almost equal to the total US power generating capacity (as of 2003) [Wilson 2003].

The transportation sector is further strongly expanding in emerging countries and still growing even in industrialized countries including the EU25. It not only comprises passenger vehicles, commercial vehicles and aviation but also maritime transport. The use of hydrogen as an energy carrier will be widely established in the transportation sector. Only passenger transport and aviation are given here as examples.

2.1.2.1. Light Passenger Vehicles

The EU25 countries dispose of a passenger car fleet of currently 212 million with an average of 460 cars per 1000 inhabitants [IRF]. Assuming an annual mileage of 12,000 km per car at a consumption of the equivalent of 7 l Diesel/100 km (34,000 kJ/l Diesel), every single car would consume $2.86 \cdot 10^{10}$ J/yr corresponding to an installed power of 906 W/car.

The total EU25 energy consumption for cars would then amount to approximately $6.05 \cdot 10^{18}$ J/yr or an installed power of $1.92 \cdot 10^{11}$ W. Assuming again the same reactor power and conversion efficiencies for hydrogen, this energy consumption would have to be produced by 640 HTGRs or 160 EPRs.

To this value, the fuel for 22 million European commercial vehicles must be added, which on average have both significantly higher annual mileage and a higher consumption than passenger cars so that the order of magnitude for fuel consumption is probably similar to the one for light passenger vehicles.

In recent years, car producers have manufactured a range of prototype internal combustion engine (ICE) and PEM fuel cell passenger cars using hydrogen as fuel, PEM cars being more energy efficient than ICE model. Currently many demonstration projects are being carried out, or are being prepared, in Europe, Japan and the United States. It has not yet been possible to develop a hydrogen storage medium allowing hydrogen-fueled PEM fuel cell cars to have range capabilities comparable to current baseline gasoline and diesel vehicles.

Other studies compare hydrogen-fueled PEM fuel cell vehicles to battery electric vehicles, showing that battery electric vehicles that were on the market in the 1990s use less energy on a tank-to-wheel basis than current fuel cell vehicles. However, current battery electric vehicles offer shorter range and take longer time to refuel than baseline gasoline and diesel vehicles and are not cost-competitive because batteries are expensive and have a short lifetime.

Proponents of battery electric vehicles hold that future advancements in battery technology may allow construction of better and cheaper battery electric vehicles or plug-in hybrid electric vehicles. Consequently, hydrogen fueled PEM fuel cell vehicles will probably see intensifying competition from advanced diesel and gasoline hybrid electric vehicles as well as battery electric and other alternative vehicle types and fuels. Furthermore, a prerequisite for the widespread introduction of hydrogen-fueled vehicles is the building-up of a hydrogen production and distribution system.

In recent years, vehicle producers have manufactured a range of prototype fuel cell buses using pure hydrogen as fuel. Another way of using hydrogen in buses is natural gas-fueled ICE buses using a blend of natural gas and hydrogen, so-called hythane, with only minor adjustments to the software in the bus. Use of hythane reduces emissions of greenhouse gases and other pollutants as compared to using natural gas, and the vehicles consume less fuel per kilometer. According to a Norwegian comparative study of energy consumption and emission characteristics of advanced buses in 2020, PEM fuel cell hybrid electric buses are expected to consume around 7 MJ per kilometer and thereby have lower tank-to-wheel fuel consumption than advanced diesel hybrid electric buses expected to be using around 9.5 MJ per kilometer. However, on a well-to-wheels basis the hydrogen-fueled PEM fuel cell bus uses more primary energy than the diesel hybrid, and related emissions of greenhouse gases depend on the type of primary energy used. In principle, the PEM fuel cell bus offers the potential of eliminating emissions of greenhouse gases and other pollutants if the production of hydrogen is based on renewable energy, and fuel cells are furthermore favorable because they eliminate emissions of NO_x and particles in the usage phase [NOU 2004].

2.1.2.2. Marine

Fuel cells are envisaged for use in marine applications. Small demonstration projects test fuel cells in small passenger boats and submarines. Hydrogen-fueled fuel cells may be used in ships that do not need long ranges and can be filled up frequently whereas ships that need longer ranges would need to use fuel cells based on fuels with higher energy densities, such as liquid natural gas, methanol or ethanol. A major driver for using fuel cells in ships could be that fuel cells offer the possibility of reducing emissions drastically. Marine transport worldwide currently contributes to 14 % of global nitrogen oxides emissions and 7 % of sulfur emissions. The European project FCSHIP – Environmental Impacts and Costs of Hydrogen, Natural Gas and Conventional Fuels for Fuel Cell Ships – indicates possible future concepts and prepares regulations for the design and operation of fuel cell technology in the marine market.

2.1.2.3. Aviation

Aviation is yet another strongly growing energy consuming sector where hydrogen or synthetic hydrocarbons could play a significant role in the future. Fig. 2-2 shows the major routes on which aviation fuel is consumed [Locke 2005]. The strong increase in the sector comes mainly from emerging countries.

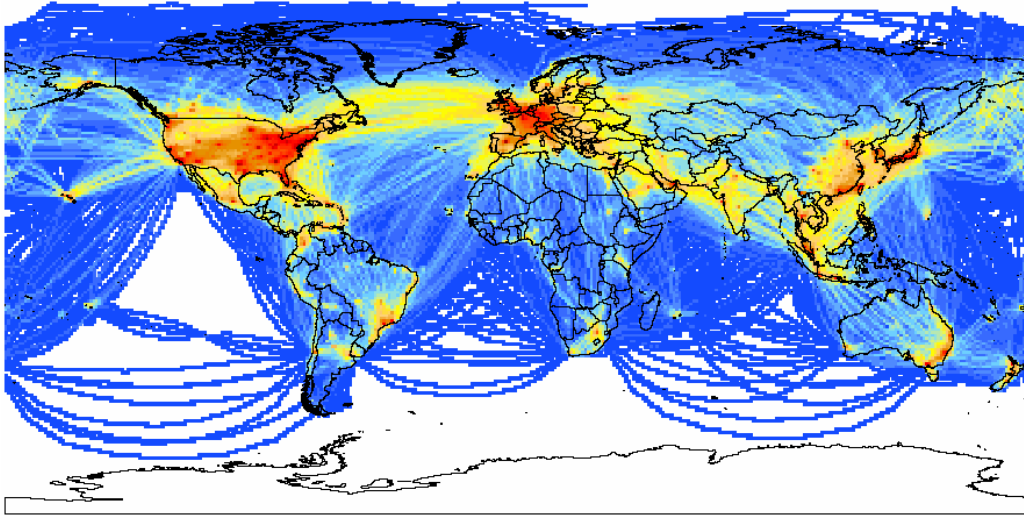


Fig. 2-2: Example plot of fuel burn on a 1 degree by 1 degree world grid [Locke 2005]

As examples, the following data is provided in [Locke 2005] for 2004: The total US aviation fuel burn amounted to $3 \cdot 10^{18}$ J/yr, which could be satisfied by 317 HTGRs or 79 EPRs. Germany has a need that is by a factor 10 lower. Data for other countries can be found in Table 2-2.

2.1.3. Summary of Potential Hydrogen Consumers and Nuclear Requirements

In Table 2-2, a summary is given on the identified main fossil fuel consumers and the estimated number of nuclear reactors when replacing fossil by nuclear hydrogen or synthetic hydrocarbons.

Table 2-2: Summary of main fossil fuel consumers and estimated requirements for nuclear power plants to replace fossil

Application	Current annual consumption [10^{17} J/yr]	Estimated number of NPP	
		HTGR ¹	EPR ²
Total hydrogen production (world)	70	309	77
Ammonia production for fertilizers from natural gas (world)	29.8	233	58
Hydrogen for refining crude oils from natural gas (world)	22.0	172	43
Passenger transport (EU25)	60.5	640	160
Small-scale combustion installations for domestic heating, hot water etc. (EU25)	120	1268	317
Air transport (USA)	30	317	79
Air transport (Germany)	3	32	8
Air transport (UK)	5.1	54	13
Air transport (Canada)	2.4	25	6

Assumptions:

¹ HTGR (600 MWt, thermal to hydrogen conversion efficiency 50 %)

² EPR (1600 MWe, electric to hydrogen conversion efficiency 75 %)

2.1.4. Transition Scenarios

Several transition scenarios for EU25 are described extensively in [EHFCP 2005b] and [Castello 2005], yet with focus on the deployment of fuel cell technology. By 2020, fuel cell applications as given in the following Table 2-3 were mentioned with road transport causing by far the largest demand:

Table 2-3: Projected main users of hydrogen by 2020 [EHFCP 2005b]

	Portable FC for handheld electronic devices	Portable generators and early markets	Stationary FC for CHP	Road transport
Cumulative FC sales until 2020	$\gg 250 \cdot 10^6$	600,000	$4\text{--}8 \cdot 10^5$	$1\text{--}5 \cdot 10^6$
Average power	15 W	10 kW	20 kW	80 kW
Total power	$\gg 3.75 \text{ GW(e)}$	6 GW(e)	8-16 GW(e)	80-400 GW(e)
HTGR ¹	$\gg 12.5$	20	27-53	267-1333
EPR ²	$\gg 3$	5	7-13	67-333

Assumptions:

¹ HTGR (600 MWt, thermal to hydrogen conversion efficiency 50 %, no losses)

² EPR (1600 MWe, electric to hydrogen conversion efficiency 75 %, no losses)

The most immediate possibility for use of nuclear hydrogen - and overlooked in most strategic documents - is certainly stationary and concerns the fertilizer and petrochemical industry, which not only consume hydrogen but also significant amounts of process heat. This combination is also easiest to implement as a high demand can be combined with large production and short transport distances, supposing that safety requirements can be met. The combination can possibly be considered economic already with low-temperature electrolysis, on the condition that the hydrogen is produced from cheap base-load electricity with intermediate storage [Miller 2005]. The combination with refineries would have a two-fold positive effect on the natural gas and crude oil prices because it eliminates natural gas consumption in the refinery (the gas could be used more efficiently in other applications) while producing lighter and cleaner liquid fuels from heavy crude oils, oil shales and tar sands that would otherwise be difficult to process.

The next possible extension of nuclear power would then concern passenger transport for which NPP can produce either electricity, hydrogen or synthetic hydrocarbons. The latter is particularly suitable for aviation.

The replacement of fossil fuels in small-scale combustion installations is probably the most difficult (although most efficient) as this would require a fully-fledged infrastructure for nuclear produced combustion fuel, except for the cases where electricity could be used instead.

Fig. 2-3 shows a projection of hydrogen production processes although no recommendation or prediction is given for the phase-out of fossil-based production processes.

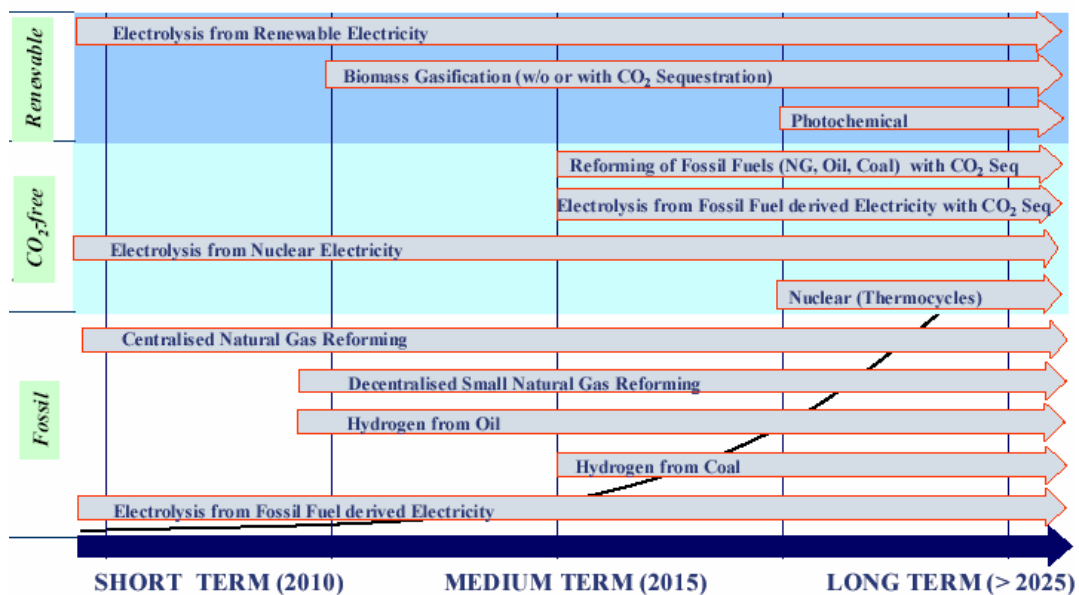


Fig. 2-3: Maturation of hydrogen production pathways [HyNet 2004]

The HyNet roadmap [HyNet 2004] predicts that electrolysis from nuclear will be among the first significant sources of hydrogen. The fact that low-temperature electrolysis from base-load electricity with intermediate cavern storage of hydrogen makes economically sense was shown in [Miller 2005] for the specific case of Canada.

Given the sheer number of NPP required for significant nuclear hydrogen or synthetic hydrocarbon production, and given the growth rates of the worldwide primary energy demand, it is clear that an increase in the nuclear contribution can only be gradual. The pressure for building NPP for hydrogen production is a direct function of the level and fickleness of natural gas prices as the main competitor. It is also clear that nuclear alone will be unable to produce the required energy alone: the capacity for building NPP in the world is too low and the lead-time to construction too long to satisfy the demand, at least in the short and medium term. Even if a hundred new NPP could be built annually worldwide for the next 10 years, the energy demand will still be higher than the supply. For a lasting effect of fissile resources, the use of the U-Pu and Th-U fuel cycles, both requiring reprocessing, is definitely mandatory.

In conclusion and independent on any detailed projection of energy demand and mix of energy sources, it seems almost impossible to question the secured long-term potential and need for nuclear power to penetrate into the non-electricity market.

2.2. Current Hydrogen Production Methods and Efficiencies

The various process paths from the primary energy to the end product and secondary energy carrier hydrogen is illustrated in Fig. 2-4 encompassing both the CO₂ emitting extraction processes from hydrocarbons and the CO₂ emission free water splitting processes.

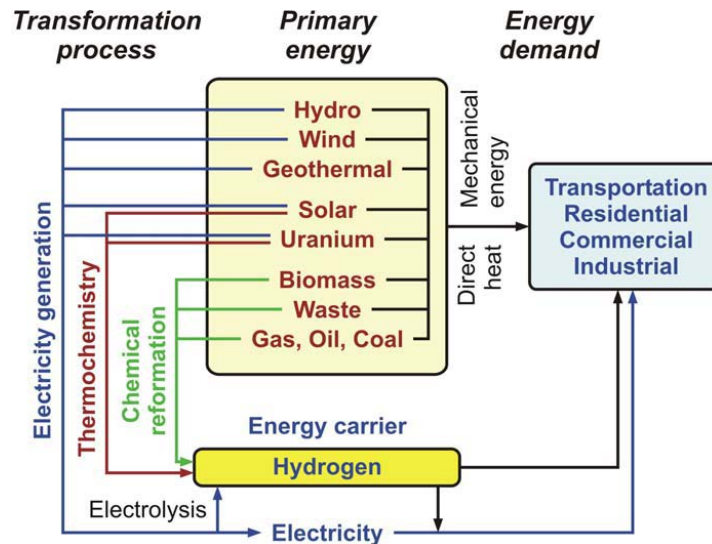
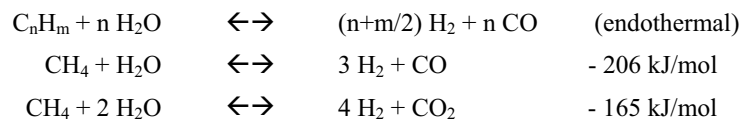


Fig. 2-4: Routes of hydrogen production [PSI 2004]

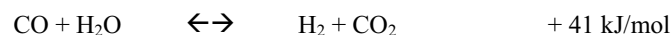
2.2.1. Steam/CO₂ Reforming of Natural Gas

The steam reforming process is the catalytic decomposition of light hydrocarbons (e.g., methane, natural gas, naphtha) to react with steam and resulting in a hydrogen rich gas mixture. A processing scheme is given in Fig. 2-5.

The reforming reactions are endothermal running at high temperatures > 500°C. Steam methane reforming (SMR) takes place at typically 850°C, and at pressures > 2.5 up to 5 MPa in the presence of an iron or nickel catalyst.



In order to increase the output of hydrogen and to avoid carbon deposition due to the Boudouard reaction, the carbon monoxide is catalytically converted in the exothermal water-gas shift reaction with steam according to



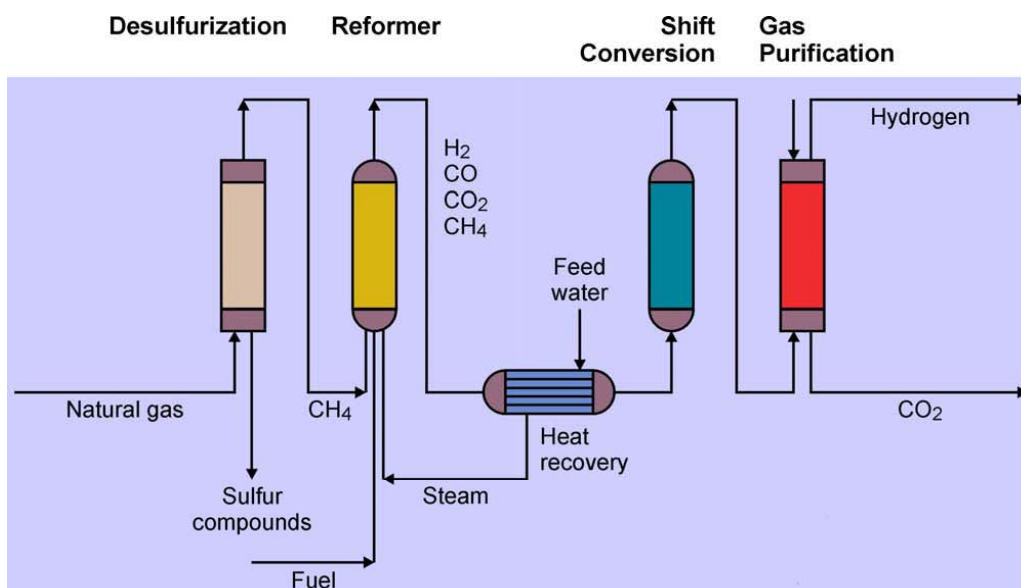


Fig. 2-5: Processing scheme of steam-methane reforming

The feed gas is desulfurized to protect the catalyst inside the reformer tubes. The reformer tubes in the furnace are heated from the outside by burning a part of the natural gas. The main processes of heat transfer are radiation and convection. Flue gas with temperatures above 1300°C passes the furnace and is used in a waste heat utilization step to produce steam and to preheat the feed gas.

The equilibrium composition of the reformer gas, which is a mixture of H_2 , CO , CO_2 , residual steam, and still unreformed feedstock, is strongly depending on the fuel characteristics, the steam-to-carbon ratio, outlet temperature and pressure, which are chosen according to the desired products. High reforming temperatures, low pressures and high steam to methane ratio favor a high methane conversion. A minimum H_2O/CH_4 ratio of around 2 is necessary to avoid carbon deposition on the catalyst which would make it inactive. If excess steam is injected, typically 300 % away from the stoichiometric mixture, the equilibrium is shifted towards more CO_2 (water-gas shift reaction, exothermal) at temperatures of 300-400°C increasing the H_2 yield and reducing the undesired production of carbon (Boudouard reaction).

The hydrogen gas needs to pass further purification steps to realize a purity of > 99 % before being used, e.g., in fuel cells. The unwanted constituents CO_2 and others are removed from the gas mixture by pressure swing adsorption (PSA) or membrane separation. The residual gas is added to the fuel gas stream to supply the process heat for the reformer. Other tail gases may be used for heat requirements. The CO_2 generated is contained in the PSA reject gas and usually vented to the atmosphere; however, a separation process is possible to capture it. Overall, the different process steps need considerable amount of energy. The total balance for such a plant is that 1 Nm^3 of methane allows the production of 2.5 Nm^3 of hydrogen, which corresponds to an overall efficiency of the process of around 65 %. It is rather difficult to get much higher efficiencies in practice.

If the steam is completely or partially replaced by CO_2 , the composition of the synthesis gas is shifted towards a larger CO fraction. The CO_2 can be either imported or taken from the reformer

outlet. The catalytic reforming of methane with CO_2 offers an environmental advantage, because two greenhouse gases are combined resulting in a product gas mixture which might be more favorable for certain applications like the synthesis of oxygenated chemicals. Major drawbacks are the rapid deactivation of conventional catalysts and the relatively high soot formation (methane cracking).

Steam reforming of natural gas is a technically and commercially well established technology on industrial scale and currently the most economical route. Reforming technology is mainly used in the petrochemical and fertilizer industries for the production of so-called “on-purpose” hydrogen. The conventional process requires the stages of desulfurization, synthesis gas production, CO shift conversion, and purification by pressure swing adsorption. Optimum pressure range is 2.5-3 MPa resulting in a hydrogen yield of 86-90 % [Uhde 2003]. Large steam reformer units with up to about 1000 splitting tubes have a production capacity of around 130,000 Nm^3/h (see Fig. 2-6). At the Fortum refinery in Porvoo, Finland, a hydrogen plant with a capacity of 153,500 Nm^3/h or 13.8 t/h of H_2 , corresponding to a stored power of 550 MW (based on HHV) will start operation in 2006. This plant is flexible in feedstock in that it can convert besides natural gas also refinery off-gas or liquid propane. Future reformer plants are designed to produce 237,000 Nm^3/h . There is even an announcement by Praxair to construct a plant with 354,000 Nm^3/h capacity [Forsberg 2005b]. Commercial large-scale SMRs produce hydrogen at an efficiency of about 75 % and a CO_2 intensity of 9.5 kg per kg of H_2 produced [EHFCP 2005a].

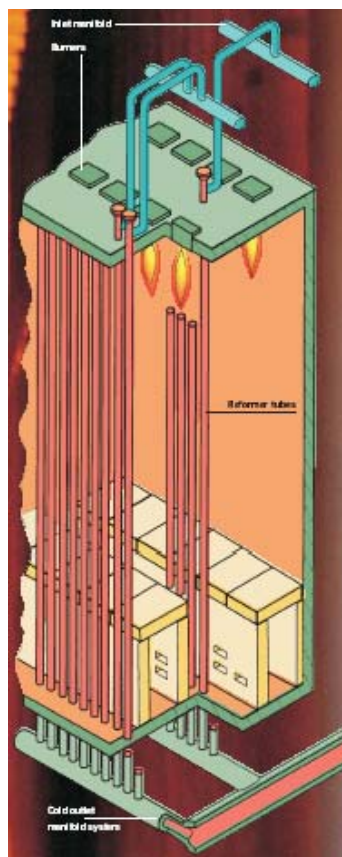


Fig. 2-6: Uhde steam reformer for light hydrocarbons [Uhde 2003]

If light hydrocarbons are used as fuel, sometimes a pre-reformation is helpful to operate the tubes under the same conditions with methane as feed gas. Naphtha-based plants are applying a pre-reforming step at a lower temperature and using a more active catalyst, partially converting the hydrocarbons to methane, hydrogen, and carbon oxides. This allows alternate feed operation and makes better use of heat in the overall process. Feed and fuel consumption savings are approx. 5 % [Uhde 2003]. Steam reforming of heavier hydrocarbons is possible, but requires a more complex process equipment and is therefore only little applied.

Characteristic data of hydrogen production by steam reforming of natural gas are given in Table 2-4 for two different plant capacities, 1000 and 100,000 Nm³/h showing that with increasing plant size, the specific costs of H₂ are significantly reduced. No major further reduction is anticipated due to only minor technological progress to be expected in future [Dreier 2000].

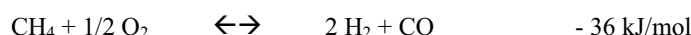
Table 2-4: Hydrogen production by steam reforming and its prospective

	State of the Art		Scenario 2025	
Technical Data				
Rated power natural gas [kW]	4500	405,000	4275	385,000
Hydrogen production [Nm ³ /h]	1000	100,000	1000	100,000
System pressure [MPa]	1.6	3.0	1.6	3.0
Water demand [m ³ /h]	2.1	58	2.1	58
Efficiency [%]	67	74	70	78
Life time [yr]	20	20	20	20
Utilization [h/yr]	8000	8000	8000	8000
Cost				
Investition [€/kW of H ₂]	675	335	675	335
Specific total cost [€ Ct/kWh of H ₂]	3.2	2.3	3.1 ¹⁾	2.2 ¹⁾

1) Figures are based on a natural gas prize of 1.4 € Ct/kWh. They do not include potential CO₂ taxation or sequestration cost.

2.2.2. Partial Oxidation and Autothermal Reforming of Hydrocarbons

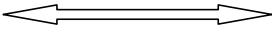
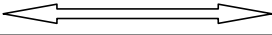
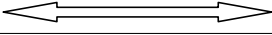
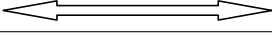
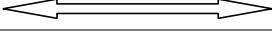
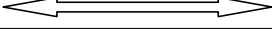
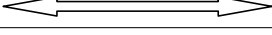
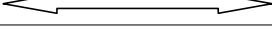
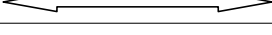
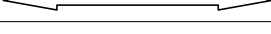
The partial oxidation (POX) of carbonaceous feedstock at the presence of water is also a conversion process at high pressures and high temperatures (950-1100°C) which produces synthesis gas (hydrogen plus carbon monoxide) and maximizes H₂ yield, if followed by the water-gas shift reaction. The H₂ mainly originates from the water, while the carbon in the feedstock provides the energy to split the water. By adding oxygen, a part of the feedstock is burnt in an exothermal reaction.



Downstream equipment is needed to remove the heat generated. The oxygen needed is typically provided by an air separation plant. POX can also be performed as catalytic reaction.

The combination of the POX process with endothermal steam reforming may lead to reactions without heat input from the outside (autothermal reforming - ATR) achieving higher efficiencies. In such a reformer, the reactor consists of a combustion zone where the heated-up reaction gas mixture is directly transferred into a fixed-bed catalytic steam reforming zone. An overall evaluation of important features of these various reforming processes is given in the following Table 2-5 [Hikita 2005, Riis 2005]:

Table 2-5: Evaluation of reforming processes

	Steam reforming	Autothermal	Partial oxidation
Start-up	Slow		Fast
Size	Large		Small
Hydrogen content	High		Low
Temperature control	Easy		Difficult
Emissions	Low		High
Efficiency	High		Low
Complexity	High		Low
Hydrogen quality	High		Low
Cost of large plant	Low		High
Cost of small plant	High		Low

The POX process has the advantage of accepting all kinds of heavy hydrocarbon feed such as oil, residues, coal, or biomass. The resulting synthesis gas with a H_2/CO ratio of ~ 2 (compared to > 3 for SMR) makes methanol synthesis an ideal follow-on process. Non-catalytic POX takes place at temperatures of 1200-1450°C and pressures of 3-7.5 MPa (Texaco process), the catalytic POX at around 1000°C. Efficiencies of about 50 % are somewhat less compared to SMR because of the higher temperatures involved and problems with the heat recovery. Disadvantages are the need of large amounts of oxygen, catalyst deactivation due to carbon deposition, the by-product CO which requires the shift reaction, the need for gas purification stages, and, if methane is used as feed, the possibility of runaway reactions or explosions due to hot spot formation.

Catalytic partial oxidation of heavy oil and other hydrocarbons is a large-scale H_2 production method which is generally applied when generating synthesis gas from heavy oil fractions, coal, or coke. It may become competitive, when cheap oxygen is available. It can be conducted in both monolith reactors and in fluidized bed reactors, but also in fixed bed micro-reactors. Plants usually include also air decomposition, unit size also in the order of 100,000 Nm^3/h . Commercial technologies available are the gasification processes of Texaco and Shell. Capacities of combined autothermal reformers (ATR) (see Fig. 2-7) are typically between 4000-35,000 Nm^3/h , a range where “normal” steam reforming exhibits high specific investment. Small-sized units of POX reforming for mobile applications are presently under development.

ATR technology was developed since the late 1970s with the goal to have the reforming step in a single adiabatic reactor. Preheated feedstock is gradually mixed and burnt in the combustion chamber at the top, where partial oxidation takes place. Steam is added to the feed to allow premixing of CH_4 and O_2 . The steam reforming step is done in the lower part of the reactor. ATR requires 10-15 % less energy and 25-30 % less capital investment [Bharadwaj 1995].

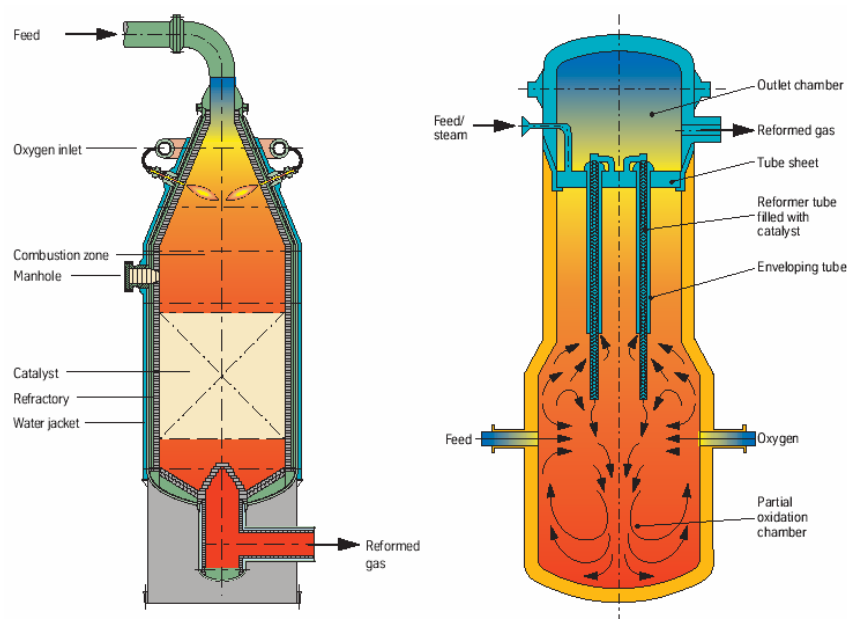


Fig. 2-7: Autothermal Reformer (ATR) (left) and Combined ATR (right) [Uhde 2003]

ATR is mainly used in large-scale plants. But there are also smaller units for local H_2 production with a capacity around $150 \text{ Nm}^3/\text{h}$. Research in reforming technologies is concentrating on finding the right balance of fuel, air, and water flows for optimal processing. If CO_2 capture technology were to be added, ATR is of advantage due to the fact that the CO_2 recovered from a separation step which works optimally at 0.3 MPa, thus saving compression cost.

For on-board reforming, multi-fuel processors in the 50 kW range have been developed. Here methanol appears to be the more attractive fuel, because it operates at lower temperatures and is more tolerant to intermittent demand. Gasoline or LPG reforming would even be more practical, since this infrastructure exists already and could allow the introduction of respective vehicles even at a lower number. Types of reactor applicable to small reformers are fluidized bed reactor, membrane reactor, short-contact reactor, heat exchange reactor, micro-reactor. Reformer unit in the very low power range like for mobile/portable fuel cell applications are presently developed and tested. The Center for Fuel Cell Technology in Duisburg, Germany, for instance, presents a 1 kW reformer for LPG or natural gas with 75 % efficiency. This unit with a 1.5 l volume employs an integrated catalytic monolith replacing the conventional catalyst pellets. The PSI, Switzerland, has operated smaller reformer units based on solar energy as the primary heat input. The facilities were a 480 kW(th) high-pressure (0.8-1 MPa) tubular reactor and a 280 kW(th) low-pressure (0.1-0.3 MPa) volumetric reactor, both operated at $\sim 800^\circ\text{C}$ with a Rh catalyst for CO_2 reforming.

Tandem reforming is the combination of a gas-heated reformer and an oxygen-fired autothermal reformer (see Fig. 2-7). If neither oxygen nor steam is available, the facility needs air separation and steam generator units.

A modification of the POX steam reforming process has been suggested which leads to an inherent capture of the CO_2 generated during fuel combustion. The principle of the so-called “Chemical Looping Combustion” (CLC) is to have separate reactors, an “air reactor” where oxygen is extracted from air by means of a solid oxygen carrier, typically a metal, and a “fuel reactor”

where the metal oxide is reduced to its initial state and recirculated. CLC has the advantage of a reduction of the exergy loss during fuel combustion, while the generated CO_2 is separated from the (diluted) air steam and thus can easily be sequestered [Ryden 2006].

Modern steam-methane reformers often use more than one catalyst at different temperatures to optimize the H_2 output. Heat losses and thermodynamic (Carnot) limitation lead to an overall efficiency of about 80 %. Catalytic autothermal reforming is ideal for fuel cell systems due to its simple design, low operation temperatures, flexible load, and high efficiency.

In plate-type reformers, plates are arranged in a stack with one side being coated with a catalyst and supplied with the reactants. These reformers are more compact, show a faster startup and a better heat transfer and therefore a higher conversion efficiency.

Advanced reforming techniques will operate at reduced reaction temperatures by means of micro-porous ceramic membranes. Membrane modules made of palladium-based alloy and a nickel-based catalyst can perform steam reforming reaction, shift reaction, and H_2 separation simultaneously (see Fig. 2-8), i.e., without shift converter and PSA stages. Therefore the system is compact and may provide higher efficiencies. The simultaneous processes allow to lower the reaction temperature down to around 550°C posing less stringent requirements to the materials. The temperature reduction is possible because part of the heat for the endothermal reforming reaction is provided by the exothermal shift reaction. Net heat required is 165 kJ/mol. Technical feasibility of the membrane reforming system was demonstrated by the Tokyo Gas Co., Japan, with test runs up to 1500 h achieving a hydrogen production rate of $15 \text{ Nm}^3/\text{h}$ and a 76 % conversion of the natural gas [Hori 2004]. Tests with a production rate of $40 \text{ Nm}^3/\text{h}$ were also conducted. Catalysts and the separation membranes are the key components which still have potential for further improvement and optimization.

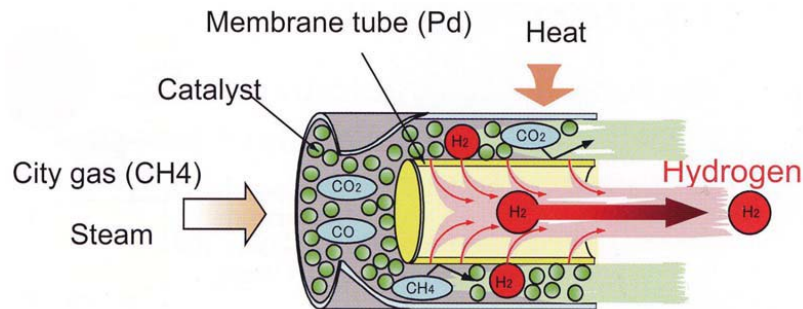


Fig. 2-8: Membrane reformer [Hori 2004, Hikita 2005]

For the high temperature range, inorganic membranes (ceramics, metals) are under development. They allow new concepts which may make the stages of air separation, POX, or PSA obsolete. "Ion Transport Membranes", ITM, with their stable oxygen defect crystal structure are operated at $> 700^\circ\text{C}$ and allows only oxygen ions to move through the membrane, which is gas-tight for all other gases. Due to the charge separation, the material must be conducting at the same time. Palladium coated metal composite membranes promise advantages in terms of mechanical strength, higher H_2 flux, high selectivity, and reduced material cost. H_2 permeation through a Pd membrane is a complex multi-step activated process typically in the range of $300\text{-}600^\circ\text{C}$. Pd membrane combines the operation of the separation unit and shift reaction in one stage, advantageous particularly for small reformers [Ranke 2004]. Conceptual designs promise cost reduction in the generation of high pressure H_2 .

2.2.3. Gasification of Coal

Coal is a solid with a high carbon content and an approximate 5 % content of hydrogen. The conversion of coal into a gas is realized by means of a gasification medium which reacts with the coal at temperatures $> 800^{\circ}\text{C}$. All organic constituents will be converted at long enough residence times. The gasification medium is either steam (steam-coal gasification) or hydrogen (hydro gasification). The process typically involves the injection of air or oxygen into the gasifier, where a part of the coal is directly burnt (POX) allowing for an autothermal reaction.

In the steam-coal gasification process, two consecutive processes take place, the partial oxidation or pyrolysis reaction, where all volatile constituents of the coal are rapidly expelled, and the much slower heterogeneous water gas reaction, where the residual organic solids are converted to synthesis gas ($\text{H}_2 + \text{CO}$) with some CO_2 and steam. The H_2 fraction can be further increased in the shift reaction. Synthesis gas output is optimal at high temperatures and low pressures. Its heat must be quickly withdrawn to avoid reverse chemical reactions.

Various types of gasification processes on a large scale exist such as Lurgi, Winkler, Koppers-Totzek, Texaco (see Fig. 2-9), which differ by the type of reactor, temperature and pressure range, grain size of the coal, and its residence time. Partial oxidation of pulverized coal by oxygen and steam in a fluidized bed takes place at about atmospheric pressure, where 30-40 % of the coal are transformed to CO_2 to supply splitting energy of water. The reaction rate strongly increases with temperature; typically temperatures up to 2000°C and pressures up to 3 MPa are selected. Main disadvantages of coal gasification are the handling of solid material streams and the large amounts of CO_2 , SO_2 , and ash requiring a complex cleaning system. New coal gasification techniques, however, can also be linked with a CLC process to reduce the impurity level in the synthesis gas.

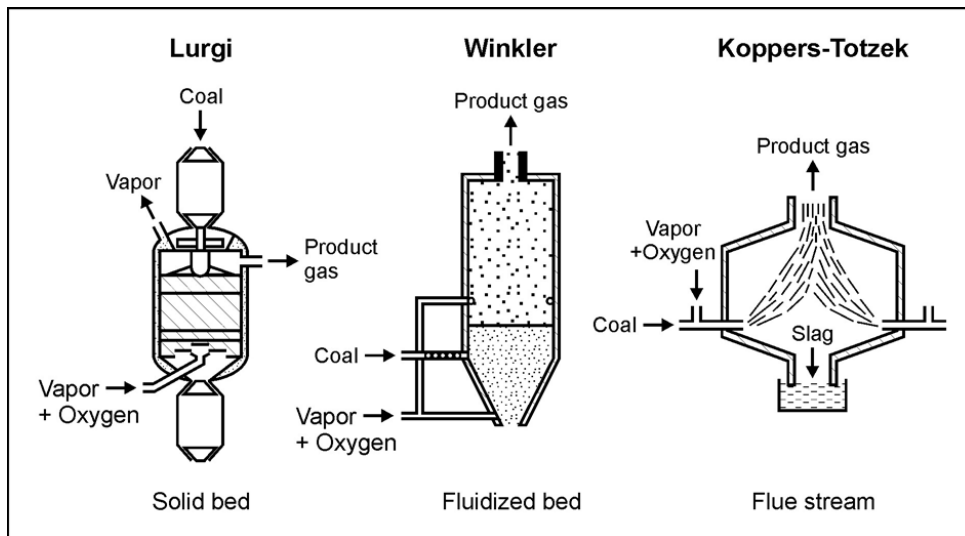


Fig. 2-9: Gas generator types for steam-coal gasification

In the hydro-gasification process, hydrogen is added to convert the coal to (synthetic) natural gas, before the synthesis gas is produced in parallel steam reforming and water-gas shift reactions. A high gasification degree can be obtained already with relatively short residence times of 9-80 min. In order to obtain high conversion rates, the CH_4 fraction should not be higher than 5 %, which

requires a low-temperature separation step. The advantage of hydro-gasification compared with steam-coal gasification is its 200 K lower pre-heating temperature which reduces potential corrosive attack. A major drawback is the large amount of residual coke of up to 40 %.

Gasification of coal is the oldest hydrogen production technology. Because of its abundant resources on earth, the conversion of coal to liquid or gaseous fuels has been worldwide commercially applied. Despite its comparatively low H_2 contents (H_2/C ratio of 0.8) and thus higher cost, steam-coal gasification is currently used to produce 18 % of the world's H_2 demand. At present, 20,000 MW of synthesis gas are being produced by coal, mainly for chemicals and power generation. However, its importance for H_2 production is decreasing, and rather its use for ammonia synthesis in the fertilizer industry or for methanol synthesis with large-scale production facilities particularly in the developing countries is enhanced. Air-blown plants have the lower capital cost vs. oxygen-blown plants, but they are suffering from much higher separation cost. Future H_2 production plants will most probably tend to oxygen-blown designs. Another important criteria for applicability and economy of coal gasification are the characteristics of the coal to be gasified. The (geologically) older the coal, the smaller is its reactivity and the higher is the temperature required. Major improvements of efficiencies could be achieved with the development of improved gas separation and purification techniques like the use of membranes.

The Integrated Gasification Combined Cycle (IGCC) is presently considered the cleanest and most efficient coal-fueled technique. A schematic is shown in Fig. 2-10. First step is the gasification of the coal followed by the Boudouard reaction to produce H_2 and CO_2 . The “entrained flow gasifier” in an IGCC usually operates at high pressures of 2-3 MPa and high temperatures $> 1400^\circ C$ to achieve high reaction rates and residence times in the order of seconds. Gasification is made with oxygen rather than with air. After separation and cleaning, the hydrogen is combusted in a gas turbine. The hot exhaustion gases can then be used in a steam cycle for further electricity production. With its gas turbine step prior to the oxygen/steam process and its intermediate stage of synthesis gas, it allows the removal of most carbon components before combustion. The separated CO_2 stream is of high purity and therefore suited for disposal. The thermal efficiency achieved can be up to 50 %.

The “cleaner” technology of IGCC was demonstrated in the 1980s with partial oxidation where the oxygen was distilled from air. Other facilities on a pilot plant scale have been following in Germany and the United States. Clean coal technology with removal of contaminants during gasification could largely eliminate the emissions of SOX, NOX, and particulates. Thermal efficiency is expected to improve by 10 % over conventional coal-fired steam turbine. But also other solid wastes can be gasified with the IGCC technology. For example, the respective plant “Schwarze Pumpe” in Germany treats a wide variety of solid (~ 1200 t/d) or liquid (~ 200 t/d) wastes ranging from waste plastic to tires, sewage sludge, household garbage for the generation of electricity, steam synthesis gas, and methanol (100,000 t/yr) [Kamka 2005]. IGCC technology represents the most advanced and efficient solution where the carbon in the fuel is removed, and hydrogen is produced in a pre-combustion process [Riis 2005].

Partial oxidation of coal is economic for coal countries (South Africa, China), only pilot plants in Germany. Under “normal” conditions, IGCC is not competitive with SMR. As of 2003, commercial IGCC plants in the power range of 250-350 MW and with efficiencies of 37.5-41.5 % are being operated in the USA (see Fig. 2-10) , Netherlands, Spain, and Japan.

The “Zero Emission Coal Alliance”, ZECA, initiative founded in 2000 in the USA, is pursuing a concept developed at the Los Alamos National Laboratory. It integrates hydro-gasification of coal, a CaO driven reforming step with simultaneous CO_2 separation, an SOFC for electricity production, and heat recovery. This process showing no air emissions and rather producing a stream of liquid CO_2 ready for sequestration promises an efficiency of about 70 % (HHV) of fuel energy into electricity [Ziock 2003]. The US-DOE goal within “FutureGen” is the verification of a zero-emission, coal fueled facility for H_2 and electricity co-generation coupled with CO_2 sequestration

with an overall efficiency of 60 % by 2015. The facility is also intended to be used as large-scale test bed for innovative technologies. The FutureGen program is now in the early stage of planning. Also Germany is planning the construction of a 400-450 MW CO₂-free IGCC plant by 2014 supposed the CO₂ storage system will be developed in parallel.

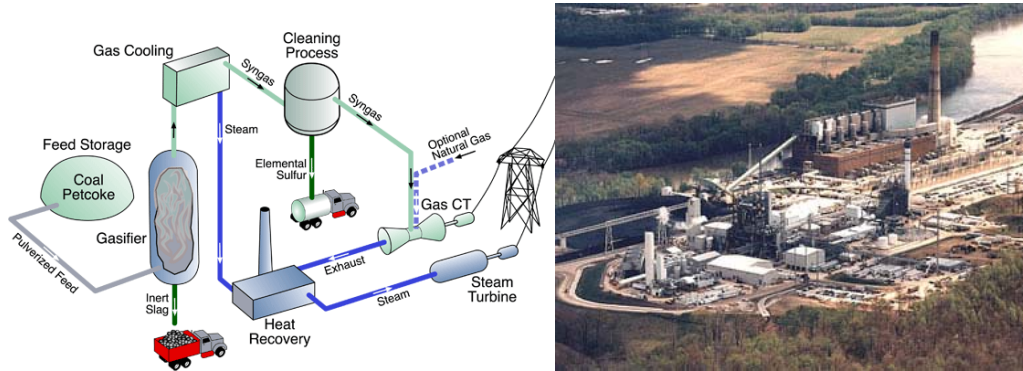


Fig. 2-10: IGCC flow diagram (left) and 262 MW Wabash IGCC plant, USA (right)

Another advanced method is the HYDROCARB coal cracking process. The coal is decomposed in a thermal cracker to carbon black as a clean fuel and hydrogen as a by-product fuel. The commodity carbon black outweighs the poor efficiency of 17 % for this method.

The characteristic data of hydrogen production in Table 2-6 are based on an autothermal hard coal gasification plant which also consumes 5 % of electric energy. For the data anticipated for the future, it was assumed that the gasification process can be optimized by lowering the fuel input by 15 % and the electricity input by 10 %, thus raising the efficiency from 59 to 69 % [Dreier 2000].

Table 2-6: Hydrogen production by hard coal gasification and its prospective

	State of the Art	Scenario 2025
Technical Data		
Rated power hard coal [MW]	486	413
Electricity demand [MW]	25	22
Hydrogen production [Nm ³ /h]	100,000	100,000
System pressure [MPa]	5.0	5.0
Water demand [m ³ /h]	84	84
Efficiency [%]	59	69
Life time [yr]	20	20
Utilization [h/yr]	8000	8000
Cost		
Investition [€/kW of H ₂]	1400	1400
Specific total cost [€ Ct/kWh of H ₂]	3.8	3.4 ¹⁾

1) Figure does not include potential CO₂ taxation or sequestration cost.

2.2.4. Coal Liquefaction for Transportation Fuel Production

Although no direct hydrogen production method, coal liquefaction is mentioned here as an alternative means for the generation of transportation fuels and other oil products applied in a number of countries as an insurance against crude oil supply problems [DTI 1999]. It is distinguished between direct and indirect liquefaction processes. In the indirect method based on the Fischer-Tropsch process from 1925, the coal is first gasified by pyrolysis to result in synthesis gas before converted to liquid products by reacting over a Co catalyst. Efficiencies are around 40 %, in modern plants up to about 55 %. In the direct method, coal is converted to liquid in a single step by dissolution in a solvent at elevated temperatures and pressures followed by hydro-cracking with H₂. Overall thermal efficiencies for the direct method are much higher, reaching the range of 60-70% in modern processes (see Fig. 2-11). Different kinds of pyrolysis (high-temperature, mild, rapid) with different liquid yields have been developed and tested in pilot plants, however, no demonstration plant has been operated so far.

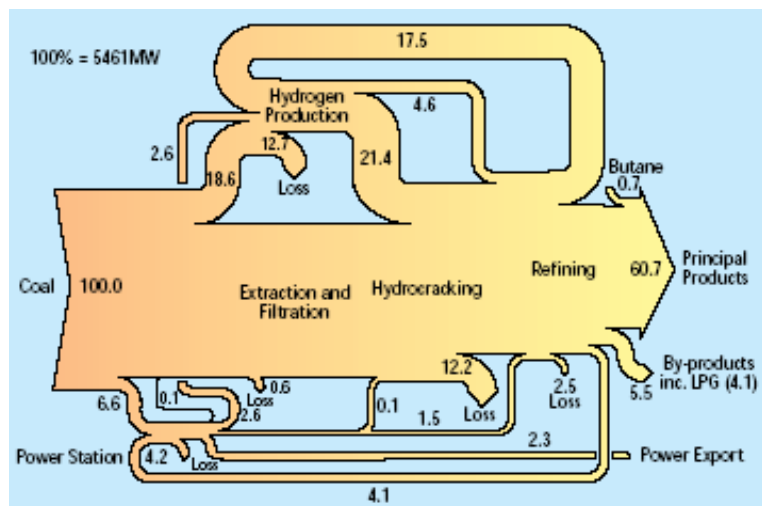


Fig. 2-11: Energy flow diagram for a "Liquid Solvent Extraction" type conceptual coal liquefaction plant [DTI 1999]

The Fischer-Tropsch process is a preferred liquefaction process because it has a large experience basis and yields high quality fuel. On the other hand, coal liquefaction is comparatively expensive particularly at the front end, where all contaminants have to be removed before processed to a gas or a liquid.

The production of coal-derived fuels started in Germany and the UK as early as around 1840. Significant R&D work was initiated during the oil crisis in the 1970s in the USA, the UK, Japan, and others, but later decreased again except for South Africa. At present, the indirect Fischer-Tropsch method applied by South Africa's SASOL company, has become worldwide the only coal liquefaction method on commercial scale. With a production of some 150,000 barrels per day of oil from coal, the SASOL process supplies about 40% of the domestic demand for liquid fuels.

China with its abundant coal reserves is experiencing a strong growth in coal liquefaction to reduce its dependence on oil imports. Several liquefaction plants based on the direct process are planned to be constructed with a total capacity of 60 million t/yr of oil. In Japan, coal liquefaction

was promoted under the Sunshine project. The so-called “NEDOL” liquefaction technology has been developed operating at relatively low pressures. A 150 t/d plant for bituminous coal is in operation since 1998. Japan is presently the only country active in large-scale process development. Within the United States DOE’s “Clean Coal Technology Demonstration Program”, a liquefaction plant with a capacity of 1000 t/d was commissioned based on the “mild pyrolysis” process.

2.2.5. Water Splitting by Low Temperature Electrolysis

There are principally two classes of low temperature electrolysis based on either a liquid electrolyte (most commonly potassium hydroxide, KOH) or a solid polymer proton exchange membrane (PEM). In both cases, the water molecule is dissociated by applying an electrical current.

In an alkaline electrolysis cell containing an aqueous caustic solution with usually 20 – 40 % KOH or NaOH, electrical energy is applied to two electrodes, which are plates made of nickel or chromium-nickel steels. Water decomposes at the cathode to H_2 and OH^- where the latter migrates through the electrolyte and a separating diaphragm, respectively, and discharging at the anode liberating the O_2 . The hydrogen gas is dissolved in the water and extracted in a separating chamber. A schematic is shown in Fig. 2-12.

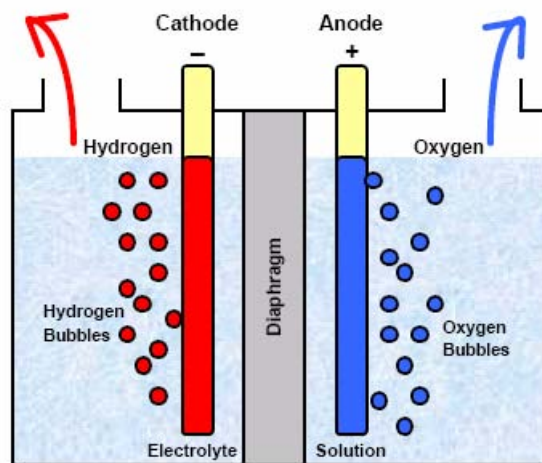
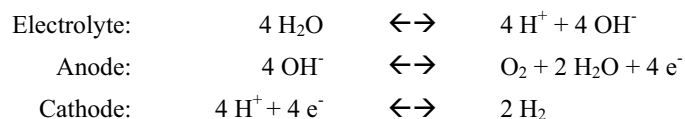


Fig. 2-12: Standard low temperature alkaline electrolysis

Operation temperatures are $< 150^\circ C$. The ideal reversible cell potential needed to split water is $1.229\text{ V @ } 25^\circ C$ which corresponds to a theoretical dissociation energy of 237 kJ/mol or an electricity demand of $3.56\text{ kWh/Nm}^3\text{ H}_2$. Caused by irreversible processes in the reaction mechanism to account for gas expansion at the electrodes and to maintain the operation temperature, however, typical cell voltages are 1.85 to 2.05 V . The electrical energy requirement is

in the order of 4 to 4.5 kWh/Nm³ of H₂ corresponding to an efficiency of 80 % and higher. The water demand is theoretically 0.8 l/Nm³ of H₂, in practice 1.0 l/Nm³. High-pressure electrolysis working at pressures of up to 3 MPa allows the saving of compression energy, if H₂ is stored as a pressurized gas or transported in pipelines, thus reducing the specific consumption of electricity.

The more advanced method is the solid polymer electrolyte water electrolysis (SPEWE) using an acidic, proton conducting (exchange) polymer membrane (PEM) as the diaphragm, thus making an additional electrolyte obsolete. Here the hydrogen ions are migrating through the membrane and recombine with electrons to hydrogen molecules. The oxygen remains in the water. The SPEWE can be operated at higher pressures and at higher current densities due to the compact design compared to cells with a KOH electrolyte. Typical operation temperatures are 200-400°C; pressures may go up to several tens of MPa. Membrane electrolysis is simpler in its design, safer, and promises a longer lifetime and a higher efficiency. The requirement of electricity will be reduced to values below 4 kWh/Nm³ of H₂. Main drawback is the limited lifetime and still high cost of the membrane [Riis 2005].

Alkaline electrolysis is the oldest and worldwide well established technology in industrial scale covering about 3.9 % of the world's production. In the chlor-alkali water electrolysis, the H₂ is actually a by-product of the chlorine production and mostly used as the thermal energy source and substitute of natural gas. A solution of salt in water is electrolytically decomposed into hydrogen and soda lye (mercury cathode) and chlorine (graphite anode). During the annual production of 35 Mt of chlorine worldwide, approx. 20 billion Nm³ of H₂ are being generated.

First alkaline electrolyzers for hydrogen production were developed by Norsk Hydro in Norway, where cheap electricity from hydro power could form the basis for this process. The large hydro electrolyzer units have a capacity of 485 Nm³/h (or ~ 1 t/d) at an availability of > 98 % and with an energy consumption of 4.1 kWh/Nm³ [Norsk Hydro 2002] (see Fig. 2-13). They usually operate at about atmospheric pressure and allow unmanned remote operation. Today's alkaline units are available across a range of capacities up to about 2 MWe. The largest electrolyzer units produce ~ 760 Nm³/h with multiple units being combined to larger capacities. Overall efficiencies achieved at that scale are about 63.5 % LHV, with compression bringing it down to about 59 %. The largest integrated installation is currently in Assuan, Egypt, with a production capacity of around 35,000 Nm³/h. The chlor-alkali electrolysis can be beneficial to the development of seawater electrolyzers which might be considered for H₂ production from off-shore wind farms. Characteristic data of various units of different sizes are given in Table 2-7.

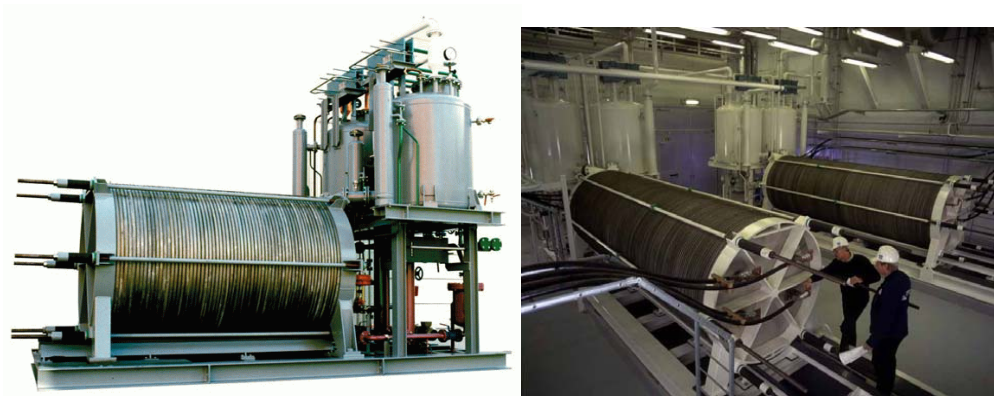


Fig. 2-13: Standard low temperature alkaline electrolysis (source: Norsk Hydro)

Electrolysis has become a mature technology at both large (125 MW) and small scale (1 kW). Efficiencies are principally independent of cell or cell stack size, which make them appropriate for small-scale use. Additional components like purification of water and products, rectifier and reprocessing of alkaline solution are necessary. Pressurized systems operating at higher pressures help many customers to save compression energy. Pressurized electrolysis up to 3 MPa is state of the art; operation at pressures up to 14 MPa was demonstrated in prototypes [EHFCP 2005a].

Table 2-7: Some data of industrial water electrolysis plants, data A-D from [Ivy 2004]

A: Proton HOGEN 380 **E: BBC**
B: Teledyne EC-750 **F: De Nora**
C: Stuart IMET 1000 **G: Lurgi (now ELT)**
D: Norsk Hydro HPE Atmospheric Type No. 5040

Parameter	A	B	C	D	E	F	G
H ₂ production rate [Nm ³ /h]	10	42	60	485			5-760
Electrolyte [%]	PEM	alkaline	alkaline	alkaline	alkaline	25% KOH	25% KOH
Type of cell	bipolar	bipolar	bipolar	bipolar	bipolar	bipolar	bipolar
Temperature [°C]				80	80	80	90
Pressure [MPa]	1.4	0.4-0.8	2.5	0.1	0.1	0.1	3.0
Current density [kA/m ²]				1.75	2	1.5	2
Voltage of cell [V]		2.1	2.04	1.75	2.04	1.85	1.86
Production pressure [MPa]	1.4	0.4-0.8	2.5	3.0			
H ₂ -purity [%]	99.999	99.9998	99.997	> 99.8	≥ 99.8	> 99.8	> 99.8
O ₂ -purity [%]				> 99.5	≥ 99.6	> 98.5	> 99.5
Energy requirement electrolyzer [kWh(e)/Nm ³ H ₂]	n.a.	n.a.	4.2	4.3 ⁽¹⁾	4.9	4.6	4.5
Energy requirement entire system [kWh(e)/Nm ³ H ₂]	6.3	5.6	4.8	4.8 ⁽¹⁾			
System power requirement [kW]	63	235.2	288	2330			
Water requirement [l/h]	8.4	42	60	485			
Conversion efficiency [%]	95	80	80	80			
Energy efficiency (HHV) [%]	56	63	73	73			

(1) incl. compression to 3.3 MPa

Low temperature alkaline electrolyzer operation is simple, highly flexible and appropriate for off-peak electricity use as well as for the intermittent operation of renewable technologies. System efficiencies of commercial low-pressure electrolyzers range from 60-73 %, can go up to 80-85 % with improvements made in the development of better electrodes and diaphragms. Also raising the operation temperature to 120-140°C both increases efficiency and lowers the requirements to the catalyst. Depending on gas purification technologies applied, H₂ purities range between 99.9-99.9998 %, those of O₂ range between 99.2-99.9993 %. Furthermore electrolyzers have also been made appropriate for intermittent operation like for solar energy. Purities directly achieved are > 99.9 % for hydrogen and > 99.8 for the oxygen [Norsk Hydro 2002]. Commercial large-scale electrolyzers produce hydrogen at an efficiency of about 65-70 % and a CO₂ intensity of 0-27 kg per kg of H₂ produced with 0 kg of CO₂ for renewable primary energy and 27 kg for a typical European electricity mix emitting 0.5 kg of CO₂ per kWh [EHFCP 2005a].

PEM electrolyzers exhibit efficiencies around 50 %, somewhat lower compared to KOH electrolysis, which is, however, compensated by the lower requirements for purification and compression. High-pressure systems are established in the smaller power range with pressures of 3 MPa achieved and efficiencies up to 80 %. Membrane development started in the 1950s-1960s for use in PEM fuel cells within the US space program. Main disadvantage today, however, is the still high capital cost of membrane manufacture preventing a significant penetration of the H₂ market up to now. In the development are plants for pressures up to 5 MPa.

2.2.6. Others

2.2.6.1. Thermal Cracking, Plasma Decomposition

Since methane belongs to the most stable organic compounds, high temperatures are required for its thermal decomposition. The search for optimal catalysts to reduce the maximum temperature has led to Ni or Fe based catalysts to decompose CH₄ in the range of 500-700°C (Ni) or somewhat higher (Fe). Activated carbon is seen as an interesting alternative for the 900-1000°C range, since it has a relatively high catalytic activity, low cost and would make an external catalyst unnecessary [Muradov 2005]. Process heat can be obtained either from an outside source like direct solar heating or from burning a part of the H₂ produced.

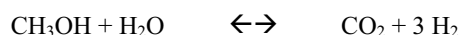
In the plasma-arc process, methane splitting takes place at temperatures in the order of 2500°C yielding solid carbon separated from the gas stream. The efficiency was reported to be about 45 %, but is expected to improve. In the Kvaerner method, an electrically powered plasma torch is being generated in a reactor to provide the decomposition energy. Hydrogen purity is 98 % prior to the cleaning step, if natural gas feed is used. In principle, all kinds of gaseous and liquid hydrocarbons can be used decomposing at 1600°C to hydrogen and carbon black with hardly any emissions.

H₂ production by catalytic methane decomposition was demonstrated already in the 1960s in a pilot plant with a capacity of 7 Nm³/h. The carbon was burnt to provide for the process heat at 815-1093°C (emitting large amounts of CO₂). SINTEF in Norway is using a 150 kW laboratory plasma torch with coaxial graphite electrodes, but without CO₂ or NOX emissions. In cooperation with Kvaerner, a 3 MW industrial-scale plant was constructed in Canada working since 1992. An input of 1000 Nm³/h of natural gas plus 2100 kW of electric energy results in the production of 2000 Nm³/h of hydrogen plus 500 kg of pure carbon and hot steam as a side product [Bakken 1998; HYWEB]. In 1999, the Kvaerner group has finally started the commercial operation of its first carbon black plant in Canada which runs on oil or natural gas and is designed for an initial annual capacity (in two units) of 20,000 t of carbon black plus 50 million Nm³ of H₂. The hydrogen is considered here a by-product and is recirculated to the plasma burner and used as process gas. The energy demand for the plant is said to be 1.25 kWh/m³ H₂ [Bellona 1999]. The conversion rate of

the hydrocarbon feed stock is almost 100 %. But also solar furnaces are under development using sunlight to provide the dissociation temperatures. Research efforts are concentrating on optimized concepts for gas injection, heat transfer, protection against undesired carbon deposition [Palumbo 2004].

2.2.6.2. Methanol Reforming

The decomposition of methanol by steam reforming takes place at moderate temperatures of 250-350°C and in the presence of a Cu/Zn catalyst.

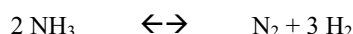


A steam/methanol ratio of > 1 (good: 1.5) results in a mixture of H₂, CO₂ and CO as the only significant products. Methanol is catalytically cracked in an endothermal process to produce synthesis gas plus traces of other gases. A two-stage membrane separation system extracts the H₂ from the CO-rich fuel that fires the cracker.

Present methanol reformers are of fixed-bed type. Drawbacks are hot and cold spots and slow response due to slow heat transfer. Improvement has been achieved by using washcoated heat exchangers. If applied in a hydrogen refueling station, methanol reforming needs a H₂ purification stage. Small-scale reformers for onboard fuel processors in FCV have been developed as an alternative to onboard storage of H₂. Various types include the plate and membrane concepts for compact design. A reasonable choice for portable fuel cell applications is the employment of micro-reactors for methanol reforming. Micro-reactor means channel sizes with a cross section of 1000 µm x 230 µm plus a 33 nm thick Cu layer as the catalyst.

2.2.6.3. Ammonia Cracking

Ammonia is also a hydrogen carrier and potential candidate H₂ source for FCV.



is an endothermal reaction with high reaction rates at > 700°C. Main impurities are NO_x and unreformed ammonia. Since PEM fuel cells need highly pure H₂, the recommended reaction temperature is 900°C. The reactor is of simple design, since no H₂O co-feed nor shift stage is required. The overall efficiency of an ammonia cracking fuel processor has been reported to be about 60 % with the remainder to supply the process heat and compensate for heat losses. For the production of pure H₂, efforts need to be made to separate N₂ from H₂ (which is not a problem in a fuel cell).

2.2.6.4. Biomass Processing

The conversion of biomass such as peat, wood, agricultural residues on the one hand or dedicated bioenergy crops on the other hand in a thermal process leads to a hydrogen containing gas mixture. Its H₂ contents is dependent on the fuel/feedstock, the availability of steam and oxygen, and the process temperatures. Processes for decomposition of the organic substances are gasification or pyrolysis with subsequent steam reforming, autothermal or allothermal (outside heat

source). The gasifiers are usually indirectly heated or oxygen-blown to avoid nitrogen in the product gas, and are operated at low pressures. The autothermal gasification in a fluidized bed results in a synthesis gas with typically 30 % of H_2 , 30 % of CO, 30 % of CO_2 , and 5-10 % of CH_4 plus some higher hydrocarbons. A shift reaction again converts CO to increase the H_2 fraction. Anaerobic fermentation of wet biomass leads to a CH_4 rich gas with only little H_2 , which, however, at a certain quality, could be used in higher temperature fuel cells (MCFC, SOFC). Although the conversion rate of biomass is high, H_2 production is highly inefficient due to the relatively low specific energy content of the biomass; only 0.2-0.4 % of the total solar energy are converted to H_2 .

The gasification of biomass or the microbial H_2 production by converting organic wastes is limited to mid-size plants for decentralized applications. Reasons are the distributed nature of biomass connected with high transportation cost, where the economy of scale does not apply. Facilities for wood treatment are on the verge of getting commercial. Demonstration pilot plants in the power range of 1 MW are being operated in various countries. Some apply an autothermal process and use air instead of oxygen. More advanced concepts perform gasification in supercritical water or apply thermochemical cycles. The technology still needs further improvements in feedstock preparation and raw gas handling and cleanup. Biomass conversion appears to be less convenient for H_2 production and is rather employed for heat and electricity production helping to reduce CO_2 emissions. Furthermore, biomass can easily be converted to a variety of valuable chemicals and liquid fuels like methanol.

2.2.6.5. Steam Iron Process

The steam-iron process, although based on coal, is a cycle process where hydrogen is generated from the decomposition of steam by reacting with iron oxide. The cycle is not completely closed since coal is consumed and CO_2 emitted. The synthesis gas produced in the coal gasification process with steam is reacted to reduce the iron oxide. In the following re-oxidation step with water, the original oxides are produced together with a hydrogen enriched gas. A continuous hydrogen production is given, if reduction and oxidation take place in separate reactors. This process has the advantage of producing H_2 at a high purity and the fact that renewables sources can easily be employed. Major drawback of this process is its poor efficiency.

2.3. Large-Scale vs. Small-Scale and Centralized vs. Decentralized Production

At present, most hydrogen is produced on-site in commercial, large-scale SMR units dedicated to the needs of the chemical and petrochemical industries. On-site production means flexible, on-purpose production with low or no transportation cost, a characteristic feature of decentralized H_2 production. In contrast, centralized hydrogen production refers to large-scale systems connected to a hydrogen delivery/distribution network transporting the H_2 to the point of use in gaseous or liquid state via pipeline or truck. Centralization allows for a secure and stable supply. Centralized large facilities are usually the result of efforts to decrease specific production cost by increasing the unit size (economy of scale) to make the product hydrogen more competitive with other energy vectors.

The use of hydrocarbons in hydrogen production systems will require a carbon sequestration functionality in order to realize the benefits of hydrogen production in general. Up to now the sequestration technology still needs to be developed further and verified in the long term. The capture, collection, and sequestration of CO_2 from many dispersed small reformer units appears to be prohibitively expensive and rather practicable only for large-scale centralized fossil fuel

handling. Distributed natural gas plants, a low-cost option presently, should therefore be replaced by electrolysis plants in the long run.

Also the use of nuclear primary energy as well as hydro-electric power only makes sense for centralized H₂ production on a large scale. Renewable energy sources (except for hydro) with their low-density energy and typically intermittent operation mode will be preferable for a dispersed system of H₂ generation plants. They can also be used to generate electricity and provide it to the grid at any place. The same applies to H₂ from biomass plants which will be limited in size simply because of the transportation of enormous amounts of biomass. Natural gas could be used for both centralized and decentralized H₂ production. A swift introduction of hydrogen into the market favors central production [EHFCP 2005a] which on the other hand will exhibit the problem of increasing the dependency on imports.

The advantage of decentralized distributive generation of H₂ is the ability to take benefit of the existing and widely available grids for electricity and natural gas. For future applications of hydrogen as part of the energy economy, the installation of a network of small-scale H₂ production units appears to be a good short-term approach for the introduction phase. These could be used to help usher in the hydrogen economy which will require some infrastructure changes for hydrogen distribution. Market prospects for stationary and mobile fuel cell applications have already led to the development of small-scale H₂ units on the prototype level to either be part of the required infrastructure for FCV or feed local grids for residential stationary fuel cell systems. Small SMR or electrolyzer units which are competitors at this scale, are attractive for early low-demand stages. They require less absolute capital investment and no transport and delivery infrastructure.

On the other hand, there are drawbacks in terms of limited efficiency and high H₂ cost (both production and primary energy), because they are lacking the advantages of the economy-of-scale factor and of the improved storage efficiency of large plants assumed a large-scale demand has built up. For smaller systems, capital cost become more significant. Furthermore operation and control of many small H₂ units require a cheap process control and high safety standards [HFP 2005]. Furthermore the H₂ production plants must meet the purity requirements of fuel cells. If connected to a pipeline grid, a problem may be seen in the mixing of H₂ streams from different sources and thus potentially different degrees of purity.

Small-scale reformers, either down-sized conventional units or specially designed units, are in the development and demonstration phase and are becoming increasingly powerful and efficient. Whether partial oxidation or autothermal reforming is an option will be depending on economic (innovative) ways of extracting oxygen from air or of separating nitrogen from the product hydrogen. Still at laboratory scale, but highly promising is the ITM technology to provide efficient small-scale H₂ units. In areas with lack of natural gas, reforming of methanol as easily transportable and storable fuel may represent an economic way of localized H₂ production. In other small-scale applications, reforming of methanol may be more cost-effective, so may be electrolysis on a very small scale.

The market for very small H₂ capacities in the range 50-500 Nm³/h is existing, but limited. A significant technology development has not been observed so far. On-board reforming of methanol is considered an alternative option to H₂ storage in an FCV which could take advantage of the already existing conventional transportation fuel distribution network. Decentralized electrolyzers which would be suitable for charging household vehicles exist as a mature technology. Consumer household vehicles can be charged during off-peak house to get cheaper hydrogen from electrolysis. However, this will result in a “well to wheels” inefficient use of energy since it requires that energy be transformed from many forms. This hydrogen will still be very expensive as compared to hydrogen produced by steam-methane reforming for which the required heat comes from natural gas. With respect to the planned network of H₂ refueling stations, a comparative cost analysis study has shown that up to capacities of 600 Nm³/h, the delivery of LH₂ by tank truck represents the most economic option [Ranke 2004].

2.4. Electrolytic Hydrogen Production and Present Grid Conditions

The main obstacle for abundant production of H_2 by electrolysis is the high cost of this route compared to petrochemical methods such as steam reforming of natural gas. At present prices of fossil fuels, H_2 from steam reforming costs about 7 €/GJ, and is by about a factor of 3 less expensive than 20 €/GJ to be paid for H_2 from electrolysis by electricity at 30 €/MWh [Dutta 1990, Lipman 2004]. Due to the high cost share of 80 % for electricity in electrolysis, the electricity has to be made available for less than 10 €/MWh for the sake of competitiveness, a level even difficult to achieve for hydropower.

A proposed alternative is the use of electricity from NPPs at off-peak periods. This operation mode will be analyzed in the following from the perspective of grid and plant behavior and market influence.

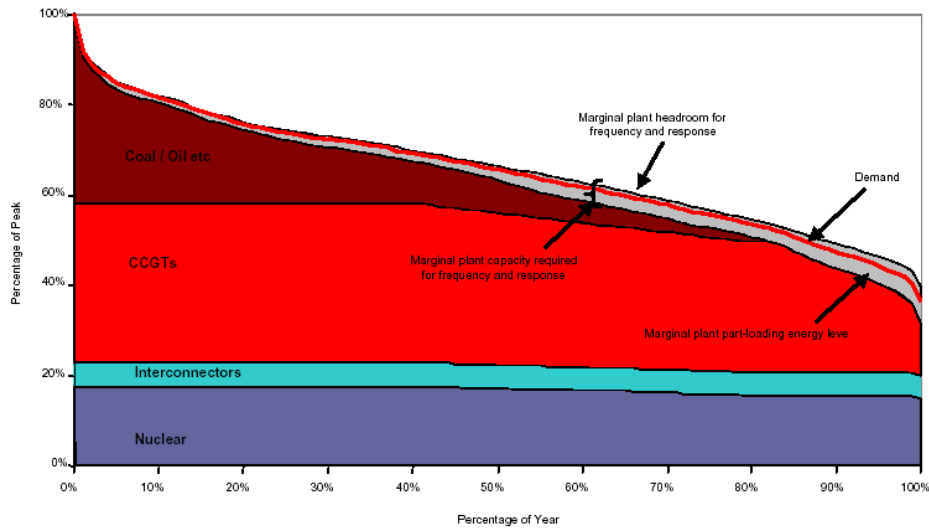


Fig. 2-14: Example of a demand-duration curve for a national grid [NGC 2001]

Off-peak electricity is defined by the demand-duration curve of the grid under consideration. Fig 2-14 shows an example, with the typical ratio of 1:1 for base load and partial load percentage. The overall demand is balanced by the park of different power plant types: NPPs are operated in the base load mode, using the advantage of low fuel cost and distribution of capital cost over continuous operation. Short temporary demand is supplied by plant with low investment/high fuel cost shares such as CCGT. As a result, for the plant mix shown in the example, there is no off-peak situation for NPPs. Obviously a large share of NPPs in the plant park is a prerequisite for nuclear off-peak electricity. A decision for deploying NPPs for off-peak operation has to compare the cost of NPPs versus the flexible fossil fired plants as GT or CCGT: As fossil fuel becomes more expensive, the use of NPPs outside the base load region becomes more attractive.

On the other hand, the optimized selection of plants in the park is also determined by the trade-off between the cost structures of the different plant types, which is given in Table 2-8 [NucNet 2002, Delene 1999].

Table 2-8: Cost structures in different plant types (1 € = US \$1.20)

	CCGT	Adv. LWR
Capital + O&M [€/MWh (base load operation)]	11	25
Fuel [€/MWh]	22	4

Considering a simple splitting of costs into a share independent of operation and another one for the fuel, the present values will result in a disadvantage of nuclear power for partial load duration shorter than 80 % of the year (Fig. 2-15). A doubling of the fossil fuel price shifts this limit to 35 %, a doubling of both fuels to 40% of the year. From this result, some partial load demand can be delivered by NPPs under economical operation, but there is obviously a need for other than nuclear sources to allow an optimized operation of the grid over the full demand range under commercial aspects. This may include the interconnection between different grids.

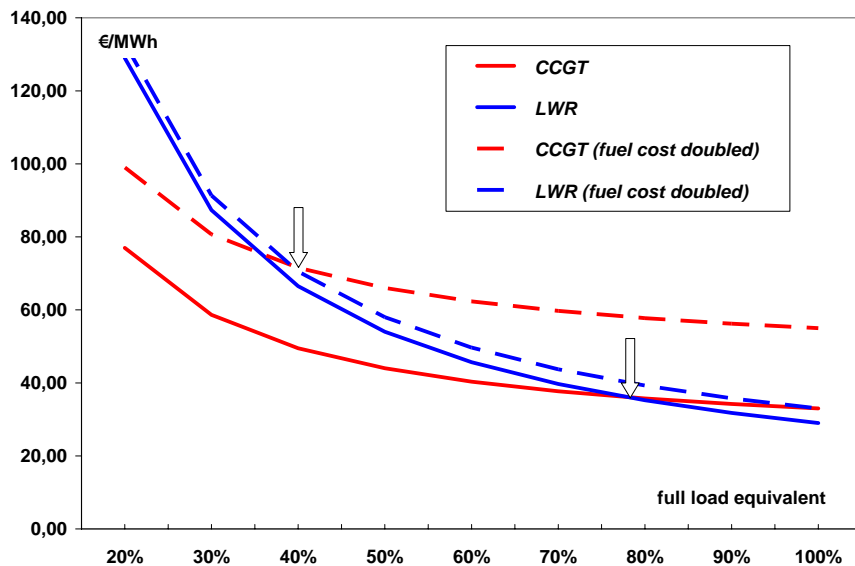


Fig. 2-15: Cost effect of partial load

The corresponding maximum nuclear off-peak capacity can be estimated from a linear 50 % → 90 % approximation of the demand-duration curve (see Fig. 2-16): The off-peak shares compared to the overall production are 1.1 % and 10.3 % for the above mentioned limit values 80 % and 40 %, respectively, i.e., a real off-peak share of nuclear electricity can be expected for the case of increased fossil fuel costs and a plant park adjusted.

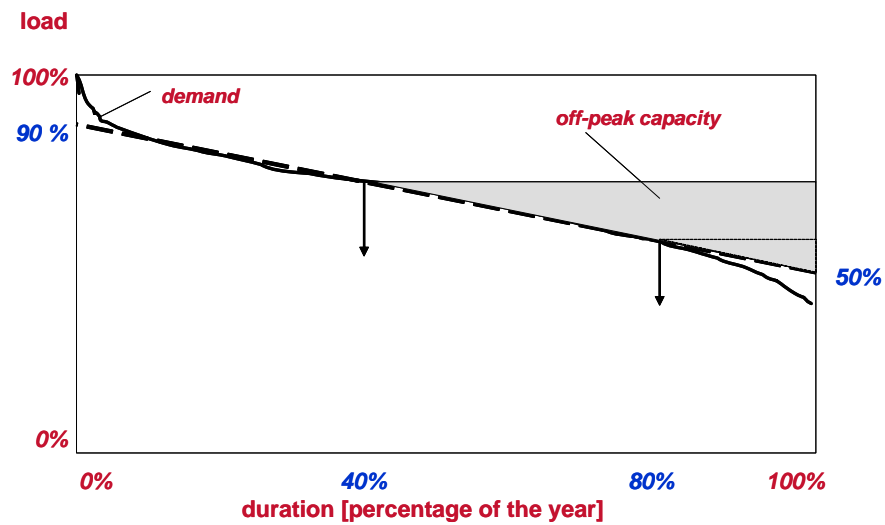


Fig. 2-16: Approximation of off-peak capacity

Although used so far in this operation mode only accidentally, NPPs are capable to follow grid demand by partial load: The characteristic speed for acceptable load changes is up to 10 % of full load per minute, the full load range can be crossed in about 1 h; for faster fluctuations the storage of the steam generators can be used [Gruhl 1977]. This should allow following the typical grid demand over the day shown by a typical example in Fig. 2-17 [NGC 2001].

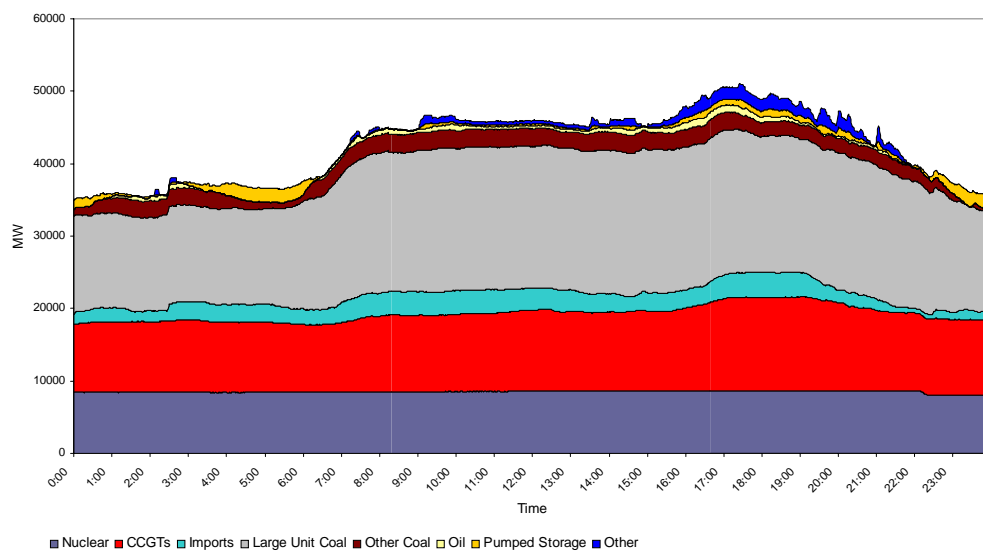


Fig. 2-17: Example of a daily demand curve for a national grid [NGC 2001]

The cost of electricity for partial load operation increases for all plant types, but for the range up to the limiting ξ values discussed, nuclear is the cheapest option. For the following analysis, the simplifying assumption is used that regular grid demand carries the whole constant costs of the plant, and the off-peak electricity has to pay the costs for the related nuclear fuel demand only. According to Table 2-8, this results in 4 €/MWh, i.e., 15 % of the usual electricity costs only.

The cost of H₂ production by electrolysis has been discussed in the preceding sections. Cost structure studies show that electricity costs are about 80 % of the total costs of 20 €/GJ for H₂ generation by high temperature electrolysis [Dutta 1990] and regular electricity cost of 29 €/MWh. This can be reduced to 4 €/MWh now.

For operation with off-peak electricity, in a first scenario the reduced duty factor of the electrolysis system has to be taken into account: For $\xi = 40$ % and the linear approximation discussed in Fig. 2-16, the H₂ plant operates in the average at a full capacity equivalent of ≤ 30 % only. This results in H₂ production costs of about 16 €/GJ from off-peak nuclear electricity. The energy losses are taken into account in the electrolyzer cost.

In contrast, H₂ from conventional steam reforming costs about 7 €/GJ at present fossil fuel costs. At a present feedstock cost share of 75 %, the price of the feedstock has to triple for reaching H₂ costs from off-peak nuclear electricity. In this case, the prerequisite for the plant park mix assumed above would be fulfilled.

The temporal behavior of the H₂ electrolysis system does not favor partial load operation. It is expected that the heating for high temperature operation has to be maintained over a longer period than the duty time from off-peak electricity and that there are limits for the exothermal variant of the electrolyzer operation [Quandt 1986].

To avoid this, a second scenario assumes continuous operation of the electrolyzer plant by temporal use of electricity at regular price. If there is a 'base demand' large enough for H₂ from electrolysis at 'full' price, an isolated view on H₂ production costs during off-peak periods results in 7 €/GJ, i.e. it is competitive to the conventional petrochemical route. To confirm this possibility, an analysis of – regional – H₂ markets and applications is required.

Different schemes for using H₂ stored for leveling source and demand fluctuations has been discussed in connection with deployment of renewables. In addition to the investment costs, the mechanisms of loss of elements in the chain back to electricity have to be analyzed for a complete picture.

The cost of H₂ storage as a compressed gas is highly dependent on the turnover rate. Assuming above-ground compressed storage, at low turnover rates (e.g., 30 days) the overall costs are estimated to be above 6 €/GJ, whereas for daily storage of H₂ storage, the costs will be – due to a different capital cost distribution – between 1 and 3.5 €/GJ [Padró 1999]. The peak demand distribution will result in a value closer to the daily turnover. Underground compressed storage – suitable for larger amounts – does not differ in overall costs. Specific storage costs in large liquefied H₂ storage systems are higher by a factor of 2, mainly caused by the energy demand for the liquefier.

From the H₂ stored, electricity can be generated by a gas turbine. For the foreseeable future, electricity from fuel cells will not become cheaper than from CCGT systems.

As an approximation, the capital costs for electricity production by a natural gas turbine plant might be used, although the technical details vary somewhat [Jordan 2005]. Since such a plant is in operation during the peak demand only, the part time operation has to be taken into account. For calculating a lower limit for the capital costs, an upper limit for the duty factor of 30 % may be assumed. The fuel costs have to be derived for H₂ generated by off-peak electricity and stored resulting in 19 €/GJ (first scenario above). Using the energy content as a basis for comparison, natural gas costs about 3 €/GJ [NucNet 2002]. Scaling capital and fuel costs in Table 2-8 above to part-time operation and from natural gas to stored off-peak H₂ results in ≥ 175 €/MWh. Even for the

second scenario the result is ≥ 100 €/MWh. This is near the generation costs of electricity by photovoltaic systems.

The resulting costs of electricity from the chain have to be compared with the respective on-peak section of the cost-duration curve of the grid. Roughly a factor of 1.6 can be derived from Fig. 2-18 for the upper half compared to the lower half of this curve. This does not compensate for the costs and losses of the electricity \rightarrow stored hydrogen \rightarrow electricity chain starting from an off-peak nuclear plant.

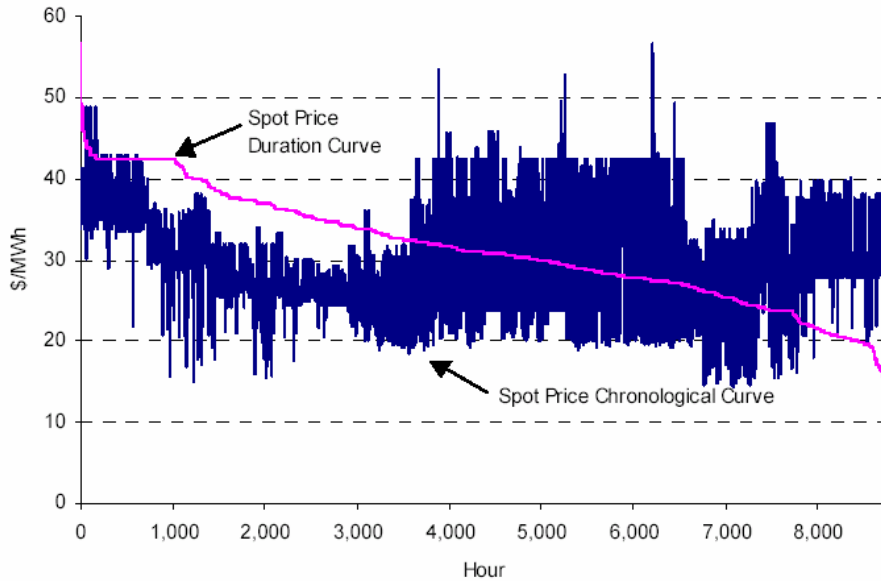


Fig. 2-18: Example of price-duration correlations [GE-MAPS]
(spot price as incremental duration curve (cf. Fig. 2-1) and actual price fluctuations as chronological record over the hours from Jan 1st to Dec 31st)

As an overall conclusion, one main obstacle for the storage of electricity by intermediate hydrogen production is the cost of part-time operation of the chain (electrolyzer, turbine plant) intended for avoidance of part-time operation of the nuclear plant.

The discussion on the development of storage facilities for large volumes of H₂ has stimulated ideas for optimizing the use of off-peak electricity surplus from nuclear plants and from wind [Miller 2005]. Leveling out the fluctuating generation of electricity from wind is a challenge for the grid management when the wind share becomes $> 20\%$. The approach of *NuWind*[®] has been summarized by: “Make H₂ when electricity price is low; sell electricity when high; dispatch electricity when needed, otherwise make H₂....; optimize threshold price for electricity to be sold or converted”.

Indeed hydrogen storage would allow a “well-defined” avoidance of low electricity prices at the spot market (cf. right hand side of the curve in Fig. 2-18), which might result in extra revenues. The scheme might become profitable, if someone buys the H₂ generated from electrolysis at a price near the level discussed above in this section – somewhat reduced by the extra revenues from avoiding low electricity spot prices.

Of course, the electrolysis and storage facilities to be provided must handle large amounts of hydrogen: 1 GW of off-peak electrical power result in $\sim 250,000$ Nm³/h of H₂ to be stored, if H₂

demand and off-peak electricity phases cannot be synchronized. The effect of storage duration on H_2 costs has been described above. Thus an overall evaluation needs more information on the emerging “hydrogen economy”.

2.5. Synergism of Electricity & Hydrogen in the Long Term

For future applications, it has to be kept in mind that electricity and hydrogen can form a symbiosis allowing an optimization of electricity generating systems with regard to the substitution of fossil energy carriers currently being used in the primary energy mix especially for load following and peak demands. This is due to the fact that electricity and hydrogen are principally interchangeable, i.e., the production of hydrogen, e.g., via electrolysis and the re-conversion of hydrogen into electricity (and heat), e.g., by fuel cells. In the following, this fundamental inter-connection is being addressed as “hydricity” [GIF 2002].

Electricity and hydrogen are also complementary to each other as electricity must be produced at the time where it is used, whereas hydrogen can be stored for stationary re-conversion into electricity or other use such as transport fuel and mobile re-conversion for driving electric motors.

Electricity is generally produced by relatively large, central station plants, although alternative technologies may make distributed production of electricity and cogeneration of heat more feasible. However, no matter what means of production is used, storage of electrical energy is generally costly and not particularly efficient. Much effort has gone into battery technology, superconducting magnets, fly wheels, pumped hydro and other means of “storing” electricity with no clear cost effective winner. Conversely, hydrogen offers flexibility in that it can be stored, although with some challenges. However, multiple economic approaches for storing hydrogen may become more feasible. Hydrogen, particularly through electrolysis, is also amenable to distributed production. When these complementary characteristics of electricity and hydrogen are coupled with a variety of production strategies, the resulting system is extremely flexible on a macro-scale and provides a large number of degrees of freedom in designing a national energy infrastructure by hydricity.

For example, current electricity production is normally defined as base load or peaking. A base load plant can have high capital costs but lower fuel and operating costs making nuclear ideal for this role. Alternatively, peaking plants, because they operate intermittently, are characterized by lower capital costs and by higher fuel costs. Natural gas peaking plants are prime examples. However, with hydricity, the distinctions between base load and peaking production will disappear and the overall system can be optimized using different criteria. The output from a large electricity generating plant may go towards meeting the entire demand during peak periods, but the electrical output can be directed towards generating hydrogen during off-peak periods. Likewise, hydrogen is a potentially attractive form in which to store energy since the hydrogen output might be distributed or alternatively used to meet peak demand and spinning reserves through fuel cells or being used in combustion engines as long as fuel cells are not yet technically or economically available. The cost of peak power is significantly higher than for electricity during low demand. This fact can favor the introduction of a new system which is predominantly focused on peak / intermediate power production, spinning reserve and load following.

As discussed in the previous chapter, intermittent, part time operation of hydrogen production, e.g. by using off-peak power, is economically problematic due to the high investments of the hydrogen production facilities. In this respect, there is a direct analogy to nuclear energy which is predominantly operated in base load. This will also apply for large-scale hydrogen production but will only be feasible in combination with high capacity hydrogen storage to cope with fluctuating hydrogen demands. Thus base load “Cogeneration of Hydrogen and Power”, CH2P, might be an

economic approach, in the long term. Fig. 2-19 illustrates the CH2P principle for a nuclear power plant which also delivers heat for high-temperature electrolysis or for thermochemical H₂ production, e.g., via Very High-Temperature Reactors (VHTR).

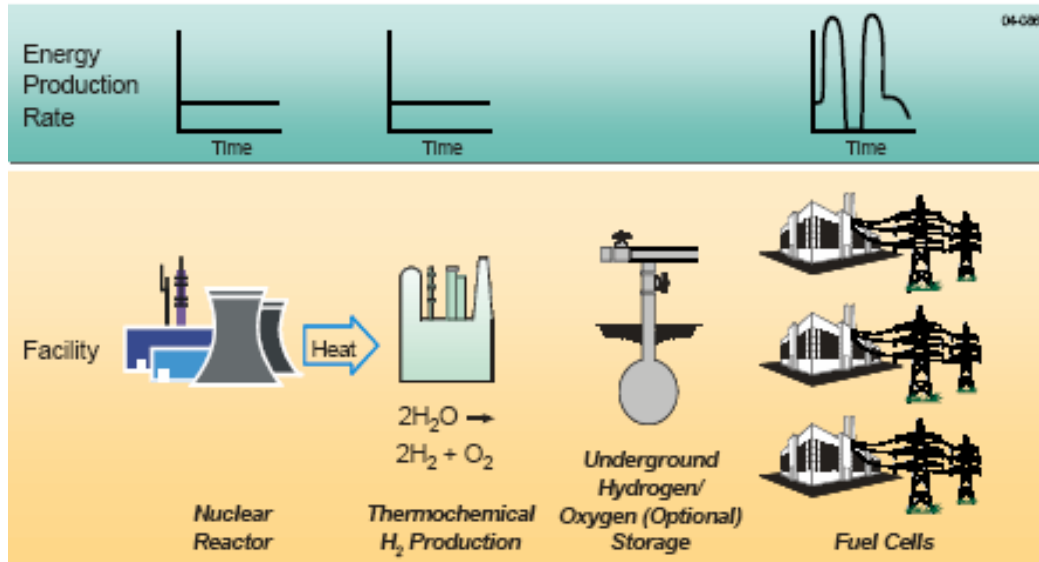


Fig. 2-19: Base-Load Cogeneration of Hydrogen and Power [Forsberg 2005a]

A large-scale CH2P technology can easily be introduced into the electricity market for substituting the combustion of oil and natural gas. This approach may also be capable to resolve the ‘Hen and Egg Dilemma’ with regard to the slow evolution of a future hydrogen market and the establishment of large-scale H₂ production units to achieve competitive cost. Cheap hydrogen (via economy of scale) will be a precondition for a mass market but a mass market is also required to create the incentives for the development of new technologies and for investments into hydrogen production facilities. In this sense, the electricity producers might find a new business case in the cogeneration of hydrogen and power, first as substitute for peak load natural gas or oil fired power plants for their own use and secondly delivering excess hydrogen to other clients like refineries, ammonia producers, petrochemistry etc. or as transport fuel for an evolving methanol or hydrogen car fleet.

A nuclear hydrogen production of the same size as the largest conventional plants presently constructed (~8.5 million Nm³/d) will need about 2400 MWt of high-temperature heat. This is compatible with a set of 4-6 VHTR modules (400-600 MWt each) assuming a conversion efficiency of 50 %. The economies of scale for nuclear hydrogen production e.g. by the Westinghouse hybrid thermochemical process have been estimated to be about a scaling factor of 0.54. This means that an increase of the power size by a factor of 4 yields capital cost of only 53 % per unit of capacity as compared to the smaller plant.

It is obvious that this approach is highly dependent from the availability of large and cheap hydrogen storage capabilities. Underground caverns, depleted oil and gas fields or aquifers are already used for low-cost storage of natural gas to cope with seasonally variable demand or buffer to maintain pipeline pressure. In countries such as UK, caverns have already been used over many decades for the economic storage of hydrogen but there are still other technologies under development for the bulk storage. The storage may also include hydrogen generated from

regenerative energy sources and thus even create a symbiosis between nuclear and regenerative energy systems. Besides, the storage requirements for the use of hydrogen from regenerative sources are very similar as is the case for the CH₂P system. Storage capacity can stepwise grow from substitution of daily peak demands towards weekly and seasonal compensations.

Typical capital cost of an underground storage for one GW-year of hydrogen (lower heating value) is estimated to be about US \$ 200-400 million (0.8-1.6 US \$/kg) assuming the same cost per unit volume for natural gas and hydrogen [Thomson 1997]. The value of the hydrogen stored in such a facility significantly exceeds the investment and even allows considerations on seasonal storage.

As long as CO₂ is not a “rare resource” (e.g., at the site of coal fired power stations), further symbioses can be taken from conversion of CO₂ and hydrogen into methanol as liquid transport fuel and storable medium. In the future, methanol storage tanks could play the same role as is currently the case with national oil reserves. It can also be used as an alternative fuel for combustion engines as well as for fuel cells. The double use of the CO₂ exhaust would also contribute to significant reductions in CO₂ emissions which is the primary goal of a hydrogen economy and not the production of hydrogen, per se.

The economic feasibility of CH₂P generation is also dependent on the projected cost and efficiencies of fuel cells. It is expected that the specific capital cost can be reduced to < US \$100/kW_{el} with an efficiency of ~ 70 %. Capital cost for fuel cells would then be much less than for gas-turbine plants (US \$ 500/kW_{el}) and 50 % efficiency). The use of oxygen for the fuel cells could further boost their performance. The fuel cells may also be used for traditional Combined Heat and Power (CHP) applications when using the waste heat for district heating or industrial process heat. Main drivers for the capital cost of such a system are first the cost of the nuclear heat source, secondly of the hydrogen production plant and thirdly for the fuel cells whereas hydrogen storage would only contribute to a minor part.

For transport fuel production, there are other advantages of the nuclear generated electricity/hydrogen duality. Currently in petroleum refining, costs of gasoline are impacted by a variety of factors. These can include foreign production schedules, changes in shipping schedule, spot market prices, weather, international events and many other influences. With hydricity, many of these uncertainties are eliminated. The opportunities will be available to draw upon a stable, reliable indigenous energy supply unaffected by the multiplicity of factors that currently impact energy prices. Hydrogen and electricity produced from nuclear also in combination with regenerative energies is an energy paradigm that means economic stability, eliminates massive balance-of-payment deficits, and offers enhanced security from international instability.

2.6. Alternative Transportation Fuels

Not only is hydrogen considered a transport fuel, which may be applied in future on a large scale, it is also a basic building block with increasing significance (and market potential) for the production of conventional liquid fuels, but also – partially – for other alternative fuels or synthetic fuels which may gradually displace the conventional ones meeting the challenges of air pollution, CO₂ emissions and supply security. If captured CO₂ is used for their production, they would even represent CO₂-neutral alternatives. Table 2-9 and Fig. 2-20 list various fuels for both ICE and FC vehicles and some of their characteristics.

Table 2-9: Alternative transportation fuels

Fuel	Main source	Molecular weight	Density [kg/m ³]	HHV [kJ/kg]	Energy per volume [GJ/m ³]
Hydrogen	Natural gas, oil, coal, water, methanol	2.02	0.08988	141,890	2.6 @ 20 MPa 10.3 @ 80 MPa
LH ₂	Hydrogen	2.02	70.8	141,890	9.9
Natural gas (methane)	Fossil resource	16.04	0.7175	55,530	8.1 @ 20 MPa 32.1 @ 80 MPa
CNG	natural gas	16.04		55,530	
LNG	natural gas	16.04	422.6	55,530	~ 20.5
LPG (propane)	refining of petroleum, NG	44.1	581	50,400	25.2 ~ 23.4
Methanol	natural gas, coal, woody biomass	32.04	793	22,700	18.0
Ethanol	biomass, grain, corn	46.07	794	29,900	23.5 ~ 22.3
Gasoline	crude oil	100-105	745	47,400	30.4-34.8
Diesel	crude oil	~ 200	832	45,840	35.7-36.2
Biodiesel	biological oil, animal fats	120-320	830-850	39,800	32.6-33.4
DME	Natural gas, coal, organic material	46	670	31,700	21.1
MTBE	isobutylene	88.15	741	7800	28.1
Toluene	crude oil, tolu tree	112	862	42,500	26.9
Ammonia	natural gas, heavy oil	17	771	22,500	17.4
Jet A	crude oil	144-226	775-830	46,500	34.2

Liquid fuels are advantageous because they are easy to handle at room temperature and atmospheric pressure. Furthermore as indicated in the above table, all hydrocarbon fuels contain more hydrogen per unit volume than pure liquid hydrogen and therefore have a higher energy density and thus perform better than H₂ with respect to handling, storage, distribution and well qualified for a wide-spread use [Bossel 2003].

Methanol produced, e.g., from coal by clean coal technologies, can be used in direct methanol fuel cells and also in higher temperature fuel cells for electricity production or even in PEMFC after a reforming stage.

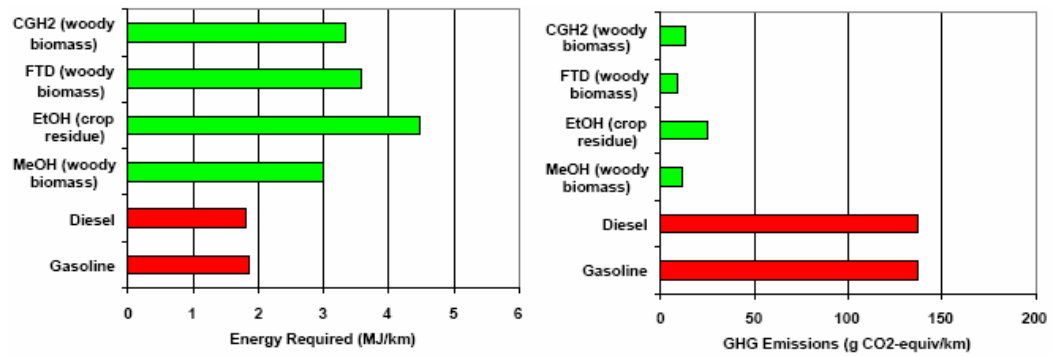


Fig. 2-20: Comparison of today's petroleum fuels with alternative non-fossil derived transportation fuels [Brusstar 2005]

3. NUCLEAR HYDROGEN PRODUCTION TECHNOLOGIES

3.1. The Nuclear Reactor as Heat and Electricity Source

In principle, all methods of hydrogen production, apart from the photolytical ones, can be coupled with a nuclear reactor to provide electricity and process heat. Conventional light-water reactors (LWR) can be employed to deliver electricity for the low temperature electrolysis process; electricity and hydrogen production are principally separated and could even be deployed at different locations. Other types of reactors with higher coolant outlet temperatures would allow the direct utilization of the hot medium which transfers its heat to the chemical process. In such cases, the H₂ production site must be in close vicinity to the nuclear site. The mutual use of the basic reactor design for electricity and process heat generation was a fundamental request in former German industrial HTGR development programs. This applied specifically to the past projects on direct gas turbine cycles (HHT), steam cycles (THTR-300, HTR-500) and respective nuclear process heat projects (PNP, NFE).

In 2002, several nuclear reactor concepts were presented as promising concepts for the next generation (Gen IV) [GIF 2002]. These innovative technologies are being designed to deliver, apart from the classical electricity, also heat, hydrogen, and/or clean water. The six concepts listed in Table 3-1 have been selected to be later boiled down to fewer options. Representing new, perhaps even revolutionary reactor technologies, they are expected to give convincing answers to the challenges of improved safety, reduced cost, minimized waste, and enhanced resistance against sabotage and proliferation.

Table 3-1: Characteristics of the Generation-IV reactor concepts

Gen IV reactor concepts	Neutron spectrum	Coolant exit temp. [°C]	Fuel	Fuel cycle	Reference size [MW]
Very High Temperature Reactor (VHTR)	thermal	1000	UO ₂ block/pebble	open	250-600(th)
Supercritical Water Reactor (SCWR)	thermal fast	550	UO ₂	open closed	3575(th) 1700(e)
Gas Cooled Fast Reactor (GFR)	fast	850	U-238	closed	288(e)
Heavy Metal (Pb-Bi) Cooled Reactor (HMCR)	fast	550-800	U-238	closed	50-150(e) 300-400(e) 1200(e)
Sodium Cooled Reactor (SCR)	fast	550	U-238, MOX	closed	150-500(e) 500-1500(e)
Molten Salt Reactor (MSR)	epithermal	700-800	UF in salt	closed	2250(th) 1000(e)

The reactor coolant and its maximum temperature are essential criteria for determining which nuclear concepts are appropriate for a coupling with hydrogen production processes. Fig. 3-1 shows coolant outlet temperatures of the Gen IV concepts ranging between 550-1000°C compared with the process heat temperature ranges needed for various chemical processes. Coolant exit temperatures of the reactor at full load must be maintained at all levels of partial load. The coolant outlet temperature needed for the chemical processes should be about 50°C above the bulk process temperature [Brown 2003]. As far as the maximum helium temperature is concerned, a value of 900°C is already suited for steam reforming applications. A value of 950°C, however, would significantly reduce the heat transfer area and improve efficiency which is a potential for the future. The maximum coolant exit temperature of 1000°C remains a challenge to both the reactor fuel and the metallic materials (reactor pressure vessel, thermal barriers, etc.).

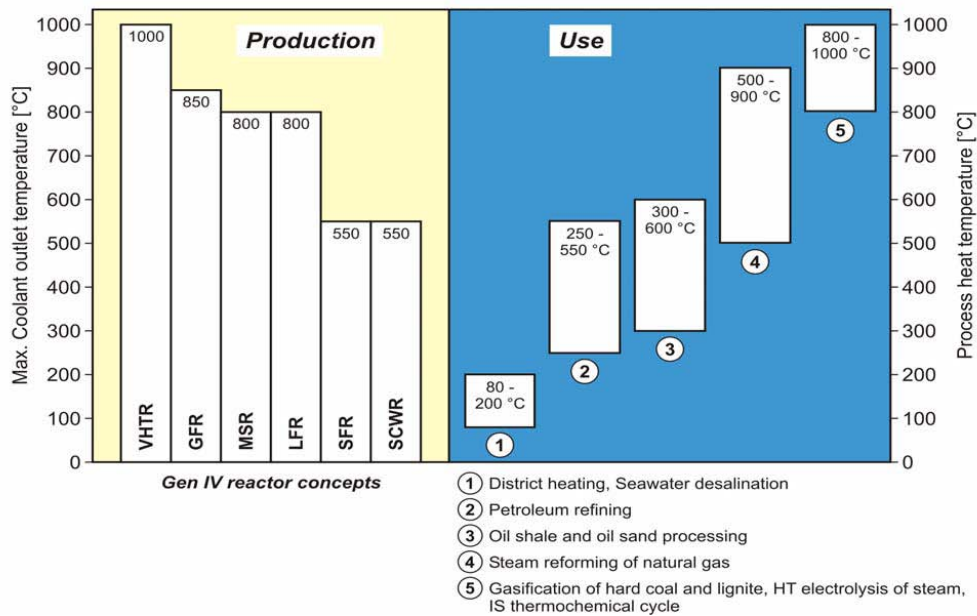


Fig. 3-1: Temperature ranges in production and use of nuclear energy

Only some adjustments were required with regard to maximum temperature and pressure levels in the primary circuit, because the chemical processes were mainly expected to be operated at a lower pressure than would be the case, e.g., in the direct gas turbine design. Another general difference is the fact that the cold gas temperature in the process heat applications are generally much lower (250-300°C) than in gas turbine applications (450-550°C) having a beneficial feed-back on the pressure vessel design and material choices. Fuel temperatures during normal operation, however, should stay below 1300°C to avoid larger release rates of fission products.

Especially for the pebble bed reactor, an additional possibility exists to realize high helium temperatures at relatively low fuel temperatures by employing the OTTO loading scheme (Once Through Then Out). The difference between maximum helium and maximum fuel temperature in an OTTO core is around 100°C. Although bearing the potential to produce in a simple way very high helium outlet temperatures, the OTTO cycle does surely not support the general goal of sustainability demanded for Gen IV reactors.

For higher temperatures, there are some advantages for the pebble fuel due to the homogeneous distribution of the coated particles in the matrix. The continuous reload also fits to the industrial practice, e.g., in refineries that do not shut down for decades. The heat market requests much smaller units than is the case for dedicated electricity generation.

As most chemical processes are performed at lower pressures some adaptation of the reactor design and of the chemical process has been necessary. Therefore, the reactor pressure had been fixed in the PNP project to 4 MPa being much beyond the pressure for electricity generating plants (~7 MPa). The choice of the pressure is also important to reduce the loads on the high temperature barriers in case of depressurization accidents either in the primary or in the secondary circuit. Other important aspects of reactor design are the power size that may be much less as compared to dedicated electricity production and higher redundancies due to smaller local heat grids and very high availability requirements as well as an optimization towards significant simplification of the nuclear island. Therefore, 600 MWt per reactor unit might be much beyond the real needs for nuclear process heat e.g. even for large refineries. A simple use of the GT-MHR or PBMR design for nuclear process heat might not be the most straightforward way to offer nuclear heat supply systems for non-electric applications although many components and operational features might be similar.

A fundamental difference between the electricity and the process heat market is the power size distribution of the energy supply system. Large plants have been favored for dedicated electricity generation, whereas in the non-electric energy market, small and medium-sized CHP plants are applied due to the limited size of heat distribution networks and due to very high availability requirements asking for redundant – modular – power supply systems. The data elaborated for different CHP applications in EU clearly show that the power size of a 200-300 MWt modular design would well fit the needs of industrial heat consumers whereas a 600 MWt GT-MHR represents already an upper limit. A modular arrangement (2-6 units) will be necessary for redundancy, reliability and reserve capacity reasons which again reduces power size per modular unit. However, smaller power size allows for simplicity and robustness by higher safety margins even at higher operational temperatures if necessary at all. More detailed investigations on the market structures are highly recommended before fixing the reference design for a nuclear heat supply system. Considering this background, it should be investigated whether steady increase of power for modular reactors being deployed for electricity generation is the correct approach for CHP applications which might need a special reactor design and system layout, e.g., with combined gas and steam turbine cycle as applied in the steadily growing conventional CHP market.

Nuclear reactor and hydrogen plant will be physically separated from each other by employing an intermediate heat exchanger (IHX) between the primary helium circuit of the reactor and the steam reformer/steam generator system. All applications with IHX require helium outlet temperatures from the reactor to be 900°C or higher to realize a compact IHX component with a larger temperature difference across the IHX. The design of a plant with 850°C coolant outlet temperature and IHX is also feasible, but at the expense of a larger heat transfer surface.

According to an assessment made by SNL evaluating the relative development requirements and considering criteria such as safety, operational issues, capital cost, intermediate loop compatibility, conclusions and recommendations were given for the different reactor types to be connected to high temperature H₂ production processes as shown in the following Table 3-2 [Schultz 2005].

Table 3-2: Nuclear reactor types and their potential connection to H₂ production processes

LWR, Organic-cooled reactor	Cannot achieve the high temperatures needed.
Alkali metal-cooled reactor	Development risk due to materials concern at high temperatures; Possibly appropriate for lower- temperature cycles.
Heavy metal-cooled reactor, Molten salt-cooled reactor	Promising, but significant development needed.
Gas-cooled reactor	Baseline choice; Only modest development needed for helium-cooled reactors.
Liquid core reactor	Significant development risk due to materials concern at high temperatures.
Gas core reactor	Not recommended, too speculative.

Through open and transparent reactor testing, it must be demonstrated that no severe core damage will result from plausible accidents applying

- core fuel and structural materials that do not melt;
- coolants that are not reactive;
- passive cooling and decay heat removal systems;
- operation and maintenance which is highly tolerant on human error;
- no off-site emergency response at any credible accident scenario;
- a complete solution for all waste streams with minimized quantities.

In the long term, generation of new fissile materials in breeding reactor will be needed meaning that reprocessing of spent nuclear fuel will be required.

3.2. The Intermediate Heat Exchanger as Coupling Component

3.2.1. Concept of an Intermediate Heat Exchanger

Any combination of HTGR with chemical processes will most probably need a decoupling between the primary circuit and the heat utilization system by the following reasons:

- Separation of nuclear island for safety reasons;
- Limitation / exclusion of radioactive contamination of the product (e.g., tritium);
- Exclusion of ingress of corrosive process media into the primary circuit;
- Near-conventional design of heat utilization system;
- Ease of maintenance and repair for heat utilization system;
- Exclusion of contamination of high industrial investments around the nuclear island.

The intermediate heat exchanger (IHX) is a component which provides a clear separation between the nuclear plant and the heat application. Under normal operating conditions, the IHX prevents the primary coolant from accessing the process plant and, on the other side, process gases from being routed through the reactor containment. The physical separation allows for the heat application facility to be conventionally designed, and repair works to be conducted under non-nuclear conditions. The flow diagram of the combination of primary and secondary circuit together with the respective T-Q diagram is shown in Fig. 3-2.

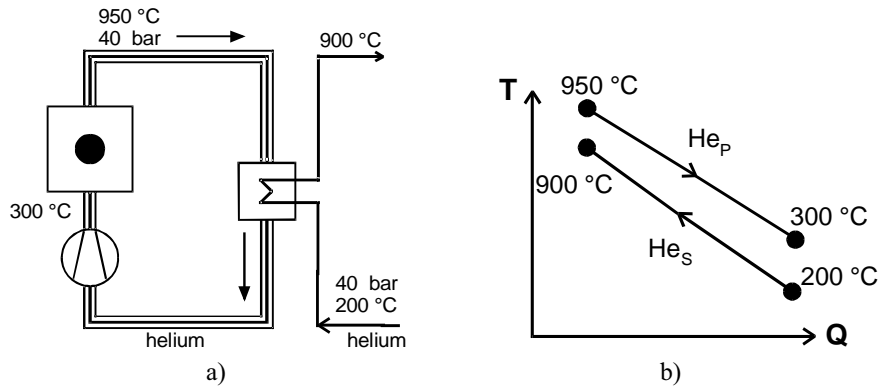


Fig. 3-2: Primary circuit of a modular HTGR with IHX
a) principle flow diagram, b) T-Q diagram for use of nuclear heat

Different technologies for heat exchanging components designated for nuclear applications have been developed in the past. For those to be applied to future nuclear systems, i.e., for high temperatures and pressures, appropriate material selection will be essential [Dewson 2005].

3.2.2. Development and Testing of IHX in Germany

The concept of a combination of the HTR-Modul with an IHX is shown in Fig. 3-3. The given example represents the German reference design of a process heat HTR-Modul in a side by side arrangement of nuclear reactor and IHX vessel for each modular unit. The thermal power of the nuclear reactor and of the IHX are 170 MW, the limitation of power is given by the requirement of self-acting decay heat removal in accidents with a total loss of active cooling. If larger thermal powers were needed, an annular core would be required. The limitation in this case is caused by the dimensions and type of reactor pressure vessel. Details of the IHX with helical tube bundle are shown in Fig. 3-4. Characteristic design data are listed in Table 3-3.

The employment of an IHX was also suggested within the PNP project with regard to the steam gasification of hard coal, for which the main characteristic data are also given in the table for comparison.

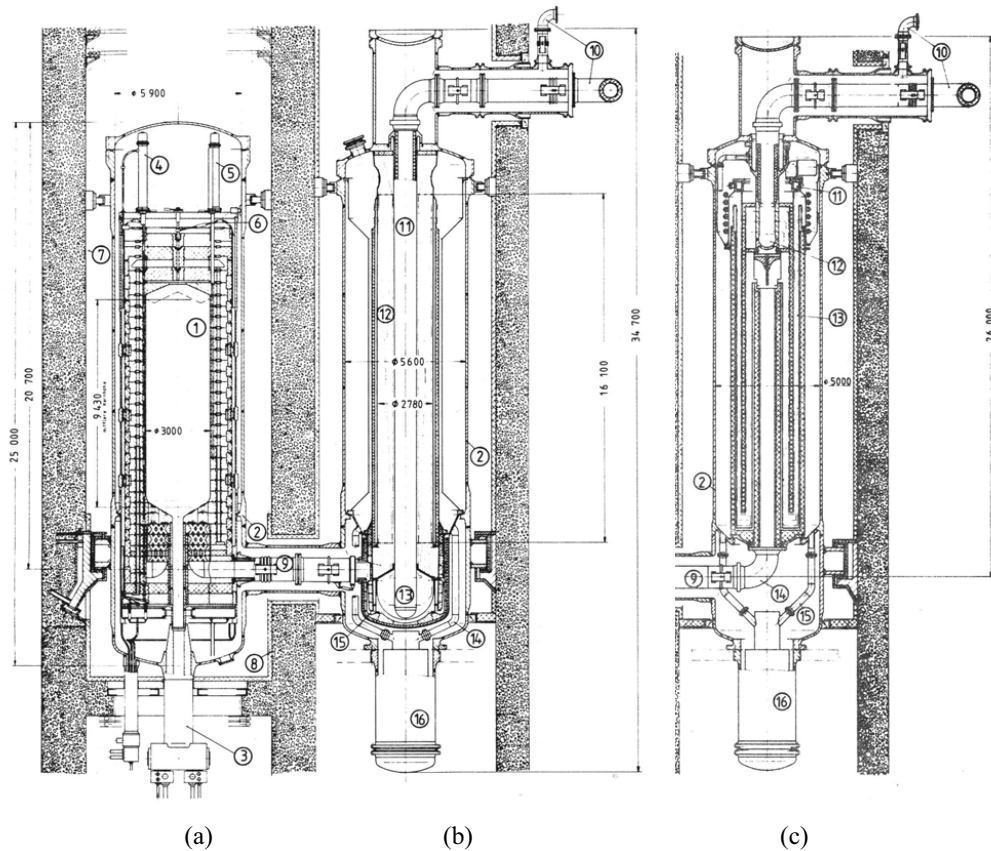


Fig. 3-3: Arrangement of HTR-Modul (a) with Helix-IHX (b) or U-tube-IHX (c) [IA 1983]

Reactor: 1-pebble bed, 2-reactor pressure vessel, 3-fuel discharge,
4-KLAK spheres shut down system, 5-reflector rods, 6-fuel loading,
7-surface cooler, 8-insulation, 9-hot gas duct.

IHX: 10-secondary pipe connections, 11-central secondary hot gas return pipe,
12-tube bundle (helical or U-tube), 13-secondary hot gas header,
14-primary cold gas duct, 15-primary hot gas duct, 16-blower.

The Helix-IHX (see Fig. 3-4) consists of a bundle of helical tubes arranged around a central return gas duct for the hot secondary helium. The support system for the tubes consists of support cylinders with star-shaped welded-on support plates serving to take the weight of the bundle. A segmental design of the support structure limits the axial relative expansion caused by the operation temperature. In the upper cold area, the mechanical loads are carried by the vessel cover. This ensures access to the secondary system for in-service inspection and repairs without the necessity to open the primary circuit.

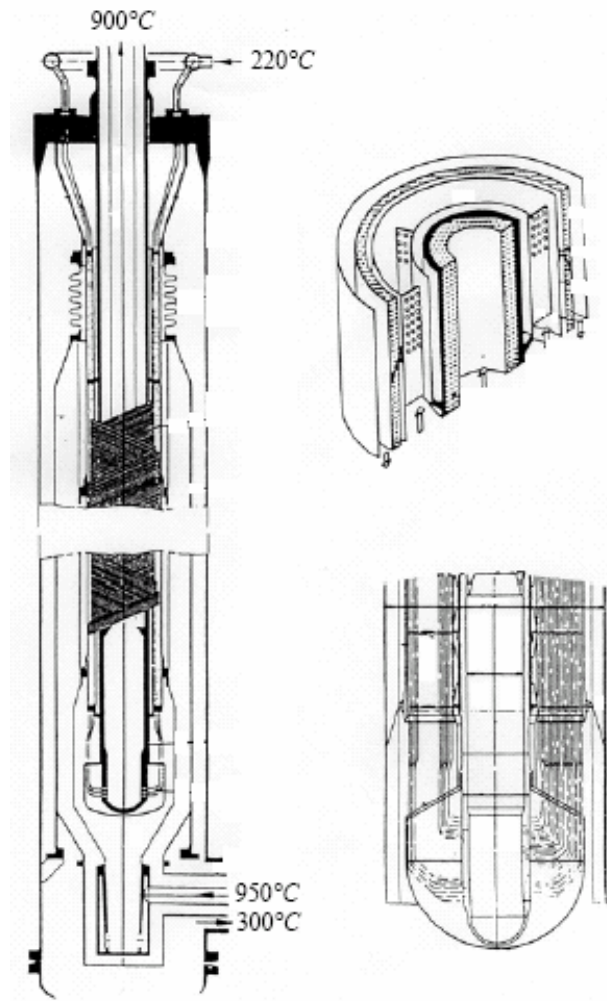


Fig. 3-4: Intermediate heat exchanger for nuclear applications [Kugeler 2005]
helical tube bundle (left), details of hot gas collector tube and support structures (right)

A facility for large component testing (KVK) was constructed and successfully operated by INTERATOM within the PNP project [Harth 1990]. In a heating system consisting of a heater with steam, a natural gas burner, and an electrical heater with a total thermal power of 10 MW, helium was heated up to 950°C at 4.0 MPa (Figs. 3-5 and 3-6). This plant also allowed the test of hot gas ducts with large diameter, of a steam generator, valves for hot helium and other components like hot headers or auxiliary plants like gas purification.

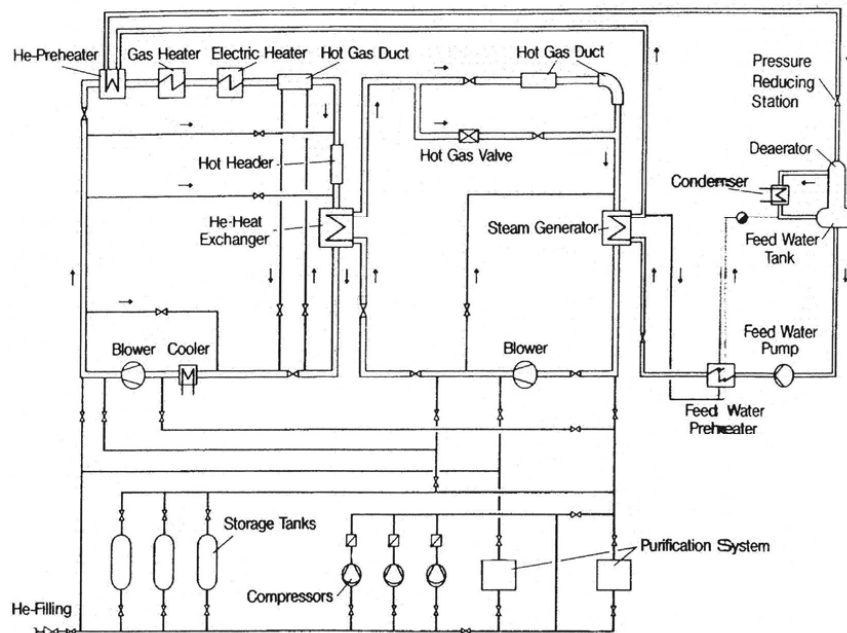


Fig. 3-5: Flow sheet of 10 MW KVK facility for testing nuclear process heat components

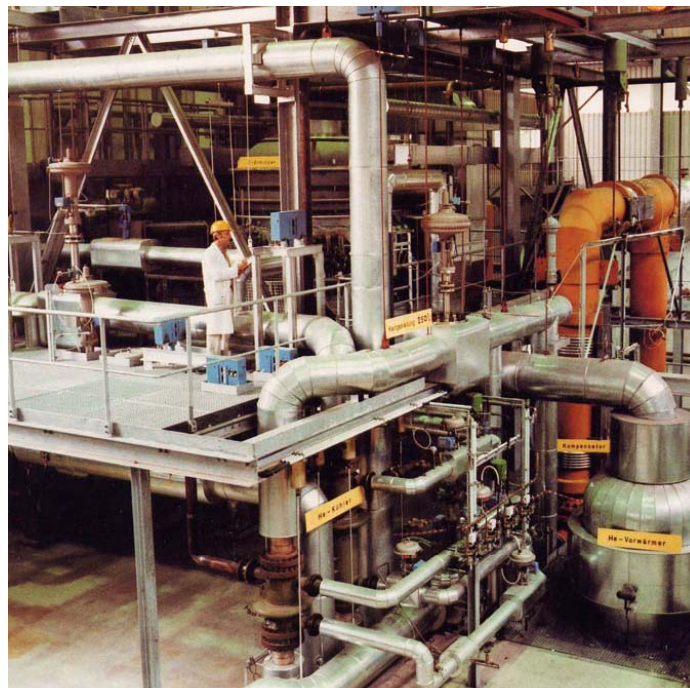


Fig. 3-6: 10 MW KVK facility for testing nuclear process heat components

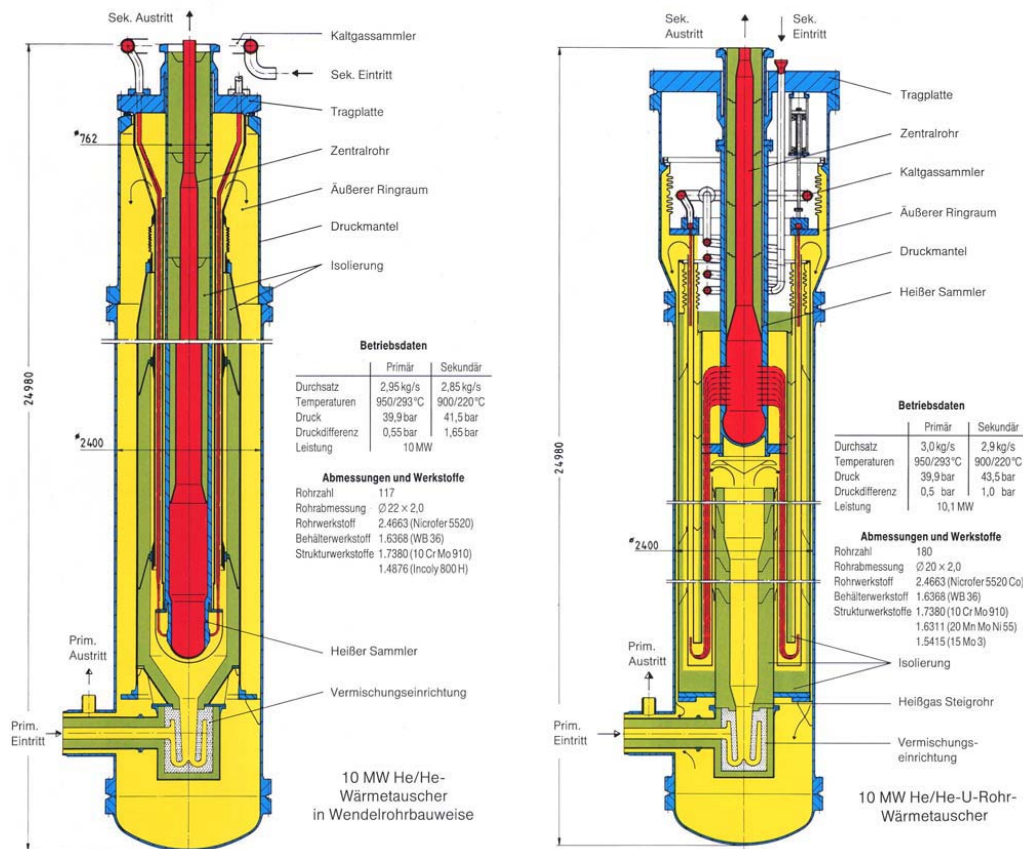


Fig. 3-7: Two IHX components tested in KVK
Helical tube bundle by Steinmüller company (left), U-tube bundle by Balcke-Dürr company (right)

Two IHX components were constructed and tested in the KVK loop, one with a helical tube bundle and another one with U-tubes. Fig. 3-7 shows schematics of both components, Figs. 3-8 and 3-9 the IHX components under construction.

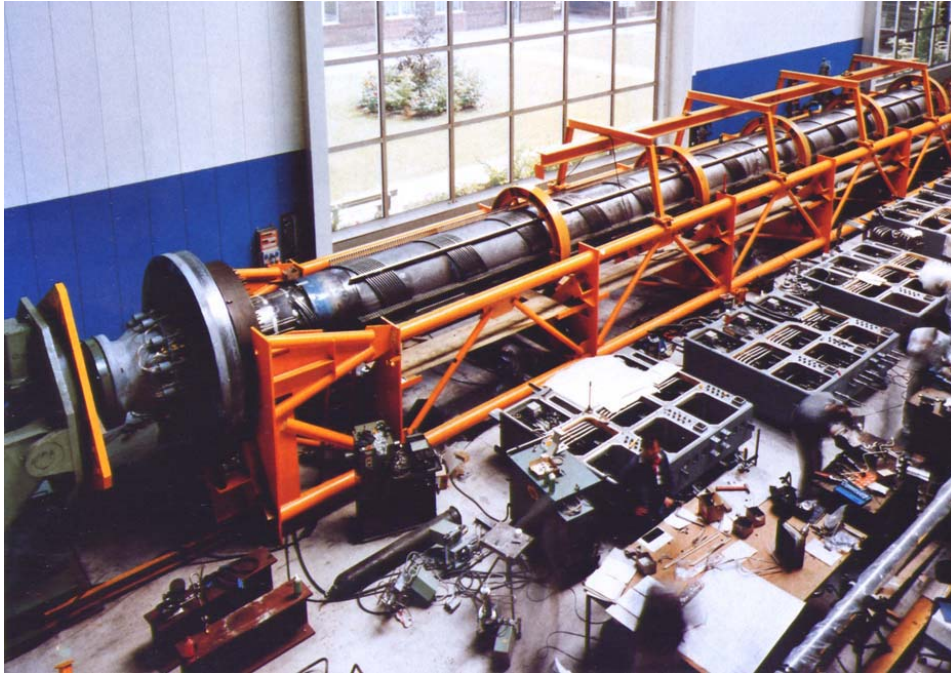


Fig. 3-8: IHX component with helical tube bundle tested in KVK (Steinmüller)

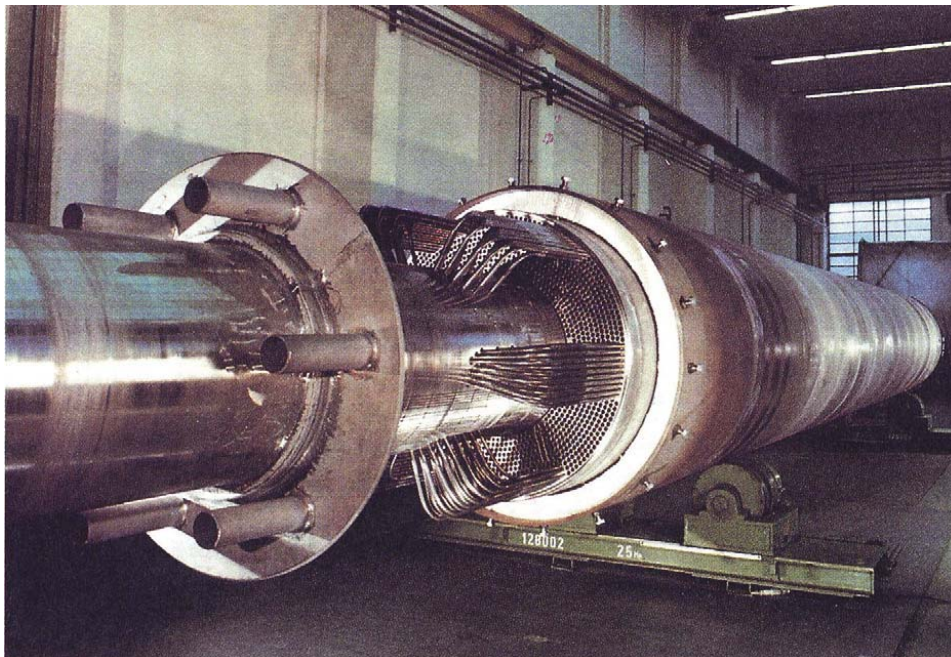


Fig. 3-9: IHX component with U-tube bundle tested in KVK (Balcke-Dürr)

Table 3-3: Comparison of data of IHX tested in KVK facility with data of IHX for nuclear application

IHX in	KVK		170 MWt HTR Modul [IA 1983]		500 MWt PNP [PNP 1981]	
	Helical	U-Tube	Helical	U-Tube	Helical	U-Tube
Heat capacity [MW]	10	10.1	171.7	171.7	125	125
Material Tube	Nicrofer 5520		Inconel 617			
Size						
Bundle outer diameter [m]	1.5	2.0	2.78	1.512	2.52	2.098(hot) / 2.815 (cold)
Tube outer diameter [mm]	22	20	22	20	22	20
Tube wall thickness [mm]	2	2	2	2	2.2	2
Tube length [m]			38.2	30.0	40.7	32.7
Number of tubes	117	180	1612	2470	1444	1900
Primary helium						
Inlet temperature [°C]	950	950	950	950	950	950
Outlet temperature [°C]	293	300	292	292	293	293
Inlet pressure [MPa]	3.99	3.99	3.99	4.05	4.0	3.99
Outlet pressure [MPa]	3.935	3.94	3.97	3.99		
Flow rate [kg/s]	2.95	3.0	50.3	50.3	36.9	36.9
Secondary helium						
Inlet temperature [°C]	220	220	200	200	220	220
Outlet temperature [°C]	900	900	900	900	900	900
Inlet pressure [MPa]	4.19	4.35	4.38	4.35	4.2	4.35
Outlet pressure [MPa]	4.025	4.25	4.19	4.19		
Flow rate [t/h]	2.85	2.9	47.3	47.3	35.6	35.6
Heat transfer area [m ²]		3500	4254	4700	4000	3900
Life time [h]	-	-	140,000	140,000	140,000	140,000

In the helical IHX, primary hot helium (950°C) entering the heat exchanger flows via a mixing and deflecting device at the bottom of the component upwards through the bundle and is cooled up to 300°C. The cooled primary helium flows back into the gap between the wall of the reactor pressure vessel and the gas shroud of the heat exchanger to the blower at the bottom of the component.

The secondary helium is entering the component at the top. After entering into a ring conduit, the helium with a temperature of 200 °C is uniformly distributed over the tube bundle and heated up to 900°C in counter flow. The cycle is closed by the hot header which is insulated on the inside. The hot helium is leaving the IHX at the top of the component.

The maximum wall temperatures in the tubes in normal operation are 920°C, the maximum pressure difference between primary and secondary side is 0.2 MPa under operational conditions. In depressurisation accidents, they have to withstand the full pressure difference in a limited time period.

The relevant data of the IHX test components are contained in Table 3-3 in comparison to the data of an IHX component to be connected to the two German concepts of a process heat nuclear reactor. Each component had a thermal power of 10 MW. The most critical part in the design of a helical tube concept, the hot header for the secondary helium, was tested in the 10 MW component in a 1:1 scale related to the large component. Both components were operated over several 1000 h without any difficulty.

The table shows that the specific data of the test components were very similar to those which are planned for the nuclear application. Gas temperatures, pressures and material temperatures in the KVK facility were even identical to those of the nuclear design.

Especially hot gas samplers as the man sensitive large components of such an heat exchanger have been tested in 1/1 scale. The tube systems had the same dimension in test and for nuclear application. The materials for the tubes, the headers, supporting structures and for the gas ducting on the primary side were the same as planned for the nuclear application.

On the basis of a broad experimental program in the KVK-plant and other related test facilities (HHV plant, EVO plant, insulation test facilities, specific facilities to test, e.g., friction, fretting, wear, materials, depressurization), the following statements can be made with respect to the feasibility of the IHX:

- Two helium heated IHX have been successfully tested on the 10 MW power level and at maximum helium heating temperatures of 950°C (primary) and 900°C (secondary), respectively. So far, the process parameters have been tested under real conditions of the nuclear application. The operation time of the helical tube bundle was more than 5000 h, that of the U-tube bundle was more than 4000 h.
- Parallel to the integral tests of the components, additional testing in KVK was carried out for a hot gas header of the helical tube bundle, for hot gas ducts (including bends and expansion bellows), hot gas valves, and a steam generator.
- The thermodynamic data of the heat exchanger designs have been confirmed by the experiments. Average heat fluxes of around 40 kW/m² can be realized at reasonable pressure drops.
- The components were tested in steady state operation and under transient conditions. Transients of ± 7 K/min were tolerated by the components without failure.
- Tests have shown leak tightness between the primary and secondary side of the IHX.
- The measured vibrations did not result in serious loads to the heat exchanger tubes.
- The bearing forces for the load transfer system of the cold header and the bundle were within the range of calculated values.
- Ultrasonic inspection of IHX tubes after 4700 h of operation did not reveal any deviations.

- Creep buckling tests on the hot gas header were carried out.
- As many as 656 cycles from 950°C to 710°C and reverse with a transient of ± 40 K/min did not cause any damage to the header.
- Tests with a pressure difference of 4.3 MPa between primary and secondary side of the hot gas header at 970°C were carried out over a time period of 455 h to simulate a depressurization accident. No damaging influence was identified.
- Insulations and gas ducting structures inside the tube bundles did not show any damage.

Overall it was stated that an IHX following one of the two designs investigated (helical tubes and U-tubes) can be designed for a power of 170 MW and can be operated for 100,000 h on the basis of the available experience. Especially the extensive wall material testing for high temperature alloys at a temperature of 950°C showed that this component can be applied in connection with process heat HTGRs (see Fig. 3-10).

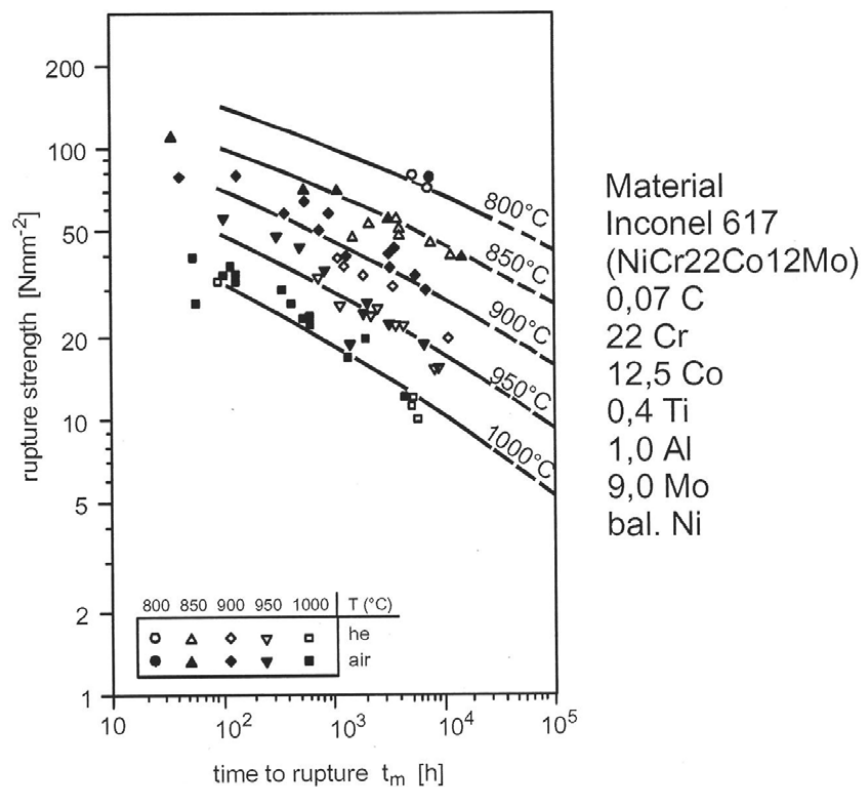


Fig. 3-10: Material diagram (source: FZJ)

Much work has been dedicated to the development of a design philosophy for components at these high temperatures. However, it still needs additional work for nuclear applications and qualification of these methods.

3.2.3. Testing of the Japanese IHX in the HTTR

The IHX used in the HTTR is a vertical, helically coiled counter flow type heat exchanger as shown in Fig. 3-11. 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.

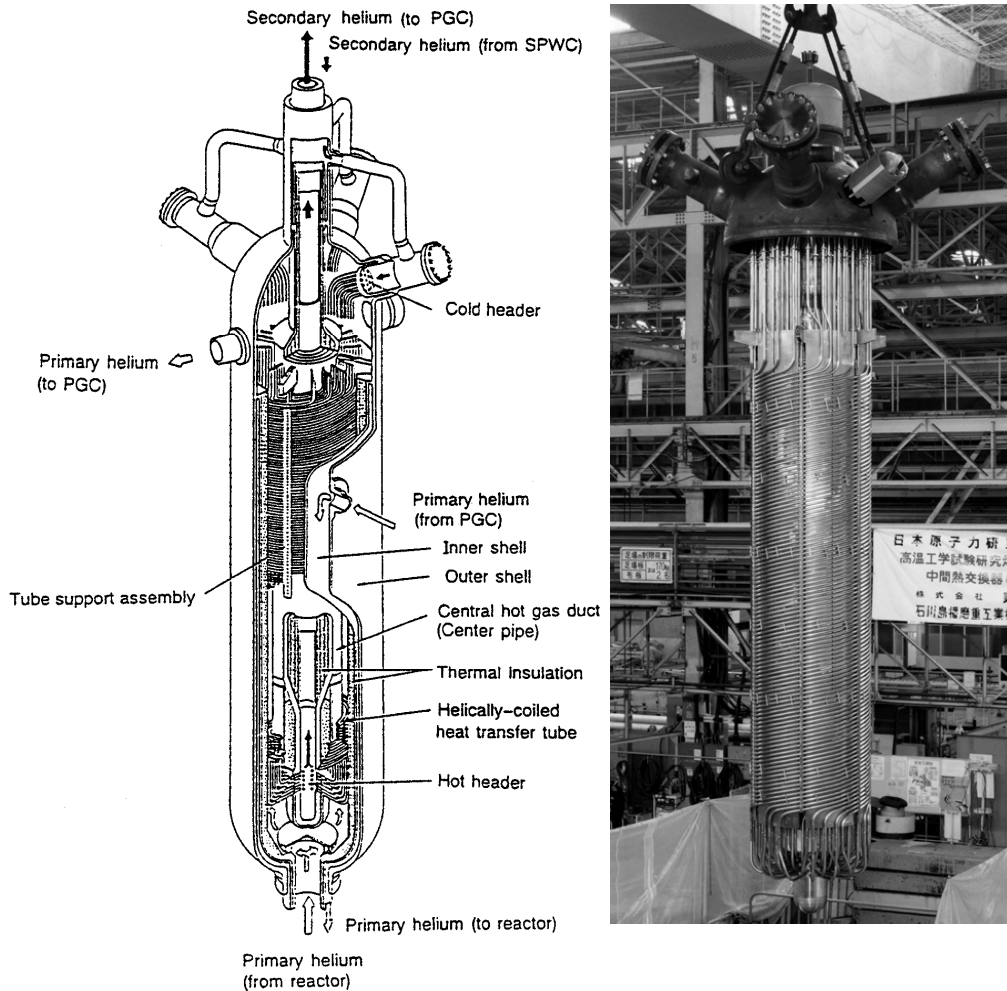


Fig. 3-11: Schematic and photograph of the He-He intermediate heat exchanger in the HTTR

A double-walled shell with a thermal insulation attached on the inside surface of the inner shell provides reliable separation of heat resisting and pressure retaining functions. Cold helium flowing through the annulus brings uniform temperature distribution throughout the outer shell, which has a

pressure-retaining function. Insulation inside and outside the central hot gas pipe keeps the heat transfer low to obtain a high efficiency.

To minimize constraints of axial and radial thermal expansions of the helically coiled heat transfer tubes, a floating hot header combined with the central hot gas duct and passing through the central space inside the helix bundle, is adopted. A tube support allows free thermal expansion of a helix in radial direction. Table 3-4 shows the major design specifications of the IHX as operated in the HTTR [Saito 1994] and as projected for the combined cycle version of the GTHTR300 [Yang 2005].

Table 3-4: Design specifications of the He-He Intermediate Heat Exchanger in the HTTR

Parameter	IHX in HTTR	IHX in GTHTR300C
Type	Counter-current, helically wound tube type	Helical
Heat capacity [MW]	9.94	168
Material Shell Tube Thermal insulation	2.25Cr-1Mo steel Hastelloy XR Kaowool 1400SHA	Hastelloy XR
Size Inner / outer shell diameter [m] Shell height [m] Tube outer diameter [mm] Tube wall thickness [mm] Tube length [m] Number of tubes	1.352 / 2.0 11.0 31.8 3.5 22 96	45 5 14 724
Maximum temperature Shell [°C] Tube [°C]	430 955	
Maximum pressure [MPa] Shell Tube (differential pressure)	4.81 0.29	
Primary helium Inlet temperature [°C] Outlet temperature [°C] Design pressure [MPa] Design pressure drop [kPa] Flow rate [kg/s]	950 389 4.06 9.2 3.4	950 850 5.02
Secondary helium (design, nominal) Inlet temperature [°C] Outlet temperature [°C] Design pressure [MPa] Design pressure drop [kPa] Flow rate [kg/s]	273 869 4.21 50.2 3.0	500 850 5.15
Design Lifetime [yr]	20	20

Mitsubishi Heavy Industries has recently developed an IHX concept for the Japanese GTHTR300 reactor design. It is of a plate fin type and consists of Hastelloy X. It is designed for a heat capacity of 105.5 MWt. Primary helium inlet/outlet temperatures are 950/340°C at a pressure of 7.9 MPa. The secondary helium inlet/outlet temperatures are 290/900°C at a system pressure of 8.0 MPa [Kamito 2005].

3.2.4. Conceptual Design of IHX for the US H₂-MHR

In the United States, the reference design for a next generation HTGR is the 600 MWt Modular Helium Reactor (MHR). For the purpose of hydrogen generation, the concept of an H₂-MHR has been developed which is to be coupled to a H₂ production technology based on either S-I thermochemical cycle or high temperature electrolysis. The intermediate heat exchanger for this facility is based on a design of so-called “Printed Circuit Heat Exchangers”, PCHE, developed by the Heatric company (see Fig. 3-12) [NERI 2003, HEATRIC].

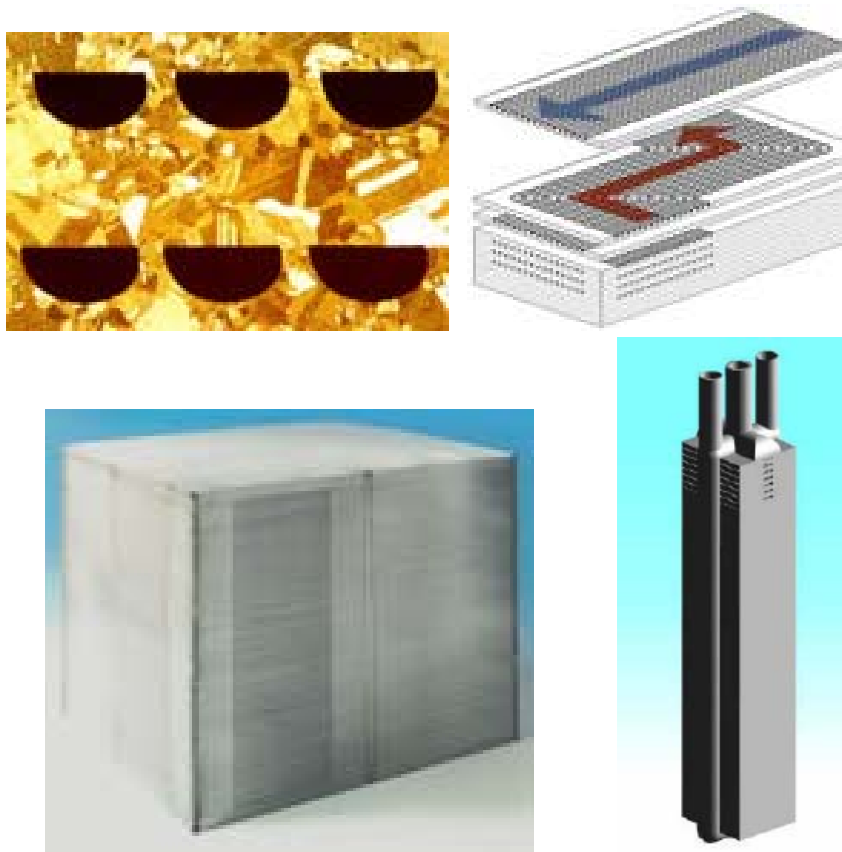


Fig. 3-12: Printed Circuit Heat Exchanger, PCHE (source: HEATRIC)

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 semi-circular 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 metal plates are stacked (top right) and then diffusion-bonded, where the metal surfaces are pressed together promoting a grain growth between the plates, thus becoming a solid all-metal core (bottom left). PCHE designs have been developed which are highly compact, highly robust (bottom right) and which have high thermal efficiencies, allowing pressures of 50 MPa and temperatures of 900°C. The basic modules can be taken to construct heat exchangers to any desired scale. The characteristic data of the IHX are listed in Table 3-5.

Table 3-5: Design specifications of the He-He Intermediate Heat Exchanger for the US H2-MHR concept [NERI 2003]
(numbers in parentheses indicate recent design changes [Richards 2005a])

Parameter	IHX in H2-MHR
Type	Printed Circuit Heat Exchanger
Heat capacity [MW]	664 (600)
Material	Alloy 800H materials
Size of vessel	
Diameter [m]	6.9
Active heat transfer height [m]	8
Wall thickness [mm]	2
Size of module	
Length [m]	0.333 (0.6)
Width [m]	0.196 (0.65)
Height [m]	2 (1.5)
Number of modules	120 (40)
Primary helium	
Inlet temperature [°C]	1000 (950)
Outlet temperature [°C]	636 (590)
Design pressure [MPa]	7.0
Design pressure drop [kPa]	40.7
Flow rate [kg/s]	320
Secondary helium	
Inlet temperature [°C]	450
Outlet temperature [°C]	975
Design pressure [MPa]	7.1
Design pressure drop [kPa]	13.1
Flow rate [kg/s]	222
Heat transfer area [m ²]	5230

For the S-I cycle based H2-MHR, the IHX as planned at the moment will consist of 40 modules of “Printed Circuit Heat Exchangers” (PCHE) with each module transferring 15 MWt. All modules plus associated manifolds will be placed into an insulated steel vessel of a size similar to the MHR vessel. For the HTE based H2-MHR, the IHX component would be much smaller due to the much less heat being transferred. The IHX could be either composed of PCHE units or follow the more conventional helical coil design [Richards 2005a].

3.2.5. IHX Designs Considered in the ANTARES Concept in France

The AREVA NP concept of a combined cycle HTGR, ANTARES, is expected to include a compact design of an IHX. The new designs currently under investigation are [Breuil 2006]

- a plate machined heat exchanger, and
- plate fin heat exchanger.

The design specifications of the present pre-conceptual phase for the IHX component of ANTARES are listed in Table 3-6.

Table 3-6: Pre-conceptual design specifications of the plate-type Intermediate Heat Exchanger for the French ANTARES concept [Breuil 2006]

Parameter	IHX in ANTARES
Type	Plate machined or plate fin
Heat capacity [MW]	608
Material	Alloy 230, Alloy 617
Size of vessel [m ³]	
PMHE	25
PFHE (Brayton Energy)	10
PFHE (Nordon)	30
Primary helium	
Inlet temperature [°C]	850
Outlet temperature [°C]	350
Design pressure [MPa]	5.5
Design pressure drop [kPa]	< 0.1
Flow rate [kg/s]	240
Secondary helium/nitrogen mixture	
Inlet temperature [°C]	300
Outlet temperature [°C]	800
Design pressure [MPa]	~ 5.5
Design pressure drop [kPa]	< 0.2
Flow rate [kg/s]	614

The plate machined heat exchanger (PMHE) consists of single plates of 1 mm thickness fabricated by drilling or electro-chemical etching, and stacked together by diffusion bonding. Since the modules are relatively stiff, their height is limited to the order of 1 m. The total volume required for ANTARES is about 25 m³.

Two variants are pursued for the plate fin heat exchanger (PFHE). The one proposed by Brayton Energy has wavy or straight fins on a flat support plate. The independent cells (see Fig. 3-13, left) allow an arrangement for the IHX module which is flexible under thermal loads. Using flow channels with 0.3 mm hydraulic diameter and a high fin density, a high compactness ($\sim 10 \text{ m}^3$) could be achieved. The second variant from Nordon (see Fig. 3-13, right) employs a different type of fins, serrated offset strip fins, on a support plate.

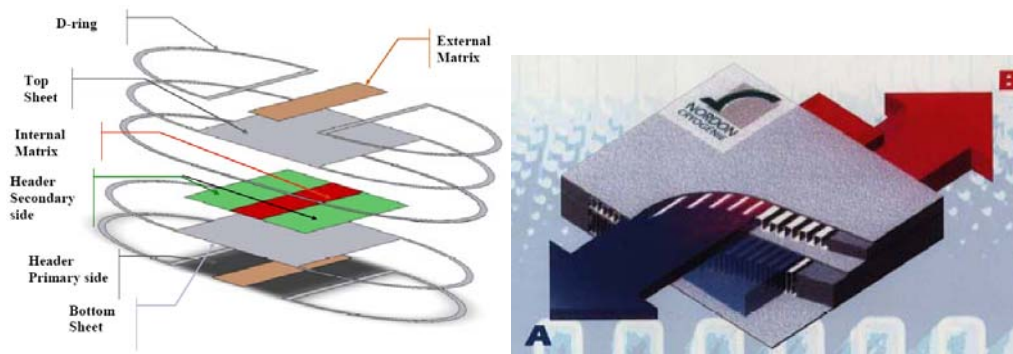


Fig. 3-13: Two variants of plate fin IHX [Breuil 2006]
Brayton Energy design (left) and Nordon design (right)

Fig. 3-14 shows a potential design of an IHX vessel for ANTARES containing in a symmetrical arrangement eight plate-type IHX modules.

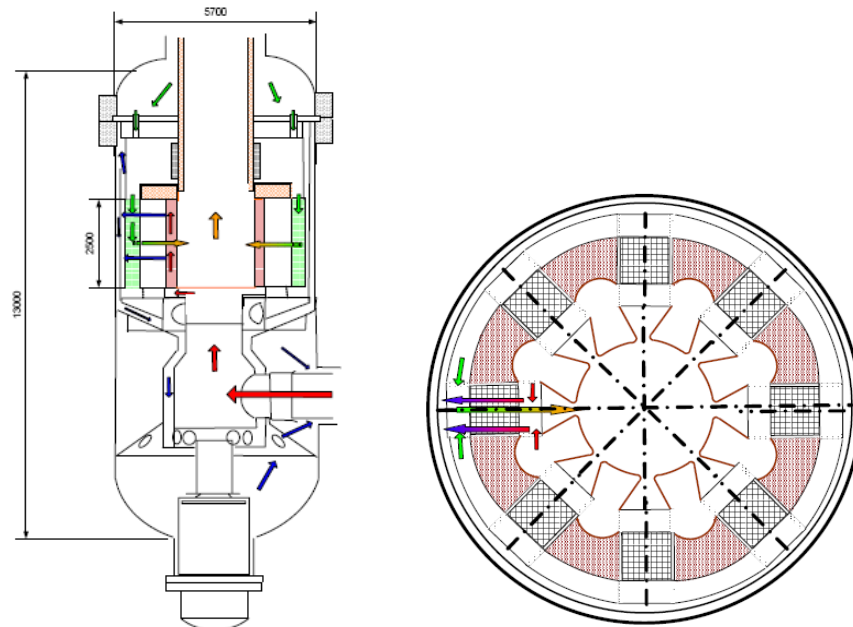


Fig. 3-14: IHX vessel with integrated plate IHX modules [Breuil 2006]

3.3. Nuclear Steam Reforming

3.3.1. Concept and Feasibility of a Nuclear Steam Reformer

In comparison to a conventional steam reformer, the employment of a nuclear steam reformer requires certain changes, since operational conditions of a nuclear reactor are not that flexible as a fossil-fueled furnace. Also safety requirements are much more stringent than for a fossil-fueled system. It is therefore desired to achieve highest effectiveness in utilizing the nuclear process heat in the whole production process system.

A large H_2 production rate is achieved, if the process feed gas rate and the conversion rate are high. The feed gas rate depends on the amount of heat input into process gas and the temperature of process gas. The conversion rate depends on temperature and pressure of the process gas.

A principle flow sheet of an HTGR with steam reformer, where all heat for the reforming process, the steam production, gas purification, and gas compression can be gained from the helium circuit, is shown in Fig. 3-15. The stages of gas purification include the shift conversion, CO_2 scrubber, H_2 separation, and a methanation reaction to remove traces of carbon oxides from the process gas. The typical temperature profiles along a splitting tube are shown in Fig. 3-16.

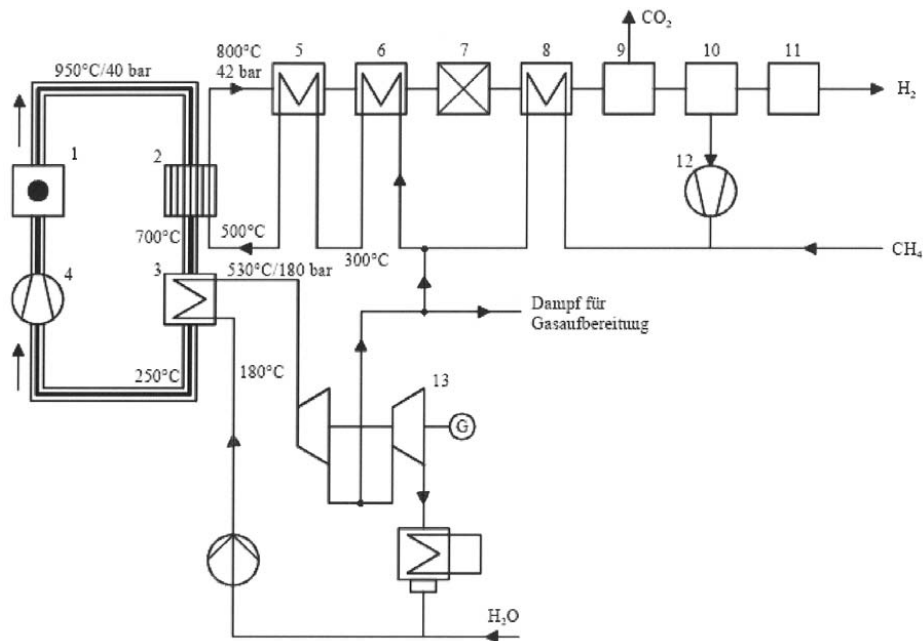


Fig. 3-15: Principal flow sheet of HTGR with SR [Kugeler 2005]

1-HTGR, 2-steam reformer, 3-steam generator, 4-He blower, 5-preheater gas, 6-preheater gas, 7-shift conversion, 8- CH_4 preheater, 9- CO_2 washer, 10- H_2/CH_4 separation, 11-methanation, 12- CH_4 compressor, 13-steam turbine plant

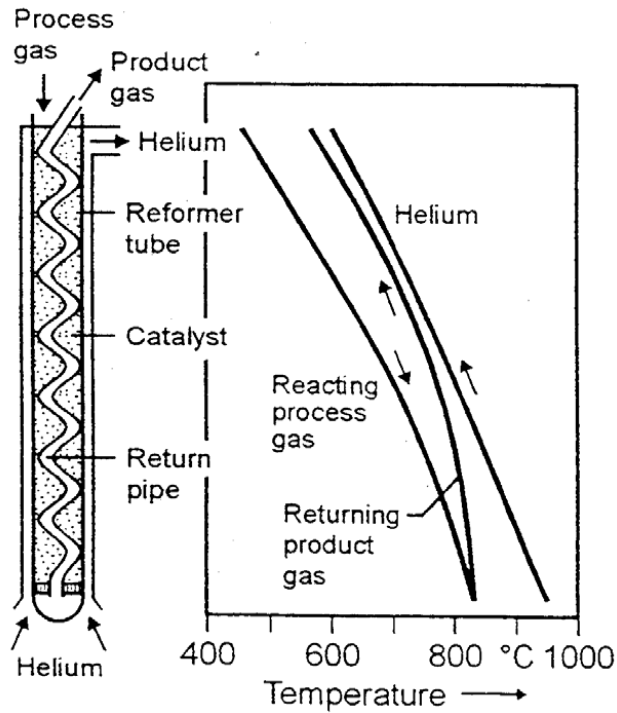
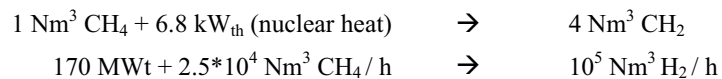


Fig. 3-16: T-diagram for use of heat in HTGR with SR [Kugeler 2005]

The steam reformer uses the temperature of the helium (4.0 MPa) between 950 and 700°C, while the steam generator is using that part of heat between 700 and 250°C. The feed gas mixture with an $\text{H}_2\text{O}/\text{CH}_4$ ratio of ~ 3 and a pressure of ~ 4.0 MPa is preheated up to around 500°C and reformed at a maximum process temperature of 800°C. A fraction of 85 % of the methane is then converted in this first step. Utilization of the heat of the reformer gas for preheating the feed gas, shift conversion, and methanation are the steps following the reformer to finally get the product hydrogen. A steam turbine plant using CO generation supplies the needed steam and electrical energy for the whole process. The overall energy balance delivers roughly the following numbers:



CH_4 as raw material is completely converted to hydrogen, the total efficiency including the nuclear heat is around 65 %. A complete life cycle analysis has even revealed that depending on operating conditions, about 40 % savings of natural gas feedstock could be achieved, if nuclear is selected the primary energy source [Spath 2001].

3.3.2. The German Approach of Helium-Heated Steam Reforming

In the German “Prototype Nuclear Process Heat”, PNP, project, the steam reformer component was included directly within the primary circuit. The employment of an IHX was deemed unnecessary. This arrangement poses much more stringent requirements to this component and its reliability than is in the case of an indirect cycle via an intermediate heat exchanger as was pursued in the Japanese HTTR project. The direct coupling to the steam reformer results in a simplified design of a process heat HTGR.

On the other hand, it could be advantageous to have a clear separation of nuclear reactor and hydrogen plant, to avoid difficult procedures in the licensing process. In principle, the question of introducing an IHX requires an optimization analysis of cost, operation behavior, safety, and licensing. The technical feasibility of the indirect coupling via IHX is governed by the components of the intermediate circuit which induces a stronger decoupling of the nuclear island from the chemical conversion plant. This design implies, however, that the maximum temperature of the reforming process is reduced at least by 50°C which is the expected temperature difference for the heat transfer inside the IHX (see also Fig. 3-2b). The direct coupling of reactor and steam reformer may at present be regarded as a long-term option.

Unlike the former 200 MWt HTR-Modul design with steam cycle and coolant outlet temperatures of 750°C, the modular pebble bed HTGR for process heat production has been designed for a thermal power of 170 MW to deliver helium temperatures of 950°C without violation of the 1600°C criteria for the self-acting decay heat removal. More than 10 years of successful and high-availability operation of the AVR at an average helium outlet temperature of 950°C have demonstrated that this temperature level is state of the art for a pebble bed HTGR core. The thermal power was reduced due to the requirement of self-acting decay heat removal from the core, i.e., for the maximum fuel temperature to stay below 1600°C in case of a loss-of-forced-convection accident. Without IHX, the helium coolant is directly fed to the steam reformer which consumes 71 MW and to the steam generator which is operated with 99 MW. Fig. 3-17 shows the 170 MWt HTR-Modul directly connected to a steam reformer.

Details of the concept of the steam reformer as a new nuclear component are shown in Fig. 3-18. It is a bundle consisting of straight tubes connected to an upper supporting plate. The tubes filled with a catalyst contain an internal recuperator and an inner return duct for the hot process gas. The upper part of the component includes the collector structures for the feed gas composed of steam and methane, and for the product gas containing H₂, CO, CH₄, CO₂ and steam. From the total heat transferred into the steam reformer, 85 % are used for the reforming process, while 15 % are taken to heat up the feed gas. The main characteristic data of the steam reformer component as was designed by INTERATOM on the basis of a simple cylindrical pebble bed core with a power of 170 MWt are listed in Table 3-7. They are compared with the respective data for the steam reformer designed for the 500 MWt PNP reactor.

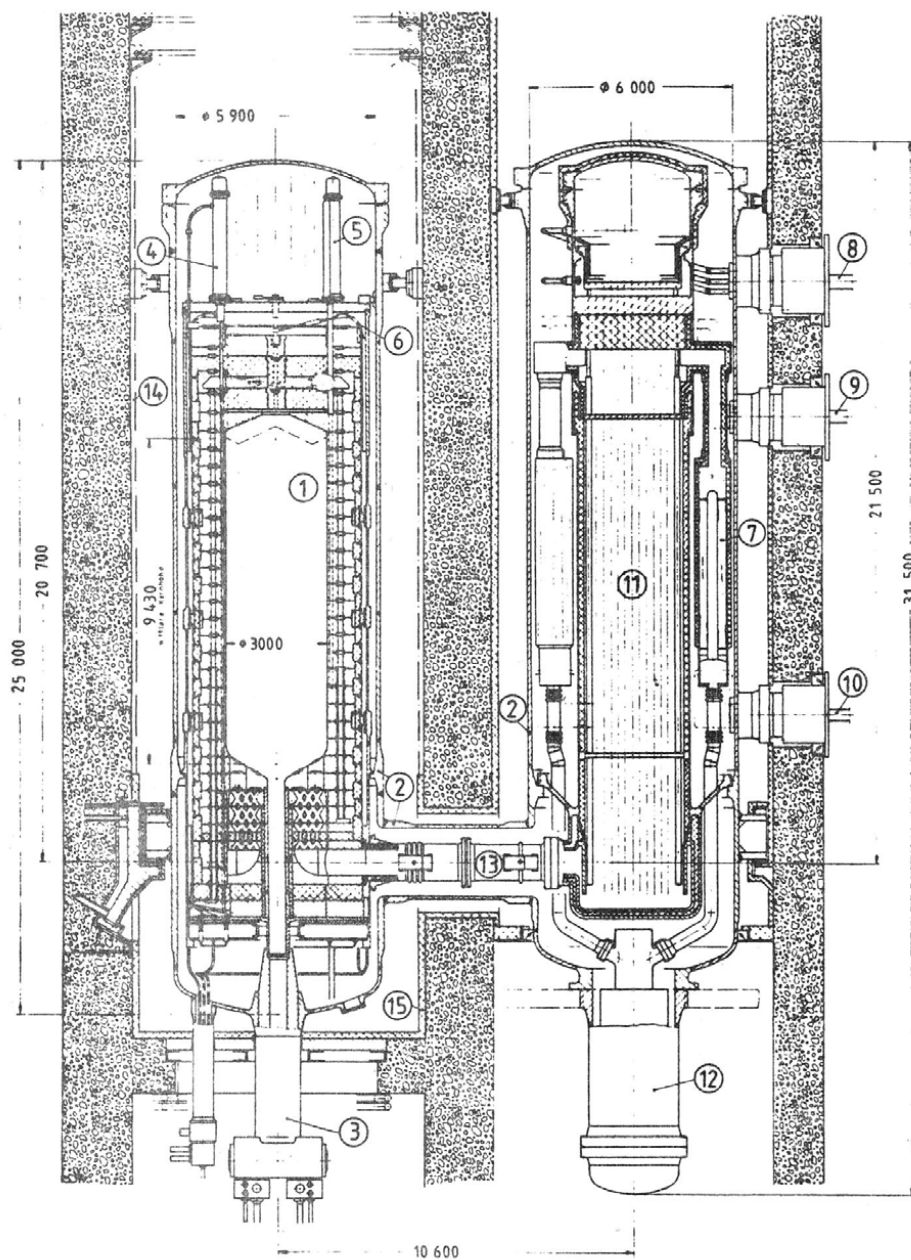


Fig. 3-17: Arrangement of HTR-Modul with splitting tube furnace and steam generator [IA 1983]

- 1-pebble bed, 2-reactor pressure vessel, 3-fuel discharge,
 4-KLAK spheres shut down system, 5-reflector rods, 6-fuel loading,
 7-steam generators, 8-reformer gas connections, 9-life steam pipe, 10-feed water pipe,
 11-splitting tubes, 12-blower, 13-hot gas duct, 14-surface cooler, 15-insulation.

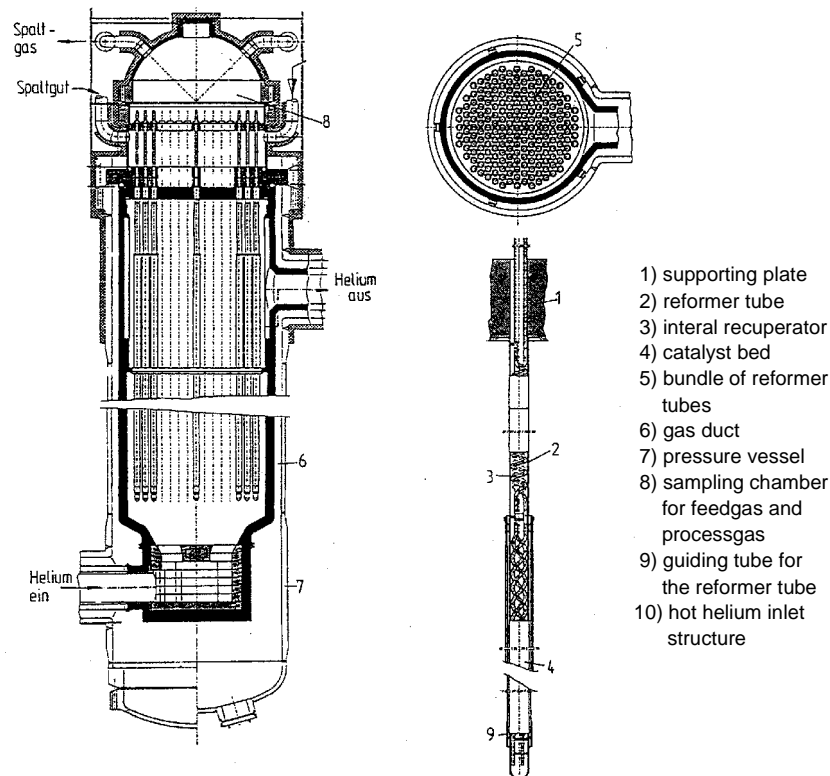


Fig. 3-18: Technical concept of a helium heated steam reformer connected to a modular process heat HTGR

Nuclear steam reforming of methane was subject of extensive R&D activities in Germany in the 1970-1980s. Within the NFE (Nuclear Long-Distance Energy Transportation) project, large test facilities were constructed and successfully operated at FZJ to study the steam reforming process and also the reverse process of methanation under nuclear conditions.

Steam reforming was investigated in the EVA single splitting tube test facility, later in EVA-II representing bundles of reformer tubes. The latter used an electrical heater with a power of 10 MW to heat up helium gas to a temperature of 950°C at 4.0 MPa (see Fig. 3-19, top). In the connected steam reformer, the heat between 950°C and 650°C was used to run the steam reforming process. In the connected steam generator (helical tubes, power 4 MW), the helium heat was used up to 350°C. Via a helium circulator, the cold helium was routed back to the electrical heater. The circuit was operated under nuclear conditions at a lower power level, but with a full-scale SR component. Also the process gas handling system was the same as in a nuclear plant.

In a methanation plant, ADAM, the product gas was reconverted to methane and steam, thus completing a closed cycle without any CO₂ emissions (see Fig. 3-19, bottom). With regard to the power input of 10 MWe, the heat release rate achieved in the ADAM plant was 5.3 MWt. The so-called ADAM & EVA system represents a long-distance chemical heat transportation system based on hydrogen as the energy carrier.

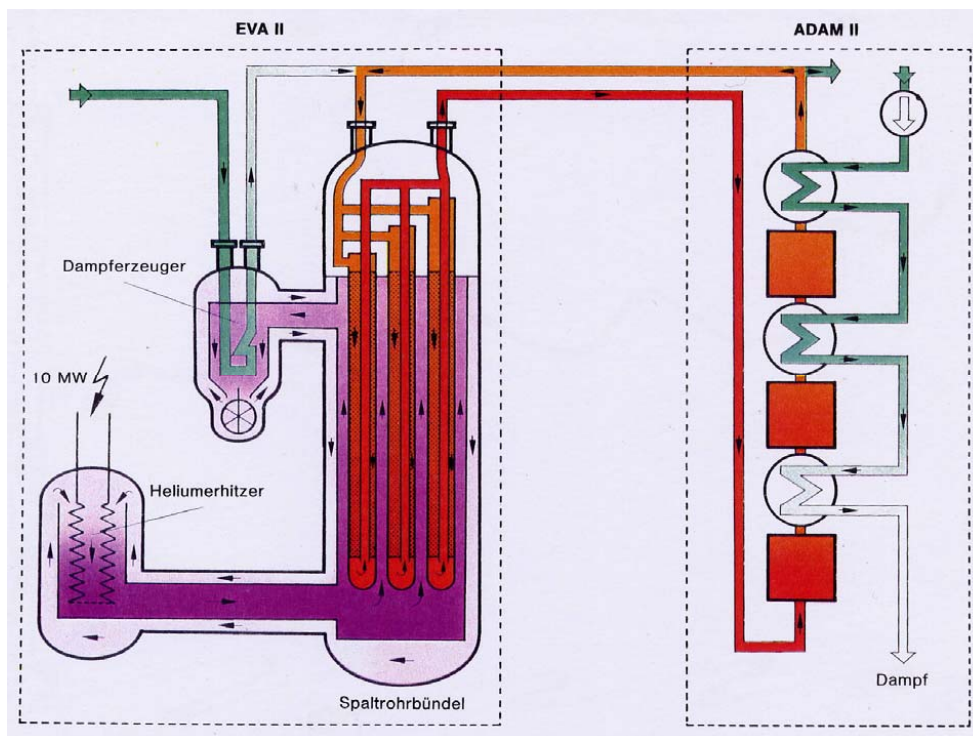
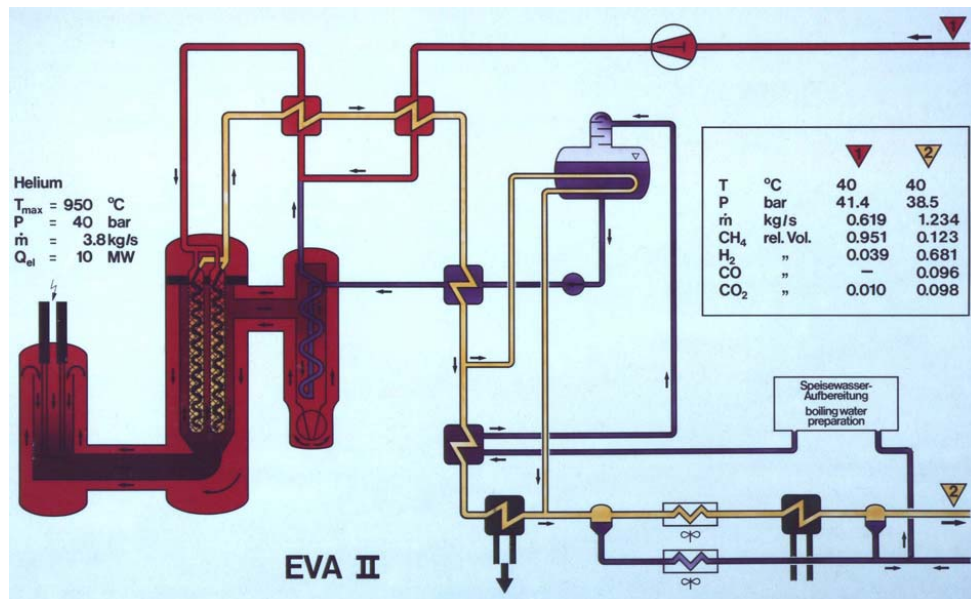


Fig. 3-19: Flow sheet of steam reforming test facility EVA-II (top)
and of combined test facilities EVA-II and ADAM-II (bottom) (source: FZJ)

Two reformer bundles have been tested in EVA-II facility: a bundle with guiding tubes (annulus design) for each reformer tube (5 MW, 18 tubes) and a bundle with baffle structures (disks and doughnuts) on the helium side (6 MW, 30 tubes). The tubes and catalytic system were 1:1 scale compared to components planned for nuclear applications. Also the loads imposed on the supporting structures were characteristic were characteristic to the nuclear case. Both have operated without any difficulties for more than 6000 h.

Fig. 3-20 shows the component with guiding tubes tested in EVA-II. Both designs of a steam reformer bundle are shown in Fig. 3-21. Data of the test objects are contained in Table 3-7 in comparison to the design data of the steam reformer for the nuclear reactor. It shows that the specific data of reformer tubes investigated were very similar to the respective design for nuclear applications.

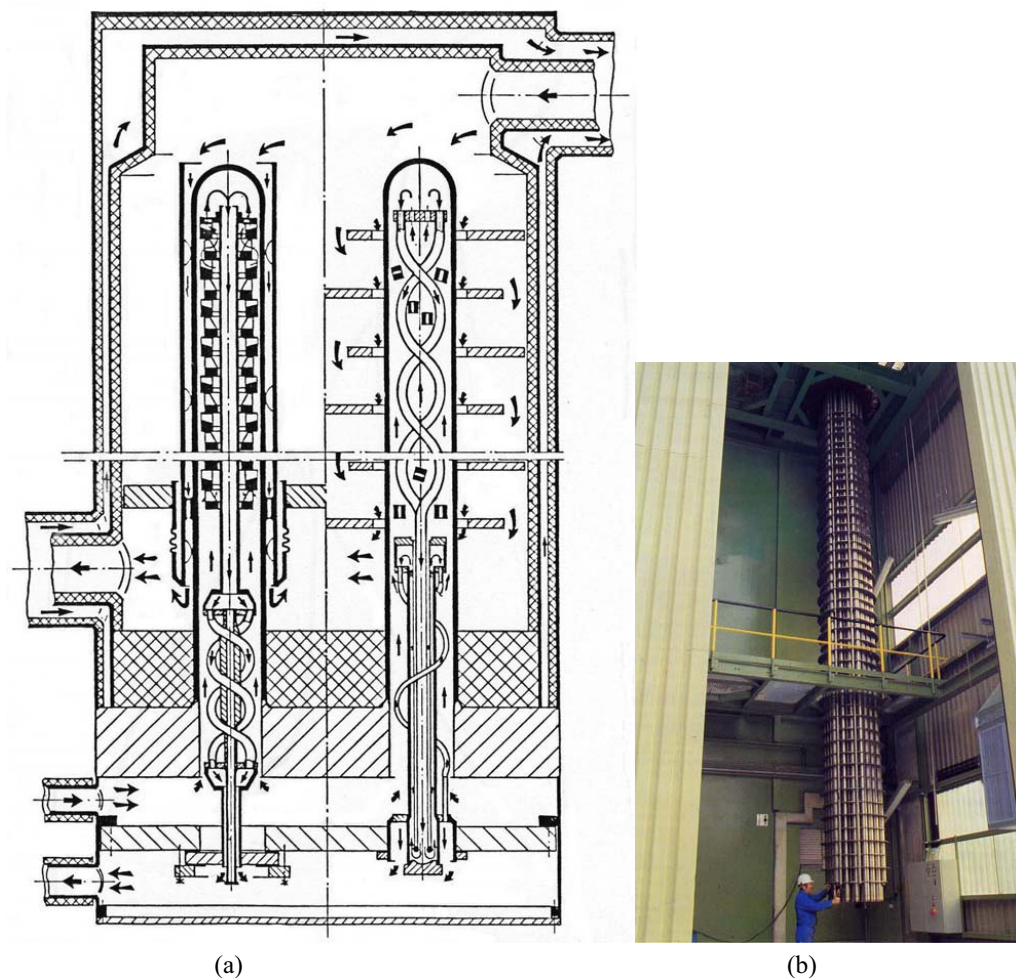


Fig. 3-20: Steam reformer bundles tested in EVA-II (source: FZJ)
 (a) schematic of annulus design (left), baffle design (right)
 (b) photograph of baffle design steam reformer bundle

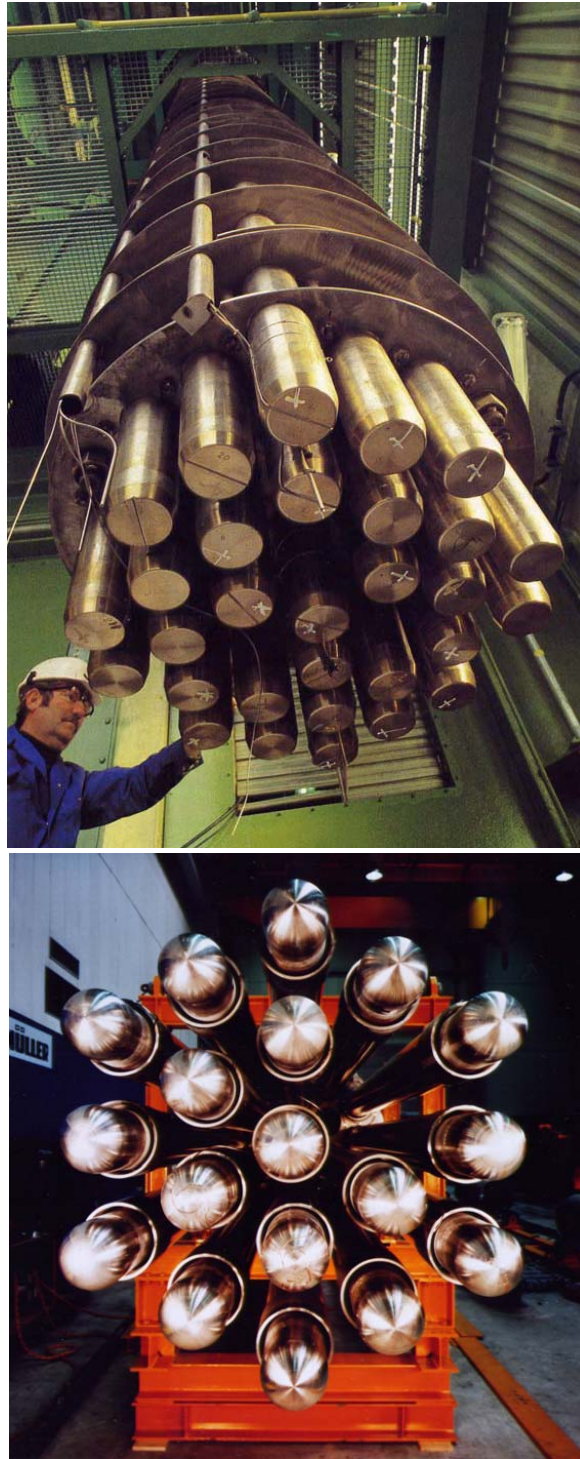


Fig. 3-21: Steam reformer bundle of baffle design (top) and annulus design (bottom)

Table 3-7: Comparison of data of steam reformer in EVA-II test facility and in nuclear application

Parameter	Test Component EVA-II		Nuclear Steam Reformer (no IHX)	
	Annulus design [Niessen 1988]	Baffle design [Niessen 1988]	170 MWt HTR Modul [IA 1983]	500 MWt PNP [PNP 1983]
Nuclear heat input [MWt]	5	~ 6	60.2	95.85
Material Tubes	Inconel 617 Incoloy 800 H	Incoloy 800 H	Inconel 617	Inconel 617
Size Shell diameter [m]	1.3	1.3	2.8	3.8
Catalyst tube				
Outer tube diameter [mm]	120	130	120	120
Wall thickness [mm]	10	15	10	10
Length [m]	13	11.5	14	17
Number of tubes	18	30	199	295
Catalyst	Raschig rings	Raschig rings	Raschig rings	Raschig rings
Primary helium				
Inlet temperature [°C]	950	950	950	950
Outlet temperature [°C]	700	650	720	700
Inlet pressure [MPa]	4	4	4.987	3.997
Outlet pressure [MPa]	~ 4	~ 4	4.947	3.960
Flow rate [kg/s]	~ 3.8	~ 3.8	50.3	73.8
Process feed gas				
Temp. inlet recup. [°C]	330	330	347	330
Temp. outlet catalyst [°C]	800	800	810	810
Pressure inlet recup. [MPa]	4.0	4.0	5.6	4.5
Raw gas flow rate [kg/s]			34.8	52.2
Steam-methane ratio	4	4	4	4
Product gas				
Temp. inlet inner tube [°C]	800	800	810	810
Temp. outlet recup. [°C]	450	450	480	462
Pressure outlet recup [MPa]			5.1	4.06
Hydrogen production rate [Nm ³ /h]				113,000
Life time [h]	10,000	10,000	140,000	140,000

On the basis of a broad experimental program (EVA-I and EVA-II facilities, additional testing of kinetics, heat transfer, materials) the following statements can be made:

- Two helium heated steam reformer bundles with about 6 MW power, consisting of 30 (18) tubes have been tested successfully. The dimensions and design data of the reformer tubes and the catalyst were the same as for the nuclear application. The maximum temperature of the helium to heat the SR tubes was 950°C at 4 MPa. Reforming temperature was 800°C and reforming pressure was 4 MPa. The operation time was more than 6000 h.
- The catalytic steam reforming reaction allows reaction rates of $10^3 \text{ Nm}^3 \text{ CH}_4 / (\text{m}^3_{\text{catalyst}} \text{ h})$ (related to the catalyst volume in the reformer tubes) at 800°C, 4 MPa and $\text{H}_2\text{O}/\text{CH}_4 = 4$.
- The heat transfer coefficient on the process side was observed to be rather high. More than 1000 W/(m² K) were realized. The heat transfer coefficient on the helium side was around 500 W/(m² K), i.e., also relatively high. It is, however, limited by the allowable pressure drop of around 40 kPa.
- The overall heat transport coefficient in connection with the chosen temperature distribution on the helium side and on the process side allows an average heat flux of 60 kW/m² for the reformer tubes.
- The conversion of methane at the reaction conditions mentioned above is in the order of 65 %. It corresponds nearly to the thermodynamic equilibrium.
- The components behaved very good in terms of thermal expansion, bending of tubes, friction, fretting and vibrations caused by flow effects.
- Components were also tested at transient conditions with changing rates for the temperature on the helium side of > 10°C/min and for the pressure of > 4 MPa/min. The rates of changes of parameters on the process side were even larger. No difficulties were encountered.
- There was no damage on reformer tubes or guiding tubes, internal recuperators or inner return pipes. The gas ducting structures, insulations and supporting structures were operated without difficulties, too. Measurement and control of process parameters during operation was easy.
- The operation with the steam generator downstream of the steam reformer never caused any difficulties.
- The efficiency of the catalyst practically was not changed during some 1000 h of operation. There was no carbon deposition on the catalyst because of the chosen $\text{H}_2\text{O}/\text{CH}_4$ ratio of 4 in the process. The handling of different catalysts was tested with procedures which can be applied to a larger component.

In summary, it can be stated that the helium-heated steam reforming process is well understood and tested on a large scale. This should allow the extrapolation by a factor of around 10 from the EVA-II plant to a helium heated reformer connected to a modular HTGR of 170 to 200 MWt. For larger powers, more reformer loops should be coupled to the reactor.

3.3.3. The Japanese Approach of Nuclear Steam Reforming

3.3.3.1. Concept of SMR for the HTTR

Unlike Germany, Japan has selected the option of coupling an HTGR to SMR by the employment of an IHX for the steam reformer process. This approach has been applied in the Japanese HTTR project. A flow diagram of the hydrogen production system based on steam-methane reforming and its potential coupling to the HTTR is shown in Fig. 3-22. The total system is subdivided by the dash-dotted line into the existing nuclear part on the left-hand side and the – presently not existing – chemical part on the right-hand side.

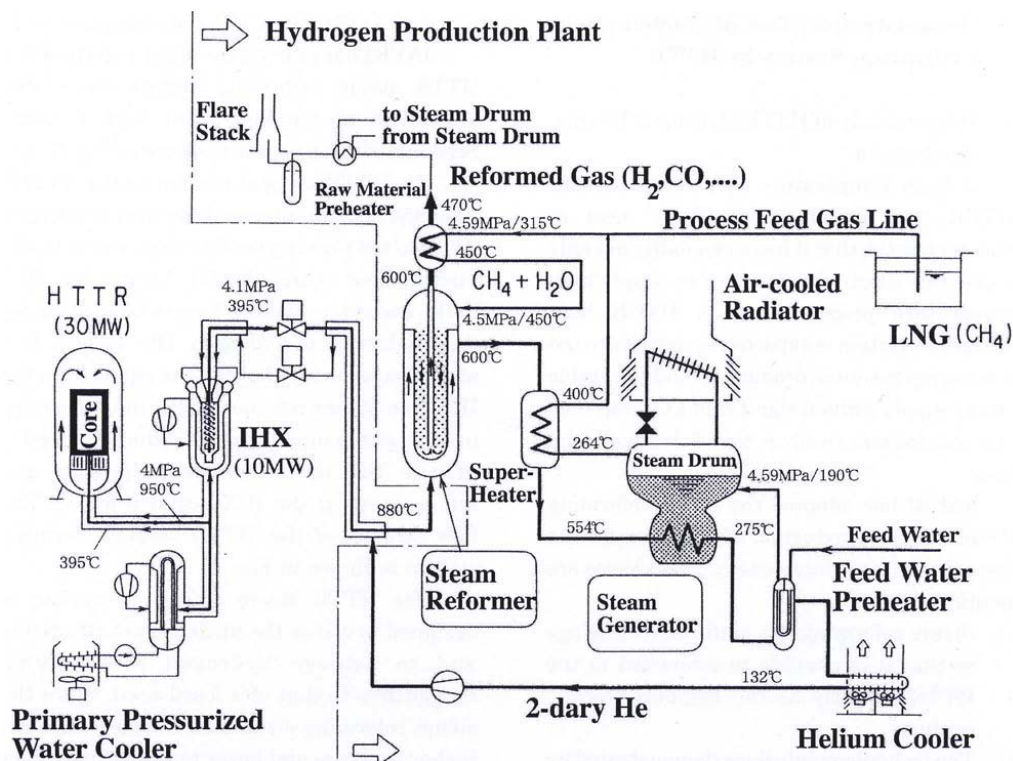


Fig. 3-22: HTTR coupled to a hydrogen production plant based on SMR

The requirements for a system with safe operation and high hydrogen production efficiency has initiated engineering design work on key components for the nuclear steam reforming process:

- A new concept steam reformer heated by helium gas from the nuclear reactor has been designed to achieve high hydrogen production performance and competitiveness to an economical, fossil-fired hydrogen production plant.
- A natural convection type of steam generator has been selected to achieve sufficient system controllability accommodating a large difference in thermal dynamics between the nuclear reactor and the steam reformer.
- An air-cooled radiator is connected to the steam generator to operate as a final heat sink during normal and anticipated operational occurrence condition.

The separation of primary circuit and chemical process avoids contamination questions in the steam reformer totally and reduces the permeation rates of hydrogen and tritium to negligible values. However, the heat fluxes in the steam reformer have values of around 40 kW/m², if the same conditions in the reforming process shall be fulfilled. The fabrication of the steam reformer and steam generator requires different standards compared to components which are directly integrated into the primary helium circuit.

With the construction and operation of the 30 MWt High Temperature Engineering Test Reactor, HTTR, (first criticality in 1999), the Japan Atomic Energy Agency (JAEA), formerly JAERI, has laid the basis for utilization of nuclear process heat for hydrogen production. The reactor allows a coolant outlet temperature of 950°C to provide process heat at 905°C outside the reactor vessel, which was demonstrated in 2004 for the first time in the world. Over several years, steam reforming of methane was considered top candidate process to be connected to the HTTR for the world's first nuclear hydrogen production. The HTTR nuclear steam reforming system will therefore be taken as an example and described in more detail.

The HTTR steam reforming system has been designed to provide about 4200 Nm³/h of hydrogen production using a Ni-based catalyst with 10 MW of thermal energy. A heat utilization ratio (defined as the ratio of output hydrogen energy to total input thermal energy) of 73 % is expected. This value is competitive to the conventional system, where the heat utilization ratio is about 80 %.

3.2.3.1. Design of Steam Reformer

The HTTR can provide high-temperature helium gas of 905 °C at the outlet of the IHX and, due to further heat loss from hot gas duct between IHX and SR, secondary helium of 880 °C at the inlet of the steam reformer. The steam reformer component is shown in Fig. 3-23.

Helium flows into the steam reformer at the bottom and then upwards outside the catalyst tubes, squeezed by multiple plates of orifice baffles transferring heat by forced convection flow (in contrast to heat radiation in the conventional design). The catalyst tubes contain packings of Ni/Al₂O₃ reforming catalysts, through which the process feed gases (natural gas, steam) are routed. The catalyst tube wall thickness is 13 mm meeting the requirements on design limits for pressure retaining components. Finally the helium, which is cooled down to 585°C, exits and flows to a superheater. The main design specifications of the SR are listed in Table 3-8.

The process feed gas mixture of natural gas and steam, after being preheated to 450°C at a pressure of 4.5 MPa enters the steam reformer at the top and then flows downwards in an annular flow between the walls of outer and inner tube through the catalyst bed, where the methane and other lighter hydrocarbons together with steam are reformed. The reformed gas having reached a maximum temperature of 830°C, flows then upwards inside the inner tubes transferring at the same time heat to the feed gas and eventually leaving the steam reformer at a temperature of 580°C and a pressure of 4.1 MPa.

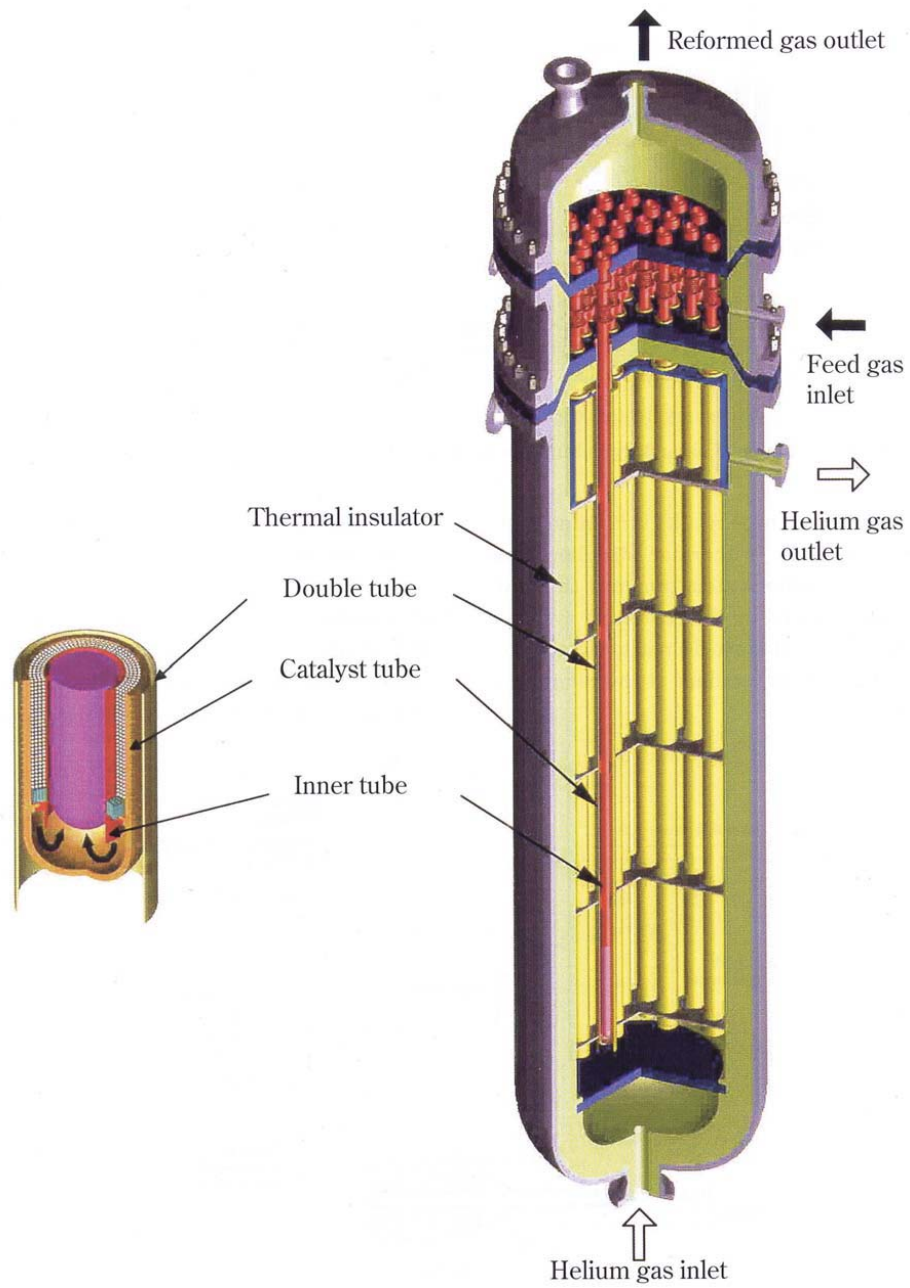


Fig. 3-23: Steam reformer component for connection to HTTR (source: JAEA)

Table 3-8: Design specifications of the steam reformer for HTTR and mock-up test facility

Parameter	Steam reformer in HTTR	Splitting tube in mock-up test facility
Nuclear heat input	3.6 MW (plus 1.3 MW from product gases)	0.42
Material Tubes Catalyst	Hastelloy XR Ni/Al ₂ O ₃	Alloy 800 H
Size Shell diameter [m] Shell height [m]	1.19 14	
Catalyst tube Outer diameter [mm] Wall thickness [mm] Length [m] Inner tube diameter [mm] Wall thickness [mm] Length [m] Number of tubes	Bayonet type, concentric double-walled tube 153.8 13 7.9 60.5 3.9 not decided yet 37	Bayonet type, concentric double-walled tube 3
Secondary helium Inlet temperature [°C] Outlet temperature [°C] Pressure [MPa] Flow rate [kg/s]	880 585 4.1 2.5	880 650 4.1 0.091
Heat transfer rate at outer surface [W/(m ² K)]	1700	
Process feed gas Inlet temperature [°C] Outlet temperature [°C] Pressure [MPa] Raw gas flow rate [kg/s] Raw gas conversion [%]	450 580 (max.: 800) 4.5 0.39 64.2	450 600 4.5 0.012
Steam-methane ratio	3.5	2.5 - 4 (3.5)
Hydrogen production rate [Nm ³ /h]	4240	120

In the HTTR steam reforming system, a steam/methane ratio of 3.5 has been selected. The required steam is about 5160 kg/h at rated conditions so that the thermal energy necessary to generate steam is 3.1 MW. The thermal energy of the product gas at outlet of the steam reformer is only 1.9 MW. Therefore, a steam generator is necessary on the secondary helium loop in order to supply this large amount of thermal energy. The superheater and the steam generator are installed downstream of the steam reformer to generate feed steam for the steam reformer. The required helium temperature at the inlet of the IHX is 160°C, requiring the addition of a feed water preheater and a helium cooler. In the future HTGR heat application system, the outlet helium gas of steam generator will be returned to the IHX directly.

The flow rate of natural gas as a feed gas is 1290 kg/h and the flow rate of steam is 5160 kg/h at the inlet of the steam reformer. The temperature of the product gas is about 600 °C. This gas is cooled down by the water cooler and separated into steam and dry gas compositions including hydrogen, carbon oxide, carbon dioxide and residual methane in the separator. The pressure and maximum temperature of the process feed gas are 4.5 MPa and 830°C so that the conversion ratio from methane to hydrogen is expected to be 68 %. As a result, 32 % of methane will remain in the product gas. In the conceptual design of the HTTR/SR, this residual methane is burned in the flare stack together with the other combustible gases.

Furthermore the reforming process requires a thermal heat input of 4.8 MW. In order to generate feed steam by the thermal energy of secondary helium gas, the helium gas temperature at the outlet of the steam reformer is required to be about 600°C, so that only thermal energy of 3.6 MW is supplied to the steam reformer from helium gas. This high-pressure and low-temperature condition is a disadvantage for the steam reforming reactions.

A new heat exchanger type concept of steam reformer is required to enhance the hydrogen production rate. It should allow

- an increased heat input into the process gas by employment of orifice baffles;
- an increased reaction temperature of the process gas at the outlet of the catalyst zone;
- an optimizing reforming gas composition to enhance the reforming rate.

The aim of reaching a heat flux density closer to that of the conventional method (Table 3-9) can be achieved by employing a helium-heated counter flow heat exchanger. Helium under pressure shows excellent heat transfer properties.

Assuming an infinitely long catalyst tube, the process gas temperature approaches that of the helium gas. But in general, a catalyst tube length limit of approximately 10 m is mandated from the viewpoint of seismic design. It is necessary to enhance the heat transfer rate in order to design for an adequate steam reformer size. There are several means for enhancement of the heat transfer such as baffles, double tubes, fins, and others. JAEA has performed an analytical comparison of the heat transfer rate and selected a double tube with a radially finned catalyst tube, for which the thermal radiation rate is more than 1800 W/(m² K).

Table 3-9: Comparison between nuclear and conventional design

Parameter	Steam reformer		
	Fossil-heated	Helium-heated for HTTR/SR	Helium-heated improved design
Process gas pressure [MPa]	1 - 3	> 4	4.5
Maximum temperature of process gas [°C]	850 - 950	≤ 750	800
Maximum heat flux to catalyst zone [kW/m ²]	50 - 80	10 - 20	40
Heat transfer	radiation	forced convection	
Efficiency [%]	80 - 85	50	78
CO ₂ emission [t/h] (Basis: 10 MW)	3	0	

The heat demand is drastically increasing when the endothermal chemical reactions start the reforming process. On the nuclear side, the helium temperature is “conventionally” increasing linearly with power output. Because of this mismatch between heat demand and heat supply during startup, an additional heat exchanger component, a so-called heat load controller, with a capacity of 2.8 MW is integrated into the steam reforming system. The controlling is achieved by adjusting the helium flow rate. Fig. 3-24 shows the relationship between nuclear power and helium temperatures during the startup phase [Hada 1994].

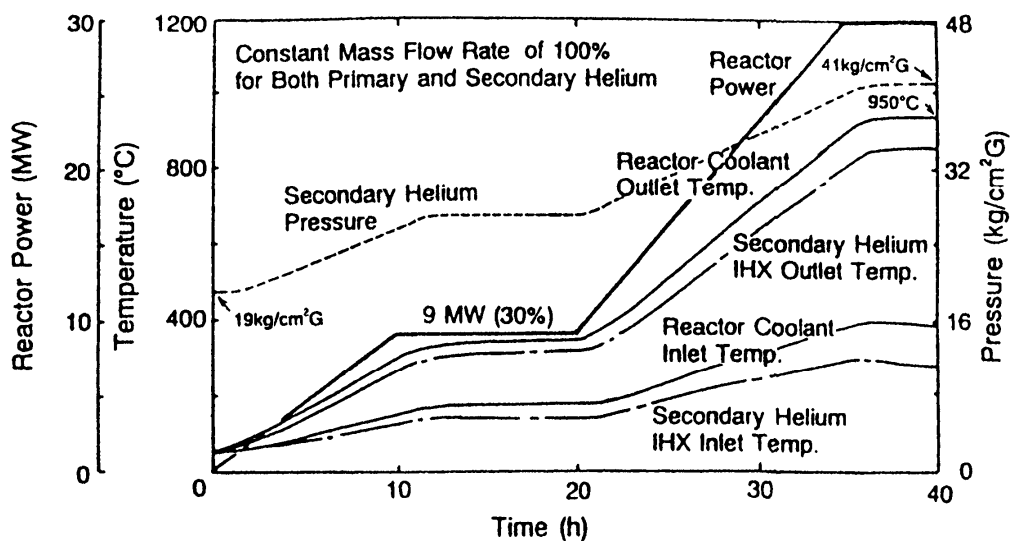


Fig. 3-24: Nuclear power and helium temperature during the startup phase

The proposed new design of a steam reformer is shown in Fig. 3-25. JAEA has adopted a bayonet type of catalyst tube, a concentric, double-walled tube which can use both the outside and inside gas flow for heating up the process gas. The thermal energy input into the process gas increases from 3.6 to 4.9 MW. These improvements are applicable not only to HTGR steam reforming system but also to other HTGR hydrogen production systems. This is because a heat exchanger type of endothermal chemical reactor is an essential technology for the production of hydrogen through the use of nuclear heat.

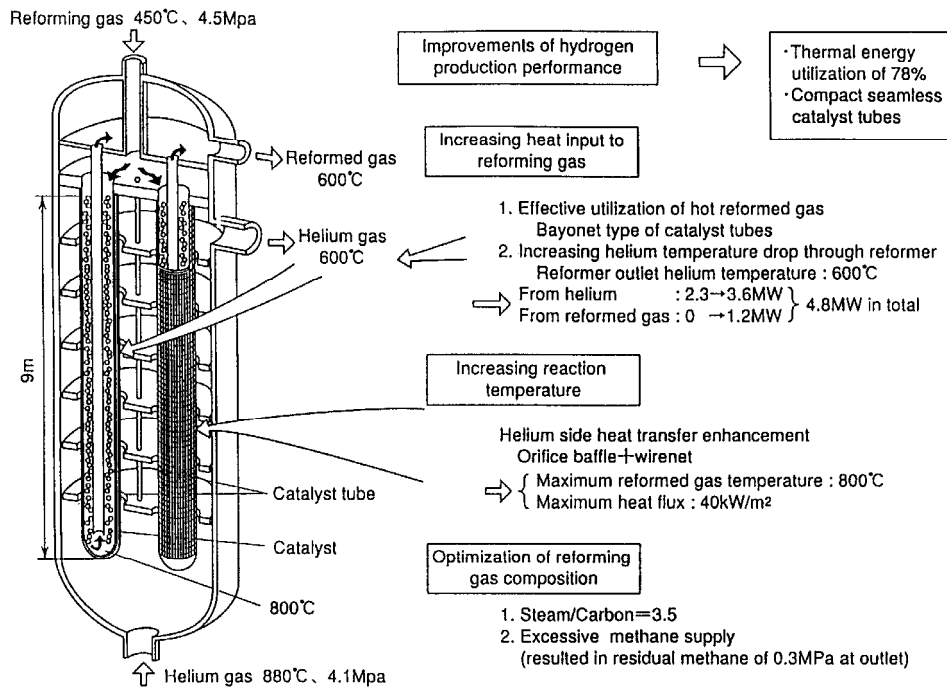


Fig. 3-25: New concept of He-heated steam reformer

The effective consumption of the nuclear process heat input of 10 MW can be seen from the following Table 3-10 [Hada 1994]:

Table 3-10: Consumption of 10 MW nuclear heat input in the SMR system of the HTTR

Heat loss	0.5 MW
Air cooler	0.7 MW
Feed water preheater	0.8 MW
Reboiler	2.7 MW
LNG preheater	0.2 MW
Superheater	1.0 MW
Process gas heater	0.5 MW
Steam reformer	3.8 MW
Σ	10.2 MW

Partial pressure conditions of the process gas components for the catalyst zone inside the reformer tubes are given in Table 3-11 [Hada 1994].

Table 3-11: Partial pressure distribution for product gases in the catalyst zone

Gas component	Partial pressure [MPa] in catalyst zone	
	Inlet	Outlet
CH ₄	0.82	0.28
H ₂ O	3.42	1.95
H ₂	0.13	1.72
CO	0.03	0.27

3.2.3.2. Experiments in Mock-Up Test Facility

The steam reforming process was tested in out-of-pile experiments under simulated nuclear conditions at JAEA, Oarai, [Miyamoto 1998]. For this purpose, a 1:30 downscaled mock-up facility was constructed. Schematics of the test plant and of the splitting tube used are given in Figs. 3-26 and 3-27. The test facility was basically used to investigate the process control technology. The test program planned to run over four years has been starting in 2002. It comprises normal startup/shutdown tests to investigate temperature and pressure fluctuations and its controllability as a function of the steam-methane ratio, in order to optimize feed flow of methane and steam according to temperature and pressure of the helium gas. In a system controllability test, potential thermodynamic disturbances at the pressure boundary between helium and methane are examined by the stepwise change of the methane and steam flow rates, in order to optimize the control system for the pressure difference (see also chapter 4.1.3.).

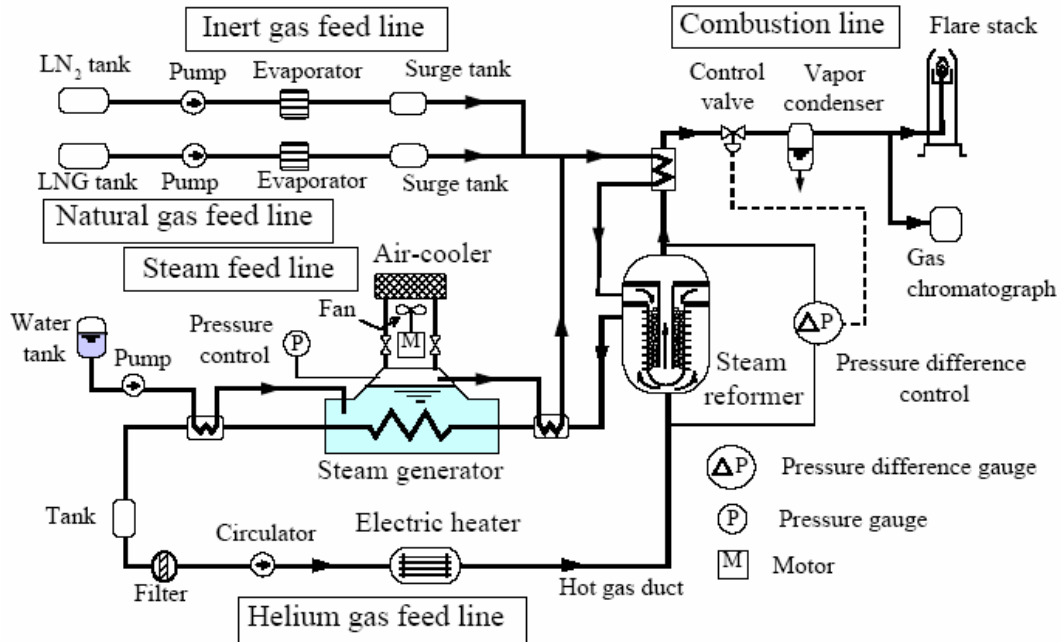


Fig. 3-26: Schematic of steam reforming mock-up model test facility at JAEA [Ohashi 2004]

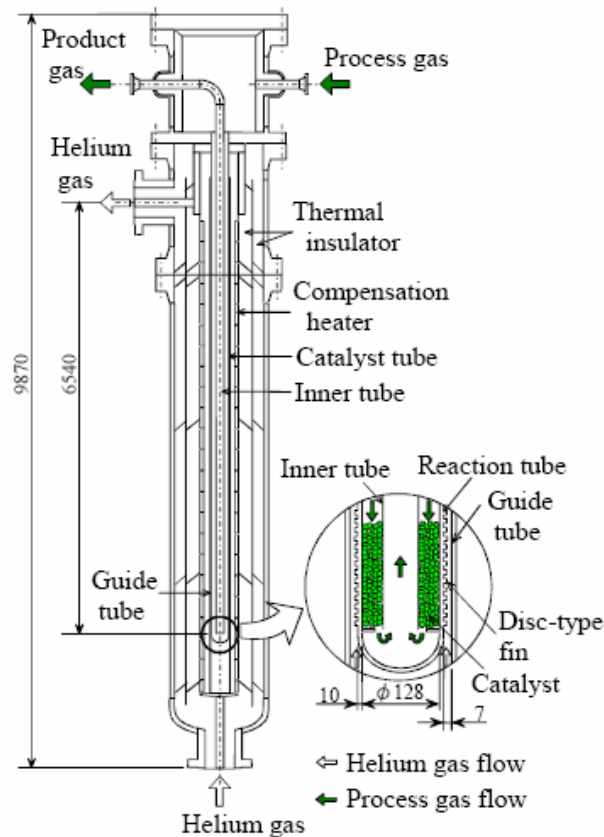


Fig. 3-27: Schematic of steam reformer tube in mock-up facility [Ohashi 2004]

Safety-related tests were conducted with the mock-up facility to examine malfunctions and accidental sequences in the process gas line including emergency shutdown. Focus was on the examination of potential thermal turbulences from the hydrogen production system to the nuclear side. The goal is that in case of an accident in the process line, the HTTR should be shutdown by the normal operation procedure rather than by a reactor scram. In such a case, the heat of the helium is to be removed via the steam generator which limits the temperature fluctuations in the helium. Characteristic data of the splitting tube in the mock-up facility are also listed in Table 3-8 in comparison with the steam reformer design to be used in connection with the HTTR. Results of these safety-related tests are described in more detail in chapter 4.1.3.

Experimental results obtained with respect to the temperature fluctuations [Hayashi 2005]:

Startup:	$\Delta T = 2.5$	@ SG outlet
	$\Delta T = 110$	@ SR outlet
Shutdown:	$\Delta T = 0.5$	@ SG outlet
	$\Delta T = 88$	@ SR outlet

demonstrating that the steam generator is capable of mitigating thermal disturbances arising from the chemical reactor in form of temperature fluctuations of the helium or pressure fluctuations of the process gas. The defined target of $\pm 10^\circ\text{C}$ for the steam generator was met.

3.3.3.2. Nuclear Steam Reforming at Lower Temperatures

Another, however “unconventional” idea of applying nuclear to steam reforming is the use of the heat of spent LWR fuel to run the endothermal process at a (lower) temperature level of 600°C. After separation of the uranium, the remaining spent fuel is packed in rods (100/80 mm outer/inner diameter) and stored in vessels (5.8 m diameter, 9 m height). Heat transferring fluids could be helium or molten salts. The decreasing decay heat production with time is compensated by reducing the coolant flow rates to maintain the temperature level for SMR. The amount of 5000 t of spent fuel corresponding to about 15 MWt was estimated to yield $\sim 13,000 \text{ Nm}^3/\text{h}$ of H_2 [Karasawa 2005].

3.4. Thermochemical (Hybrid) Cycles

Besides alkaline electrolysis, two major potential challengers have been identified to produce large amounts of hydrogen by high temperature water-splitting and are nowadays under investigation worldwide: high temperature electrolysis and thermochemical cycles. We will first describe these two types of process and then, as many options seem attractive at first glance we will give go/no go criteria to focus on the best options.

3.4.1. Overview of Thermochemical Cycles

A thermochemical cycle is a process consisting of a series of thermally driven chemical reactions where water is decomposed into hydrogen and oxygen at moderate temperatures. All supporting intermediate chemical substances are regenerated and recycled, and remain – ideally – completely in the system. The only input is water and high temperature heat. Therefore these cycles have the potential of a better efficiency than alkaline electrolysis and hence have the potential to significantly reduce the production costs from water. The cycle is called a hybrid cycle, when the low temperature reaction while thermodynamically unfavourable must be forced electrochemically. Thermochemical cycles are being investigated mainly with respect to primary heat input from solar or nuclear power.

Numerous thermochemical cycles have been proposed in the past and checked against features such as reaction kinetics, thermodynamics, separation of substances, material stability, processing scheme, and cost analysis. Some of the most promising cycles have been further analyzed in detail. However, all cycles have design challenges and all are still at laboratory scale including those based on the sulfur family which all have in common the thermal decomposition of sulfuric acid at high temperatures (see Fig. 3-28).

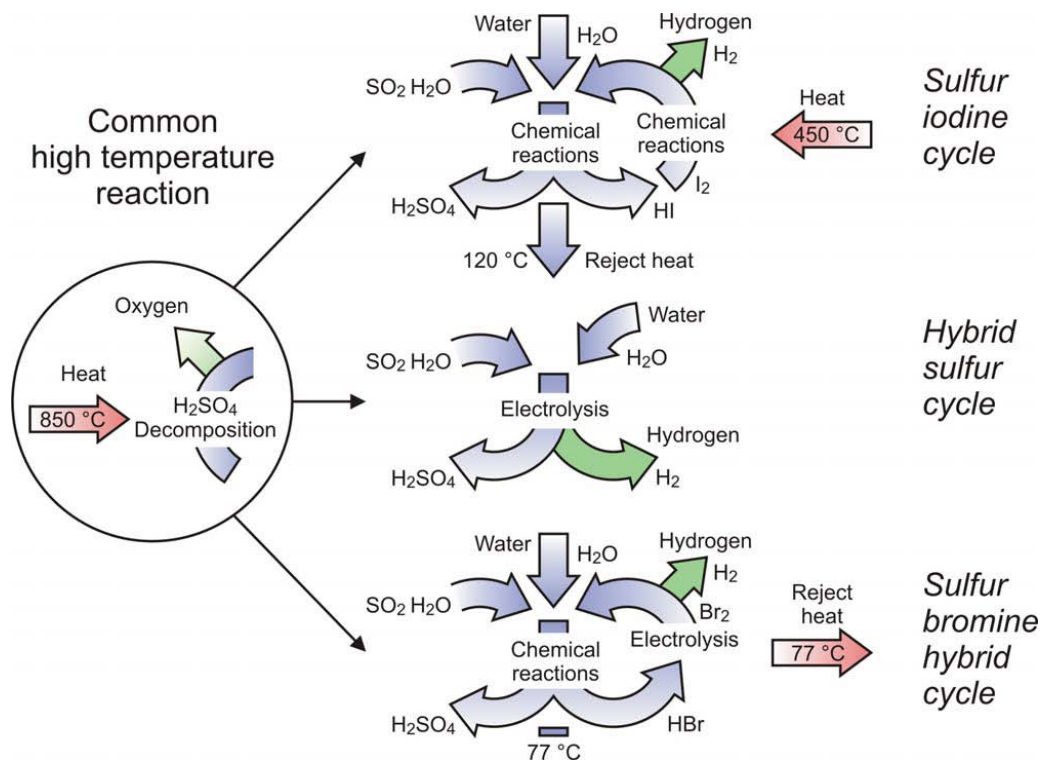


Fig. 3-28: Thermochemical cycles of the sulfur family

After the principle of a thermochemical cycle was discovered by Funk and co-workers in the mid 1960s, various process suggestions have been made. The first major program [Beghi 1986] directed by the Joint Research Centre (JRC) of the European Community in Ispra, Italy, began in the late 1960's and ended in 1983. The goal of this work was to identify potential thermochemical cycles to be coupled with a high temperature gas cooled reactor. In a first step, mercury, manganese, and vanadium based cycles were studied. Later nine cycles based on iron and chlorine were investigated. These cycles were abandoned because of the difficulty to decompose the iron chloride. Finally several cycles of the sulfur family were studied which eventually resulted in a laboratory demonstration loop of the sulfur bromine process. Associated with this experimental effort, the JRC Ispra had an important activity in the field of corrosion, the design of large-scale equipments, and the development of industrial flow sheets.

In the United States, particularly the sulfur-iodine (S-I) cycle was extensively studied with respect to individual reactions, corrosion, bench scales of the three parts of the cycle and full scale flow sheet [Besenbruch 1982]. Later the Gas Research Institute (GRI) funded a long-term systematic program to evaluate some 200 distinct thermochemical cycles, out of which 125 cycles were selected on the basis of thermodynamic criteria. 80 of the most promising were tested in a laboratory, 15 were found feasible using batch techniques with chemical reagents, and eight were demonstrated successfully with recycled materials. Within the Nuclear Energy Research Initiative, NERI, a 3 years program was conducted to evaluate the feasibility of thermochemical cycles for efficient, cost-effective, large-scale nuclear H_2 production. After various screening processes on a total of 115 cycles, the number of cycles considered worth of being investigated in more detail boiled down to two: S-I and UT-3. Further efforts concentrated on the S-I process due to the fact that UT-3 was treated in Japan [Besenbruch 2000].

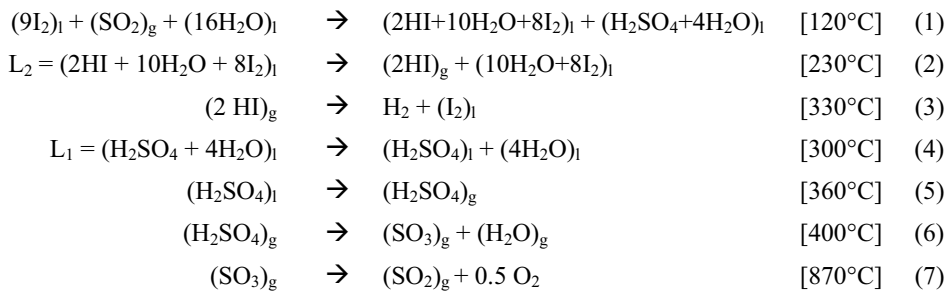
Russia made some major research on the thermochemical cycles too and finally constructed and operated a small demonstration loop of the hybrid sulfur (HyS) process.

Japan has focused its efforts on two cycles: the UT-3 cycle (iron, calcium and bromine) and the S-I cycle. In both cases, the reactions were studied separately and demonstration loops were successfully operated. Japan is the only country which has maintained a constant activity in this field until now.

The three most well known and investigated processes are the S-I cycle, the HyS (also called Westinghouse cycle), and the UT-3 cycle. They are described below in more detail.

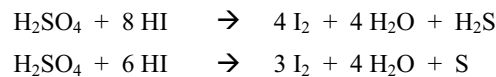
3.4.2. The Sulfur-Iodine Cycle

One cycle considered with a high priority is the sulfur-iodine (S-I) process originally developed by the US company General Atomics and later taken up and modified by different researcher groups like the Japanese JAEA. The S-I process is an all-fluid cycle and can be split into the following reactions [Vitar 2005]:



The temperatures between brackets are approximate values and depend upon the pressure which is not necessarily uniform in the different parts of the cycle.

The first reaction is the Bunsen reaction in which at the presence of the substances SO_2 and I_2 water is added. The products of this exothermal reaction are two immiscible aqueous acid phases, hydrogen iodide and sulfuric acid, given as a lighter H_2SO_4 -rich (L_1) and a heavier HI-rich (L_2) aqueous phase (see Fig. 3-29). The latter is a mixture of hydrogen iodide, iodine and water named HIX. In practice, as can be seen from reaction (1), a significant excess of both water and iodine is necessary to observe the separation of the acids. It is very important to find the optimal composition ratio of chemicals for the Bunsen reaction. A large excess is typically connected with side reactions which lead here to the formation of S and H_2S according to:



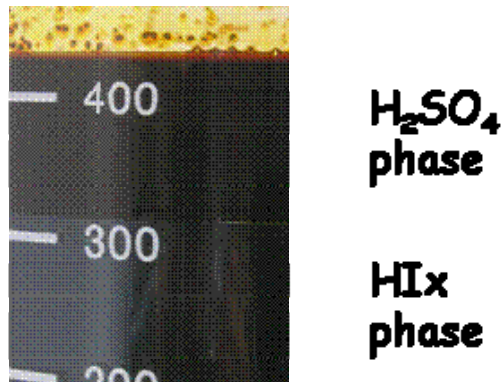


Fig. 3-29: Two immiscible aqueous acid phases resulting from the Bunsen reaction [Bae 2005]

After separation, purification and further concentration, the acids are decomposed: Reaction (2) is the endothermal decomposition of hydrogen iodine resulting in the production of hydrogen. This separation is the most critical phase and difficult because of the partial decomposition into H_2 and I_2 and the presence of an azeotrope in the ternary mixture. In reactions (5)-(7), sulfuric acid is decomposed in two stages: it first decomposes spontaneously at 300-500°C into sulfur trioxide and water (reaction (6)) with the former to further decompose at high temperatures and in the presence of a catalyst into oxygen and sulfur dioxide (reaction (7)). Sulfur dioxide, water and iodine are then recycled in L_1 . Fig. 3-30 shows the equilibrium conversion of the two acids as a function of temperature showing the need for high excess I due to the low HI conversion ratio of about 20 %.

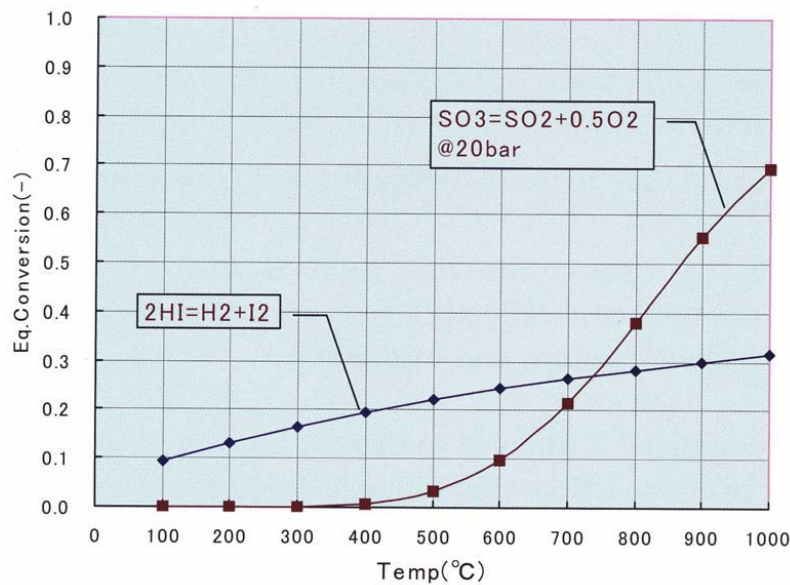


Fig. 3-30: S-I process equilibrium conversion [Tsukada 2005]

For the difficult process of HI separation, three methods are currently being investigated (see Fig. 3-31):

- extractive distillation, pursued by the US, where a third body – phosphoric acid – is added to first separate the iodine, before the HI is distilled and then decomposed with the H_2 being separated from the gas mixture by using membranes;
- electrodialysis and electro-electrodialysis, pursued by Japan, Korea, where water is removed to concentrate the HIx mixture, before excess HI can be removed by simple distillation using ion exchange membranes under the driving force of electric potential difference;
- reactive distillation, pursued by France, where HIx distillation and HI decomposition is taking place in the same reactor leading to a liquid-gas column with I_2 dissolved in the lower, liquid phase and H_2 plus water to be gained from the upper, gaseous phase.

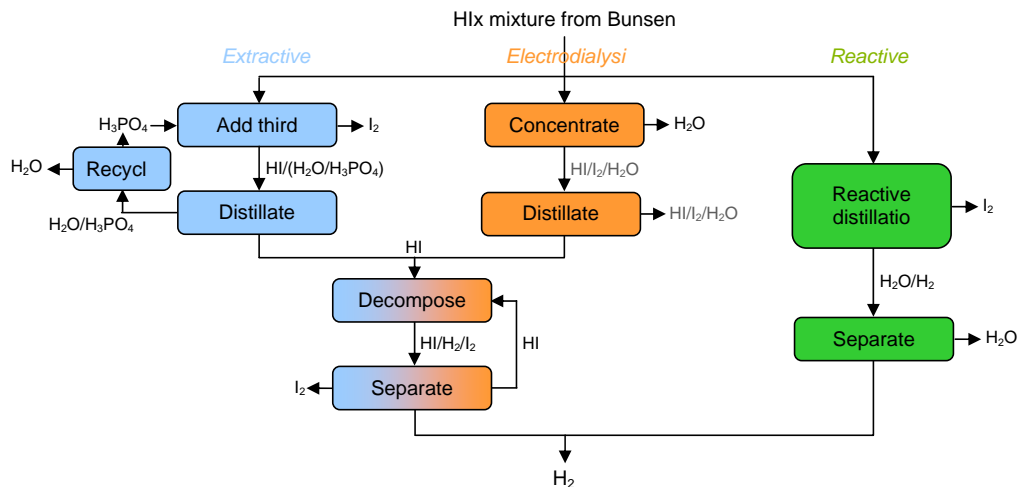


Fig. 3-31: Options of HIx decomposition (source: CEA)

The S-I cycle is among all thermochemical cycles the one with the highest efficiency quoted. Cycle efficiency vs. process temperature is shown in Fig. 3-32 [Schultz 2005]. The theoretical limit of efficiency for the total process is assessed to be 51 % assuming ideal reversible chemical reactions. Analytical studies anticipate efficiencies of 40-50 %. A best estimate was found to be around 33-36 % [Goldstein 2005].

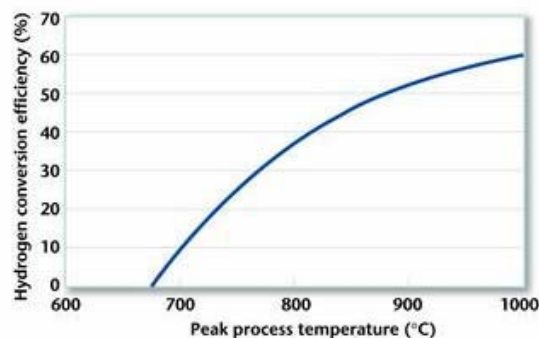


Fig. 3-32: S-I cycle efficiency [Schultz 2005]

A severe problem to solve is the extreme corrosiveness of the materials in contact with the acids at high temperatures and pressures. Respective experimental investigation is currently conducted in various institutions. The candidate materials for the sulfuric acid decomposition are, for example, Alloy 800 and Hastelloy. Other materials considered are SiSiC, SiC and SX (Fe-Cr-Si). For the hydrogen iodide decomposition, Hastelloy is foreseen and for the Bunsen reaction, Ni alloys, ceramics, Zr, Ta, glass lining are favorites.

The above S-I process could be successfully demonstrated by JAEA in a closed cycle in continuous operation over one week. The facility consisted of more than 10 process units primarily made of glass and quartz with a hydrogen production rate achieved of 30 NI/h at an efficiency of 6.4 %. The next step starting in 2005 is the design and construction of a pilot plant with a production rate of 30 Nm³/h of H₂. The pilot plant will be operated under the simulated conditions of a nuclear reactor using an existing He gas circuit to provide helium of 880°C at a system pressure of 3 MPa [Kubo 2004]². Currently tests are conducted with a H₂SO₄ purifier to remove the impurities HI, I₂, H₂O. In leak tests, a helium leakage rate of 1.5*10⁻⁸ (Pa m³)/s at the connection of the SiC blocks was measured [Kubo 2005].

For later connection of this process to the HTTR (see Fig. 3-33), a thermal power of 8.5 MWt will be taken from the IHX to allow for an H₂ production rate of 1000 Nm³/h with an expected efficiency of 43.6 % [Sakaba 2005b].

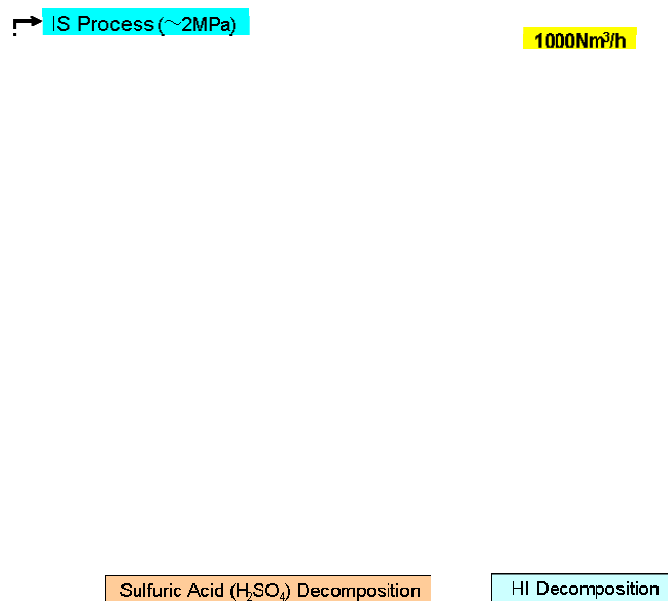


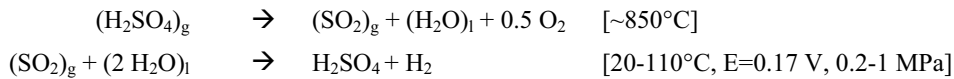
Fig. 3-33: Demonstration plant of nuclear hydrogen production with S-I cycle connected to HTTR
[Hino 2005]

A laboratory-scale S-I cycle integrated test loop at prototypical pressure and temperature conditions for a H₂ production rate of ~ 1000 l/h is now under construction by GA in collaboration with Sandia National Laboratories, and CEA-Saclay [Schultz 2005] and scheduled to be completed by 2007. Korea has started the construction of a respective test facility for a H₂ production rate of 40-100 Nm³/h.

² The designated process pressure has been recently changed from 3 to 2 MPa [Hino, JAEA, personal communication].

3.4.3. The Westinghouse (Hybrid S) Cycle

The hybrid-sulfur (HyS) process developed by Westinghouse in 1973-83 is a two-step, sulfuric acid hybrid cycle [Brecher 1977]. A mixture of SO₂ and water is reacted in an electrolytical cell at lower temperatures to produce H₂ and a sulfuric acid aqueous phase. The sulfuric acid is vaporized to produce SO₃ which is then reduced in a high temperature step to SO₂ and O₂. The electrolytic step offers the advantage of having a theoretical cell potential for SO₂ anode depolarized electrolysis of only 0.17 V per cell. In practice, SO₂ electrolyzers may require no more than 25 % of the electricity needed in the alkaline water electrolysis, however, at the expense of the need to decompose H₂SO₄ at high temperatures in order to recycle the SO₂. Still the net thermal energy requirement is significantly less than for conventional water electrolysis. The HyS process is a variation of the S-I process where the reactions (2) and (3) are replaced by the electrolysis of sulfur dioxide:



The Westinghouse process (see also Fig. 3-34) is simpler in the design, because the use of corrosive halides is not required and instead uses only one common raw material, sulfur. The raw material flow is quite limited when compared to other thermochemical cycles. Optimum efficiency was observed at a 65 % concentration of sulfuric acid in water. The main advantages of this cycle are the fact that there are only two reactions, and the ability to have the hydrogen production separated from the nuclear plant.

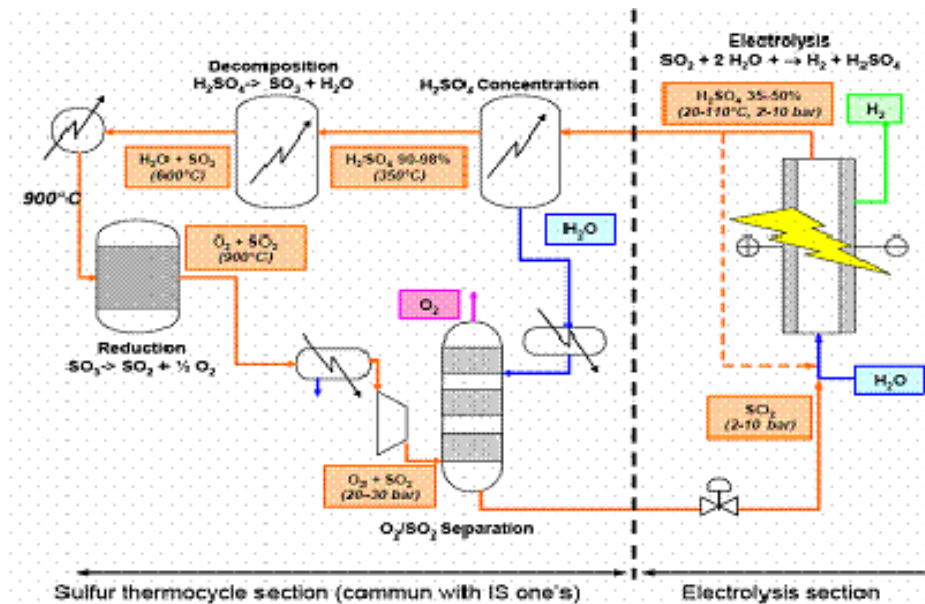


Fig. 3-34: Schematic of Westinghouse hybrid-sulfur cycle (source: CEA)

A major task is the optimization of the cell design, materials selections and operating conditions to improve performance and overall durability. The main drawback is in the use of the electrolytic step. The most important issues for increasing the competitiveness of this cycle are to reduce the cell voltage (currently about 0.6 V against a theoretical demand of 0.17 V) and the cost of the SO_2 electrolyzer. Other technical issues to deal with are the sulfuric acid decomposition step and the SO_2/O_2 separation system. Special caution has to be taken to avoid sulfur deposition on the anode or H_2S formation in the electrolytic step which requires the use of SO_2 perm-selective membranes.

Westinghouse demonstrated a 120 NI/h production on a laboratory scale. FZJ in cooperation with JRC Ispra successfully realized the operation of a three-compartment electrolysis cell at 80°C and 1.5 MPa in a 600 h run. The H_2 production rate was 10 NI/h. FZJ also verified the heat consuming step of sulfuric acid splitting on bench scale under HTGR conditions at 4 MPa and with 950°C heat from an electrical furnace [Weirich 1984]. A schematic of the potential connection to an HTGR is shown in Fig. 3-35.

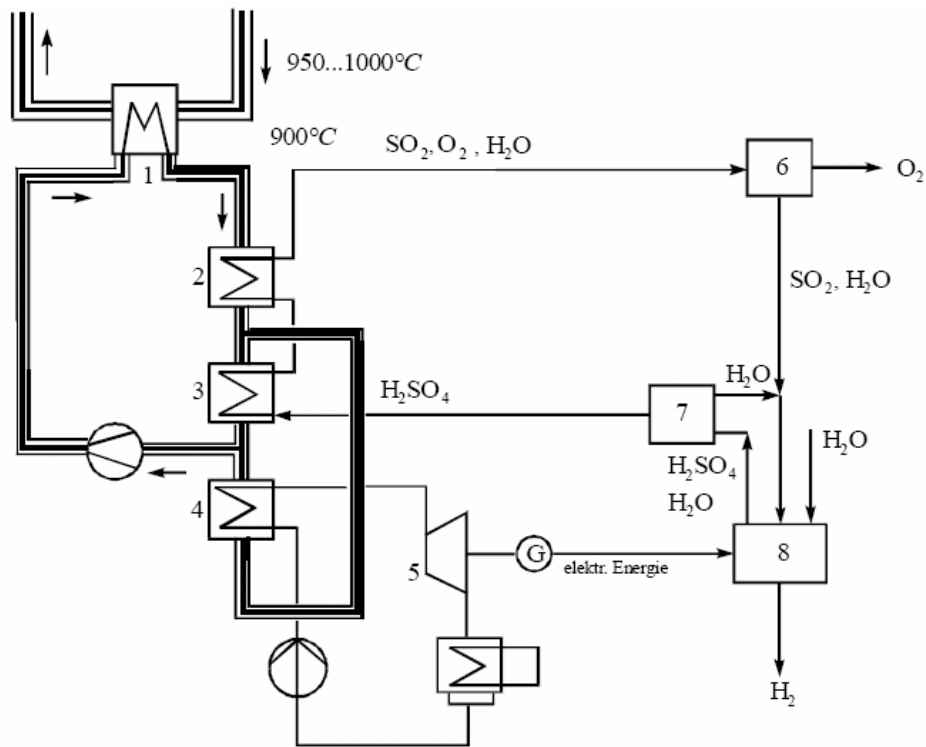


Fig. 3-35: Diagram of combined system of HTGR plus Westinghouse HyS cycle [Kugeler 2005]

Recent work at the SNL resulted in further process improvements employing the process analyzer software AspenPlus. The net efficiency for hydrogen production by the HyS hybrid cycle connected to a helium cooled reactor providing heat input at 900°C has been assessed to be 48.8 % (HHV) [Schultz 2005].

3.4.4. UT-3 Cycle

Another promising thermochemical process is the calcium-iron-bromine or UT-3 cycle which was developed at the University of Tokyo (UT). It consists of four gas-solid reactions in four separate reactor furnaces containing the solid reactants CaBr_2 , CaO , Fe_2O_3 , and FeBr_2 which are present as spherical pellets. Only the heat carrying gases are passed through the reactors which eases material flow control. Maximum heat input required is up to 750°C . Hydrogen and oxygen are removed from the recirculating streams via semi-permeable membranes. A major drawback are the still low reaction rates [Teo 2005].

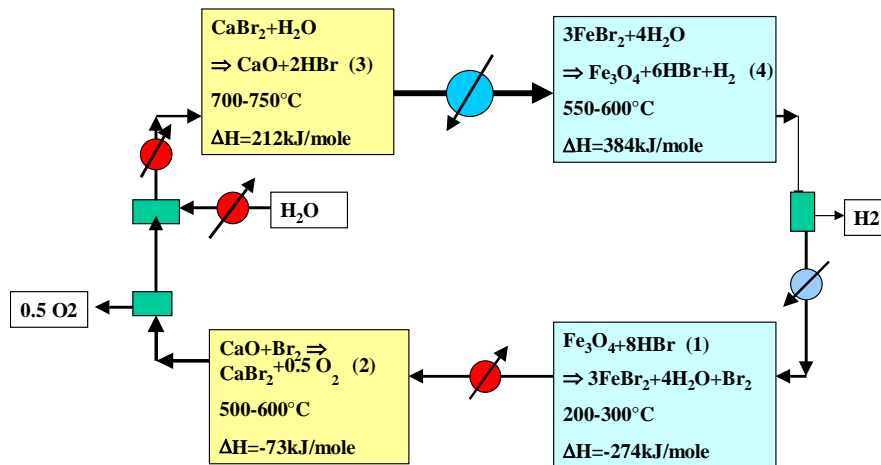
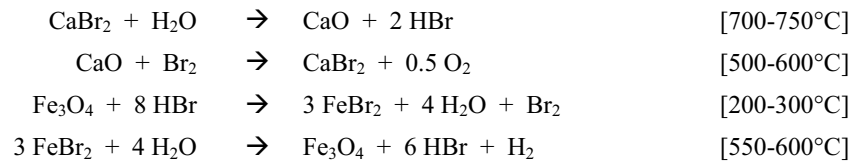


Fig. 3-36: Schematic of UT-3 process

The process with four reactors connected in series in a loop is shown in Fig. 3-36. Stages (1) and (3) produce reactants for stages (2) and (4). After one cycle, the reactors are switched and the direction of the cycle is reversed. This may pose some coupling problems with a permanent HTGR heat source because of the variations of the temperature and heat demand of the four reactors.

The thermodynamics of the above reactions are considered favorable, the overall efficiency, however, is limited to about 40 % due to the melting point of CaBr_2 at 760°C [Schultz 2002].

The physico-chemical approach forecasts some difficulties because of the sintering of the solid reactants [Lemort 2003]. With regard to the second and the last reaction, thermodynamics require the operation at very low pressure and at high temperature what is in contradiction with the volatility of the bromides. One way could be to operate with over-stoichiometric conditions, which

supposes a large amount of gases and will probably conduce to too large reactors because of the too low pressure level required by thermodynamics.

The UT-3 cycle was successfully operated for a few hours in the pilot plant MASKOT (Model Apparatus for the Study of Cyclic Operation in Tokyo).

At the Argonne National Laboratory, a modified UT-3 cycle is being investigated as is shown in Fig. 3-37. In the first step, the CaBr_2 is reacted with steam of 1030 K in a solid-gas reaction to split the water. The dry product HBr is then delivered to the H_2 production taking place in a plasmatron where in an endothermic reaction the HBr is decomposed by electricity into the products H_2 as a gas and Br_2 as a liquid, which are expected to be easily separable. In the exothermic oxygen production step, the initial CaBr_2 is regenerated [Doctor 2003].

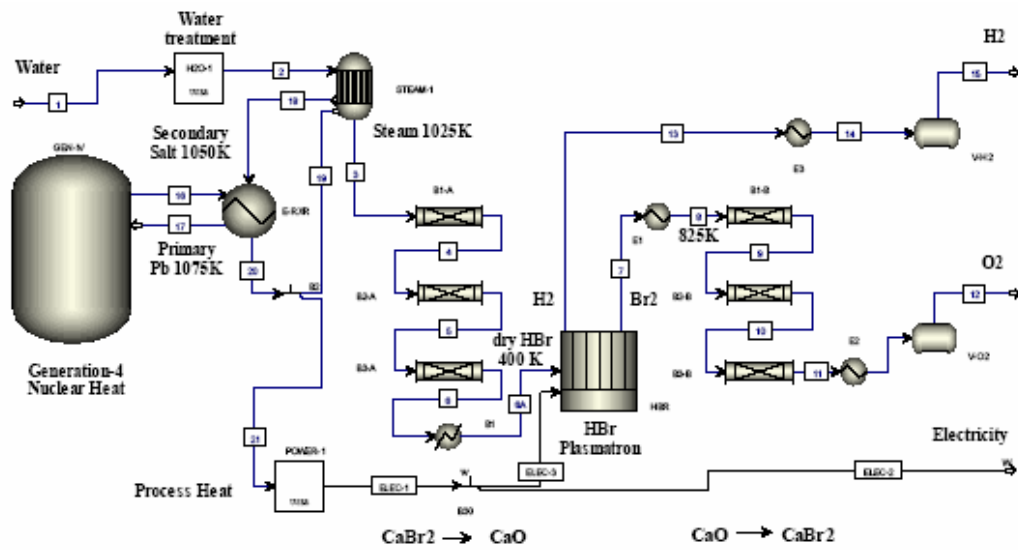


Fig. 3-37: Modified UT-3 cycle as used in STAR-H2 [Doctor 2003]

3.4.5. Other Approaches

Interest has been continued and revived also for other thermochemical cycles as alternatives to the above described. In particular, those operating at lower temperatures, like around 550°C, promise fewer demands on material behavior and lifetime plus they may become feasible in connection with other types of nuclear reactor in addition to the HTGR, such as the Fast Breeder Reactor (FBR), Molten-Salt Reactor, or the Advanced Supercritical Water Reactor (SCWR).

Fig. 3-38 presents a classification of nuclear reactors in terms of power output and coolant temperatures showing that there are two coolant options in the high temperature range: high-pressure gases and low-pressure liquids [Forsberg 2005c].

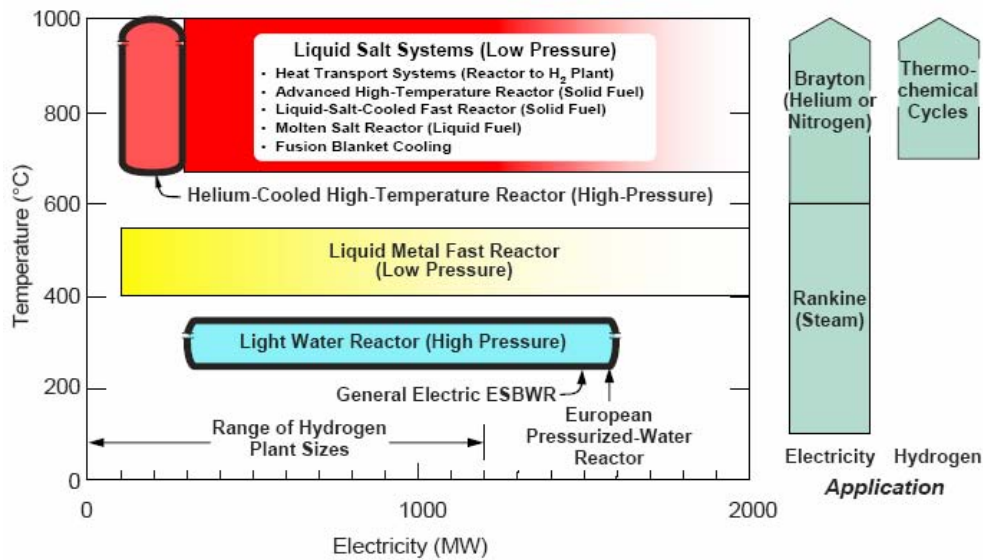
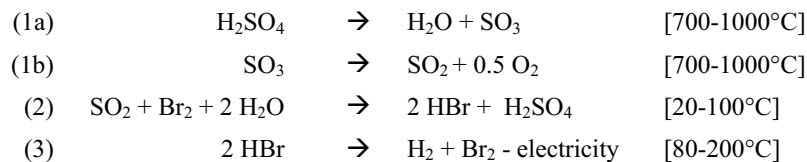


Fig. 3-38: Type of nuclear reactor vs. coolant temperature and power output [Forsberg 2005c]

3.4.5.1. Mark 13 Process

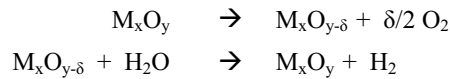
From the three cycles of the sulfur family shown in Fig. 3-28, the one not yet described is the sulfuric acid-bromine hybrid cycle, also called Mark 13. The reactions of this cycle are



with reaction (3) as the electrochemical step. The S-Br hybrid cycle using only liquid or gaseous species, was successfully tested in 1978 in a lab-scale facility at the JRC Ispra, Italy, demonstrating a H₂ production rate of 100 l/h over 150 h with an efficiency of 37 % [Van Velzen 1991]. The system was also operated with a 1 kW solar heat source.

3.4.5.2. Metal/Metal Oxide Processes

In analogy to the steam-iron process of coal decomposition, there are also materials which can act as effective redox pairs in a two-step water splitting process. In the first step, a metal oxide is reduced delivering the oxygen; in the second step, the reduced compound is reacted with water extracting its O₂, while the H₂ is liberated. Two-reaction processes offer the potential for high efficiencies at low cost. Pairs of materials investigated are Fe₃O₄/FeO as the most representative, but also Mn₃O₄/MnO, ZnO/Zn, CoO₃/Co[•]O, Ce₂O₃/CeO₂. The water splitting is done at temperatures < 650°C, whereas the reduction step requires much higher temperatures around 2000°C. Major drawback is the decreasing activity over repeated oxidation/reduction cycles. The general reaction scheme is



Two-step metal-oxide cycles are under investigation at the PSI, Switzerland, in connection with solar-concentrated heat input and at INET, China, in connection with nuclear heat input. Main research efforts are on finding new metal oxides with a comparatively low decomposition temperature. Most promising cycles were found to be the pairs ZnO/Zn and Fe₃O₄/FeO. The INET is examining in detail MFe₂O₄ with M = Cu, Ni, Co, Mg, Zn, Mn having decomposition temperatures in the range of 800-860°C. Hydrogen generation has been demonstrated for Cu whose cycling capability was also proven [Zhang 2005]. The ZnO/Zn is a promising cycle because of its comparatively low decomposition temperature. Metal-oxide cycles are often considered in connection with solar thermal reactors which can easily achieve temperatures of 1500-2500 K.

The PSI, Switzerland, has realized a 60 kW sunlight concentrating device providing a temperature of 2200°C for the water splitting process based on the metal iron with an expected efficiency of about 20 % (see Fig. 3-39).

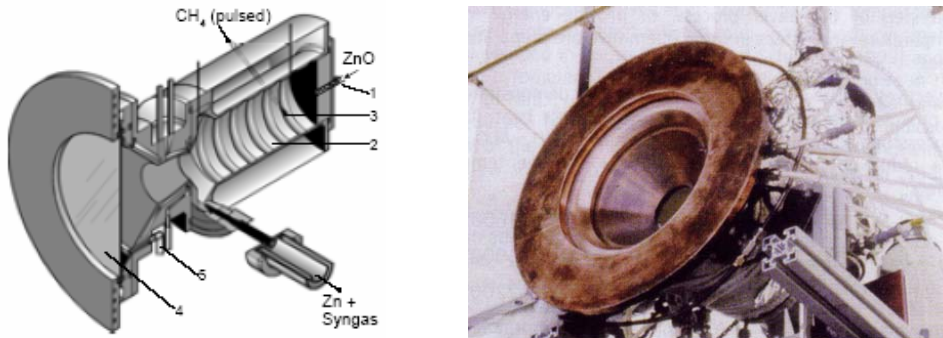


Fig. 3-39: Solar prototype reactor for ZnO/Zn two-step cycle processes [Kräupl 2000]

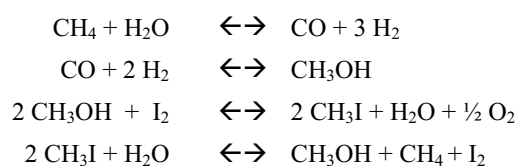
- 1) spiral-type feeder, 2) reactor cavity, 3) primary CH₄ inlet, 4) quartz window, 5) secondary CH₄ flow for window protection

3.4.5.3. Methane-Methanol-Iodomethane Cycle

One of the hydrogen production methods pursued within Korea's Nuclear Hydrogen Development and Demonstration project is the methane-methanol-iodomethane (MMI) thermochemical cycle [Shin 2004].

Fig. 3-40: Schematic of the Methane-Methanol-Iodomethane (MMI) cycle [Chang 2004]
solid line: mature status, dotted line: early R&D status

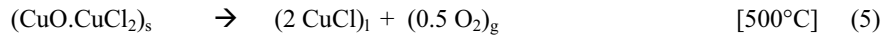
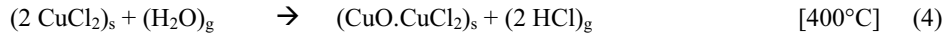
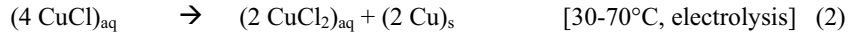
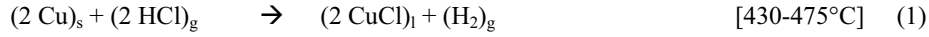
The MMI cycle consists of four chemical steps, of which the steam-methane reforming step (right-hand side of Fig. 3-40) for synthesis gas production to be used for the methanol synthesis is already proven technology. Still at an early R&D level is the realization of the iodization of methanol which can be done either by electrochemical or by catalytic reaction. For the latter option, an SO₂-catalytic chemical reaction pathway is considered possible, where the produced sulfuric acid is decomposed with the SO₂ recycled to the MMI cycle [Shin 2006].



Basic experimental works were conducted both under atmospheric conditions and under pressurized conditions up to 1.2 MPa and up to 150°C in a 100 ml pressure reactor. Works on the MMI cycle are presently discontinued in Korea, but it is still considered a potential backup solution [Shin 2006].

3.4.5.4. The ALTC-1 Process

A low temperature thermochemical hybrid cycle to be connected to Gen IV “CANDU Mark 2 SCWR” type reactor is being pursued at ANL, USA [Suppiah 2005]. The nuclear reactor is designed to provide a temperature of 530°C to the chemical reaction. The copper-chlorine (Cu-Cl) cycle, named ALTC-1 according to “Argonne’s Low Temperature Cycle 1” can be operated at a maximum temperature of about 500°C:

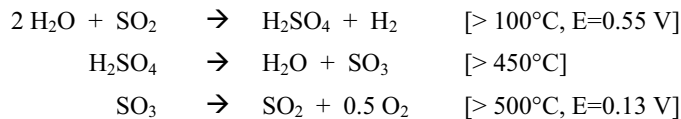


The above cycle promises an efficiency as high as 41 % and has the advantage of employing less expensive raw materials compared to the S-I cycle. The electric energy demand was assessed to be 39 % of the total energy. The viability of all reactions and, in particular, the H_2 and O_2 generation was demonstrated at ANL on a bench-scale level. What still needs to be checked are side reactions and completeness of the reactions.

The attractive analysis of this cycle, with an efficiency over 40% published by ANL [Lewis 2005] assumed, in the absence of relevant data, the CuCl_2 enthalpy of mixing with water to be negligible. A closer examination of the ANL analysis shows, however, using the first law of thermodynamics that this enthalpy of mixing cannot be neglected and must be over 100 kJ/mol. Furthermore some experimental values of the enthalpy of mixing of CuCl_2 with HCl in aqueous solutions [Kosa 1994] can actually be found in the literature. Based on these data, the missing enthalpy of mixing is estimated to range between 100 and 200 kJ/mol. Taking into account an estimated but probably realistic factor of 2 for losses in the CuCl_2 separation process, the result is a minimal amount of additional energy between 200 and 400 kJ/mol, which reduces the efficiency of the cycle to a maximum value around 30 %. Indeed, recent calculations conducted at UNLV [Khan 2005] have led to a revision of the efficiency below 30 %. There are also other issues with the cycle, like the probably incomplete character of reaction (4) in the proposed range of temperature. In conclusion, the Cu-Cl cycle appears to have a low efficiency. With already one (maybe two) electrochemical steps, it seems unlikely that it could be competitive with alkaline electrolysis.

3.4.5.5. The HHLT or JNC Process

In Japan, the former JNC (now JAEA) has suggested a thermochemical and electrolytic “Hybrid Hydrogen Process in the Lower Temperature Range”, HHLT, also called JNC process, to be connected to a sodium-cooled Fast Breeder Reactor (FBR) [Nakagiri 2005]. The process is based on the Westinghouse hybrid cycle, which usually requires high temperatures $> 800^\circ\text{C}$ for the SO_3 decomposition step. At an FBR typical coolant outlet temperature of 500°C , the decomposition fraction of SO_3 is not more than ~8 % which, however, can be raised significantly, if electrolysis by ionic oxygen conductive solid electrolyte is applied (see Fig. 3-41). The reaction equations are:



where the required voltage for the SO_3 splitting reaction is expected to be $< 0.2 \text{ V}$ (0.13 V theoretical) at 500°C .

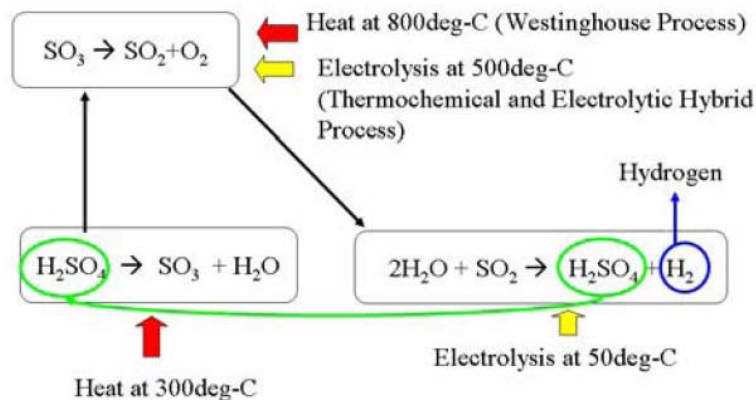


Fig. 3-41: JNC cycle as modification of Westinghouse cycle [Chikazawa 2005]

For the conceptual design of an FBR providing 395 MWt and 82 MWe, hydrogen could be produced at a rate of $47,000 \text{ Nm}^3/\text{h}$ assuming an overall efficiency of 42 % [Nakagiri 2004]. Drawback of this cycle, however, and adding to the investment cost is the employment of two electrolyzers, one in a high-concentration acid at lower temperatures and one in a corrosive atmosphere at higher temperatures.

3.4.6. Limits of Number of Reactions and Separation Energy Cost

An important factor to consider when evaluating a thermochemical cycle is the number of reactions involved. With decreasing maximum temperature, the number of required reaction steps increases. This leads to a decreasing efficiency and on the other hand to the need of separating the reaction products and of transporting the products/reagents between the reaction stages. Irreversibility and costs of each step are cumulative, quickly lowering the overall efficiency and increasing the final cost of the global process.

A minimal irreversibility (or driving force) is necessary to drive each step, especially at low temperature, which is around 40 kJ/mol for a chemical reaction as well as electrolysis. To this irreversibility, it needs to be added the minimal enthalpy to separate the reactants from the products with a high purity (corresponding to the term of ideal mixing entropy) with a minimal amount of 20 kJ/mol in practice to achieve purity of at least 10^{-3} molar.

In the case of low-temperature exothermal reactions, this additional heat can not be recovered in other steps of the process and has to be added to the final heat demand. Therefore it is doubted whether cycles with at least two exothermal chemical reactions at low temperatures or cycles with

five or more reactions will ever be competitive. Also it is obvious that cycles with two or more electrolytic steps will not be viable because of the cumulative over-voltage and large investment costs needed.

The separation work required has a strong influence on the efficiency of the cycle processes. Ideal are those where the real work is not far away from the theoretical values which can be approached the higher the “low-temperature” value is. A typical separation problem is the energy needed to separate hydrogen from water vapor (for example at the exit of a high temperature electrolysis system operating with a voltage near the theoretical reversibility point). The easiest way is to cool the water-hydrogen mixture to ambient temperature. But doing so, the heat of vaporization of water will usually get lost and can not be recovered directly to heat the water at the entrance of the electrolyzer.

Some reactants are particularly difficult to separate. For example, the energy needed in practice to separate halogen acids (HBr, HCl, HI), which are commonly proposed in thermochemical cycles, is much higher than just the enthalpy of mixing. This separation energy loss, however, can be bypassed, if the products are kept together when being processed in the next step (for example using a reactive-distillation column to produce H_2 from water/HI instead of trying to distillate HI before decomposing HI in water and hydrogen), but rarely are the optimum concentrations of species similar in different process steps.

Following the second law of thermodynamics, to achieve the best efficiency, heat must be provided at the highest temperature compatible with the heat source, as the efficiency of the process is limited by the Carnot law applied to all temperature levels balanced by the heat needed at these levels (and not simply Carnot law applied to the highest temperature level as commonly used). Therefore endothermal reactions at low temperature levels should be avoided. Free enthalpy of each reaction must be negative or very near zero to avoid too large dimensions of apparatus (a typical negative example in this respect is the UT-3 process).

3.4.7. Chemical Elements and Compounds Involved in Thermochemical Cycles

One of the major questions in a thermochemical cycle process is whether the reactions are as expected or whether there are undesired side reactions which is influenced by both the excess input of chemicals and by the residence time in the reactor. This can theoretically be answered by applying computer models to determine simultaneous equilibria. The fact that in some processes the low-temperature step rather than reducing the hydrogen reduces other elements has led to the proposal of using a hybrid process replacing the low-temperature step by an electrolysis step.

Computer models also allow the determination of the required excess (over-stoichiometric) masses which become necessary due to the formation of stable interim compounds, but on the other hand enhance side reactions. Therefore it is necessary to find a compromise between completion of reaction, phase separation, side reaction, and energy loss.

An often neglected aspect is the abundance of a given element (see Table 3-12). If relatively low, these rare resources may be depleted rapidly and the cost of extraction/purification of raw material will increase significantly as supplies dwindle.

The minimum maintenance cost is the flow loss running in the oxygen and hydrogen product typically around 0.01-0.1 % of hydrogen production (lower limit of typical industrial standard in chemical industry). This gives the order of magnitude of resources needed.

Worldwide hydrogen production through thermochemical cycles should be around 10^8 mole/s to have a significant impact on the transportation sector (assuming an optimistic 40 % efficiency for fuel cell and transport and storage). The assumption of maintenance losses for the various chemical elements applied of (at least) around 10^5 mol/s would amount to a total annual loss of about $3 \cdot 10^{10}$ moles. This has to be compared with the worldwide production of the respective raw material and typical world wide reserves estimations.

Table 3-12: Availability and production cost of various elements used in thermochemical cycles

Element	Annual production [t/yr]	Resource [t]	Resource base [t]	Price (2000-2004) [US \$/kg]
Europium	97	100,000	200,000	4800
Selenium	1500	82,000	170,000	13 to 66
Tantalum	1270	43,000	150,000	66
Bismuth	3800	330,000	680,000	6.8 – 8.2
Mercury	1750	120,000	240,000	1000 - 22,500
Iodine	20,000	15,000,000		15

For the example of selenium, several cycle proposals are based on this element. It is attractive because it belongs to the same column of the Mendeleev table of oxygen and sulfur. Selenium by itself is not expensive as it is a by-product of copper mining. However, estimations of world reserves are around 100,000 tons or $1.25 \cdot 10^9$ moles. In contrast, the expected consumption of Se due to operational losses represent 24 times the world total reserves. A minimal worldwide reserve that is 20 times the expected annual operational losses (final value depending on element weight and stoichiometry of reactions) seems necessary to insure the industrial feasibility of the project (see Table 3-12). Other elements with relatively low worldwide reserves are tantalum, europium and bismuth. Proposed cycles based on these elements may not be industrially feasible either.

For uncommon elements like iodine, a deep analysis seems necessary. It was already mentioned above that the S-I process is practicable only when operated at over-stoichiometric mixtures with high excess of iodine (and steam). Nine moles of I_2 are necessary to produce one mole of H_2 . Based on present market prices for iodine, it means that 1 kg of H_2 requires the handling of a US\$ 20,000 worth amount of iodine. This again puts stringent limitations to the iodine loss in the S-I process with respect to the H_2 cost making iodine recovery one of the key issues. The amount of iodine to be handled in a 600 MWt plant has been assessed to be 3000 t [Vitart 2005].

Another issue is the toxicity of the various elements. Examples are the toxic gases SO_2 , SO_3 in the S-I process, or bromine in the UT-3 process. Also cycles that contain Hg, Se or Cd may be limited in their industrial application because of the very low allowable releases (e.g., per U.S. EPA regulations).

3.5. High Temperature Electrolysis

3.5.1. High Temperature Electrolysis Versus Alkaline Electrolysis

Another principal variant of electrolysis considered promising for the future is the high temperature electrolysis (HTE). Unlike water electrolysis, the total energy demand of electrolysis in the vapor phase is reduced by the heat of vaporization which can be provided – much cheaper – by thermal rather than of electric energy. The electricity input is decreasing with temperature as can be seen from Fig. 3-42, and is by 35 % lower compared to conventional electrolysis in the high temperature range of 800-1000°C. Also the efficiency at this high temperature level is significantly better.

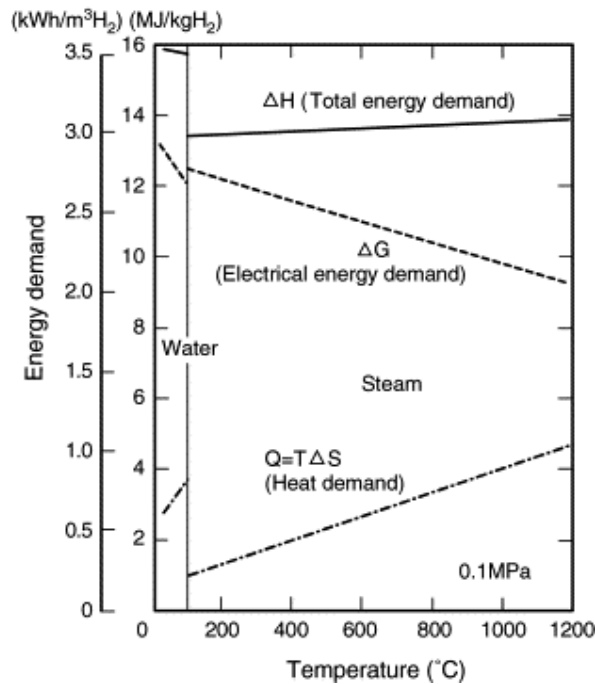
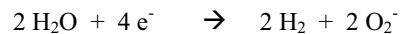
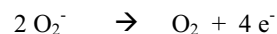


Fig. 3-42: Energy demand for water/steam electrolysis [Hino 2004]

Essentially the electrolytic cell consists of a solid oxide electrolyte with conducting electrodes deposited on either side of the electrolyte. A mixture of steam and hydrogen at 750-950°C is supplied to the cathode. At the cathode-electrolyte interface, it comes to the reaction:



Oxygen ions are drawn through the electrolyte by the electrical potential, until they combine to O₂ at the electrolyte-anode interface:



The oxygen flows then along the anode (lanthanum-strontium-manganite), while the hydrogen-steam mixture passes along the cathode (nickel-zirconia) on the opposite side of the electrolyte.

At these high temperatures, all reactions proceed very rapidly. The steam-hydrogen mixture exits and is then passed through a separator to separate hydrogen. The feed gas stream to the HTE cell contains a 10 % fraction of hydrogen for the purpose of avoiding Ni corrosion which is in the top layer of the cell (see Fig. 3-43).

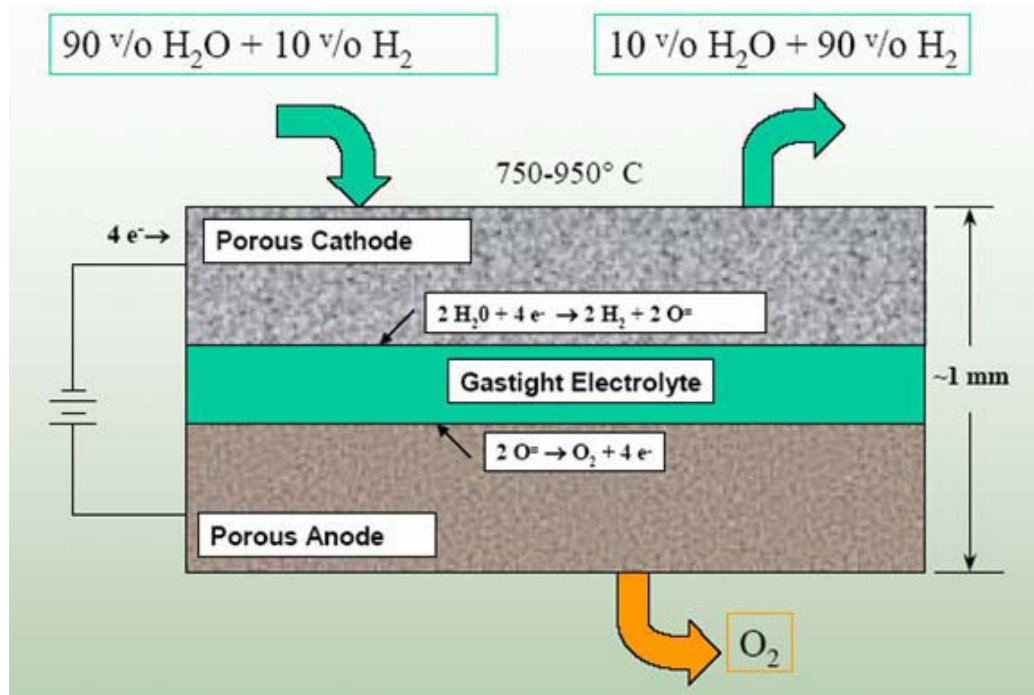


Fig. 3-43: Schematic of a planar steam electrolysis cell

Using these thermodynamic data, one can deduce the minimum primary energy necessary to electrolyze water by applying a penalty of $1/\eta$ (where η is the heat to electricity conversion efficiency) to ΔG , and adding it to $T\Delta S$. This leads to the two upper curves shown in Fig. 3-44, where two values of η have been assumed, respectively 33% for near term reactors and 50% for long term reactors.

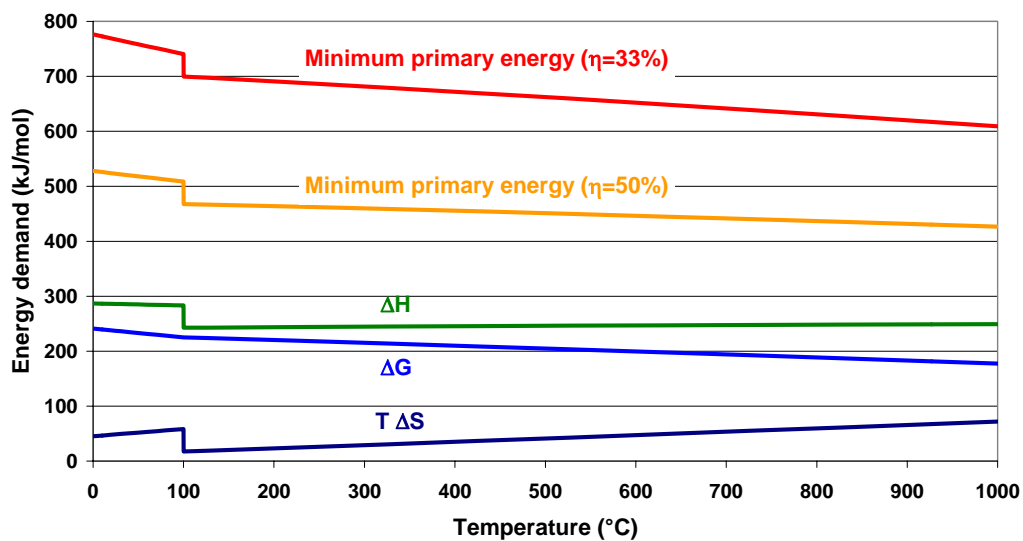


Fig. 3-44: Minimum primary energy demand for water electrolysis

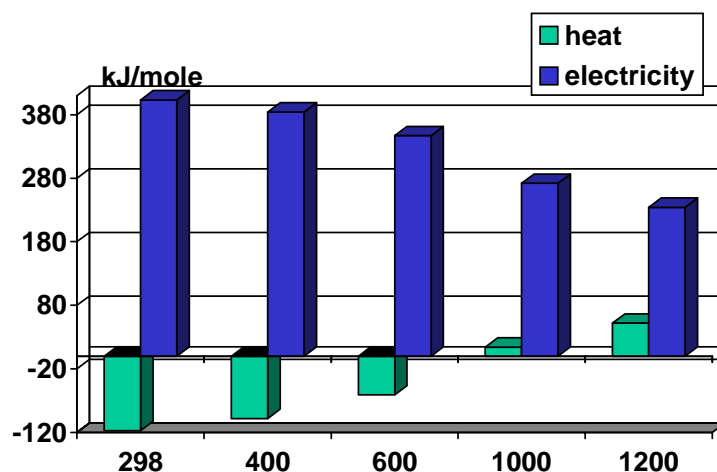


Fig. 3-45: Heat and electricity demand in preliminary HTE simulations for one mole of hydrogen production (source: CEA)

One can clearly see on this graph that a large part of the energy demand reduction is related to water vaporization, which does not require high temperature heat. Once vaporization is achieved, the extra heat necessary to reach the high temperature required to have a high enough ionic conductivity in the electrolyte may be also be provided by self heating of the cell thanks to its electrical resistance.

With respect to the electricity requirements, values of 2.6-3 kWh/Nm³ of H₂ are expected (see Fig. 3-42). The heat for steam production to supply the cells, however, must be added. The process with the production of hot steam and cell operation at high temperatures requires especially a solid oxide membrane electrolyte. For example, Y₂O₃ stabilized ZrO₂ acting as both separator and electrolyte is used, where oxygen ions start migrating when electricity is applied. Steam electrolysis at about 800°C will have an efficiency in the range of 35-45 % approaching that of thermochemical cycles. The efficiency will even rise to around 50 % at 900°C. It needs a water cleanup system (recycling in stationary systems). A major problem was identified to be the small size of the electrolysis cell with a thickness of not more than 2.5 mm. HTE corresponds to the reverse process of the solid oxide fuel cell; respective devices could be operated in both modes. HTE may therefore take benefit of on-going R&D efforts in the SOFC area.

High-temperature electrolysis of steam was investigated in the 1980s by the German Dornier company in the process called “HOT ELLY”. An electrolysis tube consisting of 10 solid oxide cells (10 mm length each, 13 mm diameter) in series with porous thin layers as electrodes was used to produce H₂ at a rate of 6.8 NI/h at 1000°C and achieving an efficiency of 92 % [Dönitz 1982, Erdle 1995]. It also confirmed the lower electricity requirements. Still total production costs (80 % of which for electricity) were too high, so that the project was eventually discontinued.

HTE was later also tested by JAEA in a bench-scale facility with the main aim to derive design data on the process characteristics. The experiments were conducted in a serial arrangement of 12 tubular cells. Hydrogen yield at a temperature of 850°C was 4 NI/h, which was increased to 7.6 NI/h for a temperature of 950°C. Tests were also started with planar electrolysis cells. Efficiencies achieved were still at a very low level., e.g., from a solar furnace or a high-temperature nuclear reactor [Hino 2004]. This activity at JAEA has also been terminated in the meantime.

The INL in the USA is presently conducting an experimental program to test solid oxide electrolysis cell stacks combined with materials research and detailed CFD modeling [O’Brien 2005]. In the most recent test in 2005, an H₂ production rate of 162 NI/h over 197 h could be verified using a 22 cell stack (Fig. 3-46) and no problem in stack performance was observed. A 1000 h test is planned to be conducted in 2006 [Herring 2005].



Fig. 3-46: A 22-cell stack as was used by INL in the summer 2005 experiment [Herring 2005]

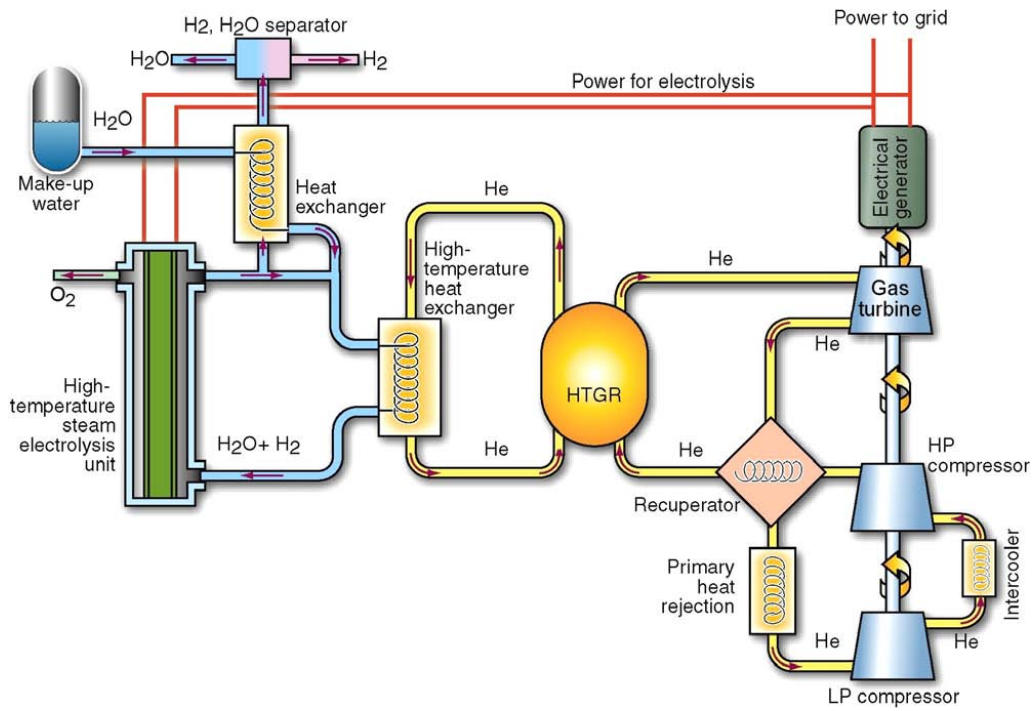


Fig. 3-47: HTGR + HTE [Herring 2003]

A schematic of a nuclear hydrogen production plant using high temperature electrolysis is shown in Fig. 3-47. The reactor (in this case an HTGR) supplies both the electricity and the steam to the electrolytic cell. The steam generator supplies very superheated steam to the cells at a temperature of 750-950° C, and a pressure of 1-5 MPa. The input gas contains both steam and hydrogen in order to maintain reducing conditions at the electrolytic cathode. Design parameters for the HTE cells are given in the following Table 3-13 [Herring 2004]:

Table: 3-13: Electrolysis cell characteristic design of conceptual HTE plant

Design parameter	Value
Active cell area	100 cm ²
Electrolyte Material Thickness	Scandia stabilized zirconia 10 µm
Anode Material Thickness	Strontium-doped lanthanum-manganite 1500 µm
Cathode Material Thickness	Nickel-zirconia cermet 50 µm
Bipolar plate Material Thickness	Nickel-aluminum 2500 µm
Total cell height	4.06 mm
Number of cells per stack	500
Stack height	2.03 m
Total number of cells	12*10 ⁶
Total active area	120,000 m ²
Total hot volume	26 x 15 x 5 m ³
Cell operating conditions	850°C, 5 MPa, 0.2 A/cm ² , 1.1 V
Water consumption	22.3 kg/s
Hydrogen production (LHV)	2.5 kg/s or 238 t/d @ 50 % overall efficiency

3.5.2. Main Technological Issues

Two design concepts are possible, a planar and a tubular one (see Fig. 3-48). In the HOT ELLY process in Germany [Erdle 1995], the tubular design was first selected for the purpose of mechanical stability and also the fact that fabrication of thin-walled ceramic parts with large surfaces was not state of the art. The tubes were vertically deployed on support tracks which contained steam input and hydrogen output ducts. The top right part of the figure shows a respective 2 kW HOT ELLY module. As technology progressed, the Dornier company moved after 1987 over to the fabrication of planar electrolysis cells.

JAEA has tested both planar and tubular versions. The planar design appeared to be the most effective, but also the most fragile one. The limits of the performance of the tubular design were attributed to the presence of the tube which limits the vapor flow and ohmic losses in electrical connections. Current densities were demonstrated between 0.3 and 1 A/cm².

The constraints induced by thermal cycling and the variation of dilatation coefficients are major remaining problems. Also equipment costs are still very high: current evaluation reveals investment cost to be around twenty times the cost of alkaline electrolysis, but it is expected to decrease rapidly.

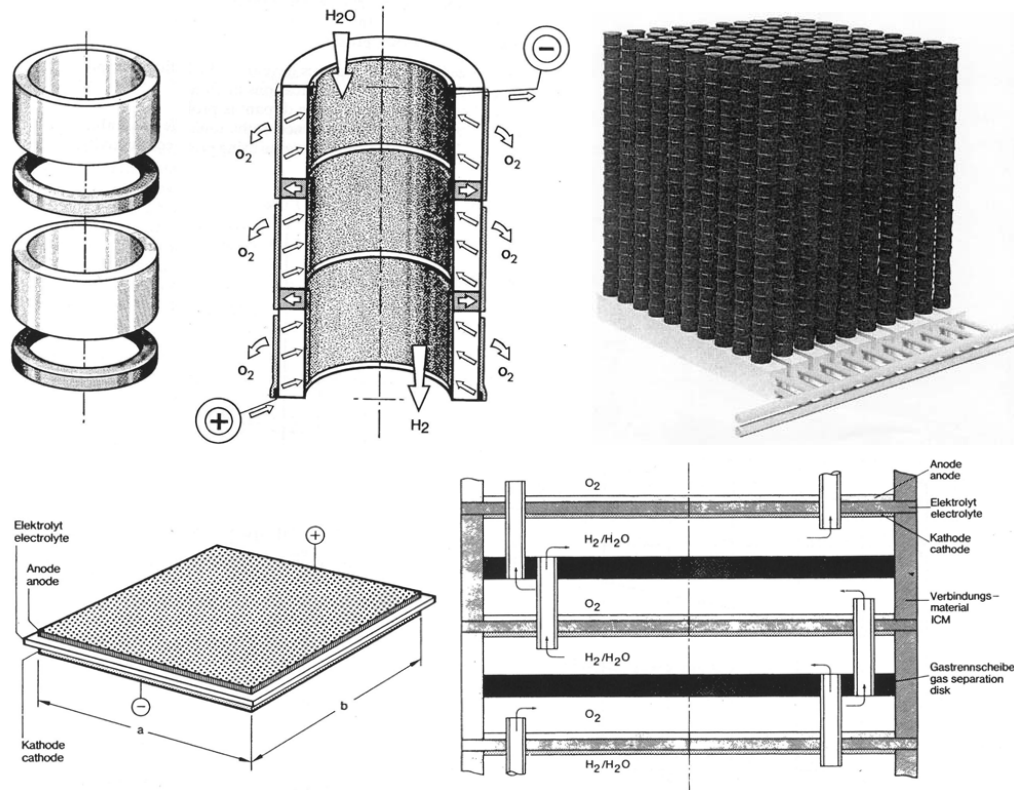


Fig. 3-48: Tubular (top) and planar (bottom) concepts of SOWE cell stacks [Dönitz 1982]

3.6. Key Factors for Innovative H₂ Production Processes

3.6.1. Efficiency

To evaluate the different hydrogen production methods, the basic thermal hydraulic relationships are used that govern the efficiency of thermochemical processes for hydrogen production as well as that of electricity generation in combination with electrolysis.

Fig. 3-49 shows the relationships of ΔH and ΔG , respectively, as a function of temperature for electrolytic water splitting (Case A, left) and for a fundamental two-reaction thermochemical method (Case B, right). As a result, the final conversion efficiency is fundamentally independent of any route of conversion technology assuming the same conditions for input and output [Ogawa 2003].

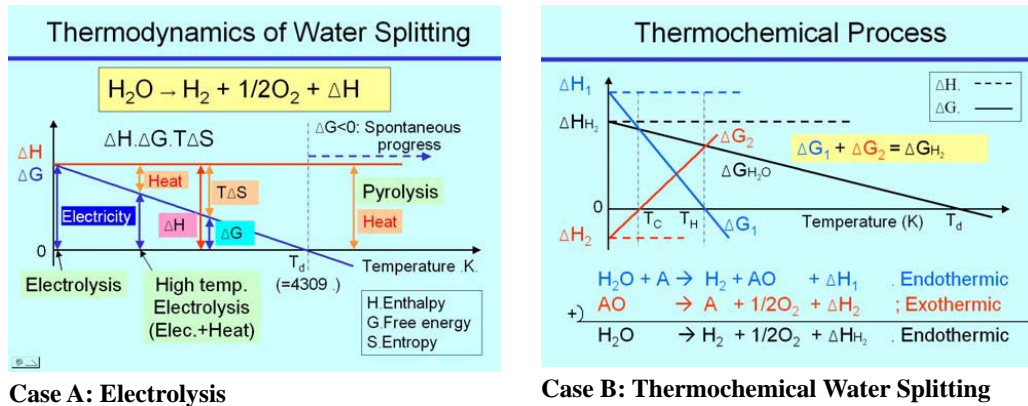


Fig. 3-49: Analysis of hydrogen production by electrolysis and thermochemical processes [Ogawa 2003]

Both processes – electrolysis and thermochemical cycle – are governed by the same Carnot law depending only on the upper (T_H) and lower (T_L) operational temperature as well as on the dissociation temperature (T_D), which is 4309 K for autothermal water splitting. The efficiency at temperatures ≥ 4309 K, i.e., the ultra high temperature one-step direct thermal splitting of water, is 100 %. At lower temperatures, it can be determined by [Ogawa 2003]:

$$\eta = \frac{T_H - T_L}{T_H} * \frac{T_D}{T_D - T_C}$$

This formula has a direct impact on the choice of technical options:

- Operational temperatures of the heat source and the process should be as high as technically feasible;
- Thermochemical processes or electricity generation at lower temperatures will always be inferior with regard to the thermal efficiency η ;
- Since the dissociation temperature is extremely high, water splitting always needs several successive processes to provide the dissociation energy, i.e., electricity generation plus electrolysis (plus heat) or a follow-up of different endothermic and exothermic chemical processes at lower temperatures.

Enhancing the efficiency for electricity generation and for electrolysis by temperature increase is one option for improving hydrogen production. Electrolysis can be done remotely and decentralized or with direct coupling to the reactor by using high temperature steam, the so-called “hot electrolysis” route, shown in Fig. 3-50. This process can benefit extensively, for example, from the solid oxide fuel cell development which is the inverse process using the same functional elements.

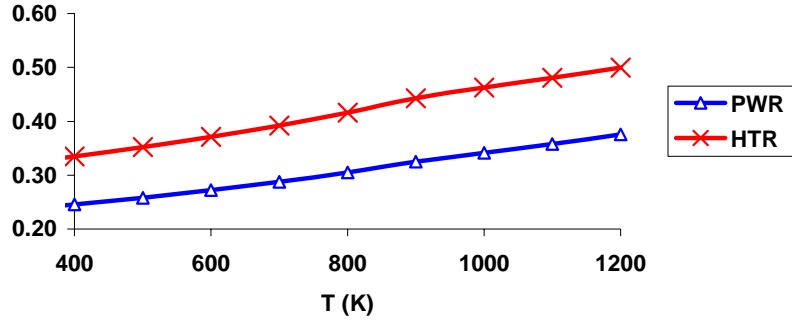


Fig. 3-50: Efficiencies of HTE using electricity from HTGRs or LWRs

Another very efficient and cost-effective way to produce hydrogen using nuclear energy is through the use of high temperature heat provided by a reactor to a thermochemical water splitting cycle. This heat is utilized through a set of coupled chemical reactions that result in the decomposition of water into hydrogen and oxygen. Energy is introduced via endothermal high temperature reactions and rejected via exothermal low temperature reactions. The chemical reagents are recycled within the process and the net effect is to convert heat energy and water into hydrogen energy.

The high temperature processes for the production of hydrogen are at the first glance attractive due to high efficiency perspectives induced by the basis equation of thermodynamics second principle : $\eta = (1 - T_c/T_h)$, where T_h is the high temperature value and a T_c cold temperature value, often taken equal to the ambient air temperature. However, all these processes also require a certain quantity of electrical or mechanical energy. Electricity can be produced either by diverting a fraction of the heat generated from the high temperature source, from another source, or from the grid. In all the cases the electricity is produced from a heat source with an efficiency of η_{el} . An amount of work equal to W , needs at least an amount of heat $Q' = W/\eta_{el}$. In every case where the same primary source is used to produce the work and the heat (Q) necessary for the process of production of hydrogen, it is necessary to take at least $Q_T = Q + (W/\eta_{el})$ from the heat source. If these quantities are normalized to a unity of produced hydrogen, it is possible to define a global efficiency [Vitart 2005]:

$$\eta_T = \frac{HHV}{Q_T} = \frac{HHV}{Q + (W/\eta_{el})}$$

where HHV is the enthalpy of formation of liquid water at ambient temperature or the higher heating value. Such a global efficiency must be compared to the global efficiency of the chain:

High temperature heat source \rightarrow electricity \rightarrow production of hydrogen by alkaline electrolysis, for which it is easy to estimate a global efficiency:

$$\eta_{T, alkaline electrolysis} = \eta_{el} * \eta_{electrolyzer}$$

where $\eta_{electrolyzer}$ is the system efficiency of the electrolyzer, namely 73 % for the recent Norsk Hydro process, which gives:

$$\eta_{T,alkalineelectrolysis} = 0.73 \eta_{el}$$

In the case where both heat and electricity are produced from the same primary energy source, a first key for a high temperature process for the production of hydrogen is that its global efficiency should be higher than the global efficiency of the alkaline electrolysis using electricity produced by the nuclear source of reference.

If the primary energy source considered is a nuclear one of the generation IV concepts, this key fits very the recommendations of GEN IV: “The Economic Goals of Generation IV Nuclear Energy Systems”, as adopted by Generation IV International Forum (GIF), are to have a life cycle cost advantage over other energy sources, and have a level of financial risk comparable to other energy projects.

All the systems related to the Generation IV concepts will need important R&D expenditure, they can be justified only in the context of very large-scale hydrogen production, for which the basic economic rules will apply.

If the nuclear source is a Very High Temperature Reactor (VHTR), the high temperature processes could be either thermochemical cycles or high temperature electrolysis. In both cases the complexity of the processes implies that capital and maintenance costs would be higher than for alkaline electrolysis and the financial certainly higher, because they are non-proved processes. The efficiency of the production of electricity (heat-to-work conversion) can be estimated in the range $0.45 \leq \eta_{el} \leq 0.5$.

The upper bound of the global efficiency of high temperature process for the production of hydrogen according to the above reaction scheme can be estimated to 51 % [Vitar 2005]. Further refinements of the flow sheet with more realistic values for component efficiencies will lead to a lower overall efficiency. However, it will have to be greater than 33 %, but preferably 36.5 % or more.

3.6.2. Heat Matching with Heat Source

For nuclear heat sources, cycles with a reaction taking place at a temperature > 1200 K should not be considered because it would be higher than what Generation IV concepts are expected to provide. Also the range of 1075-1200 K is very questionable because of the exponential increase in cost of materials with increasing temperatures.

It is also very important to match the Q/T diagram of the heat source. For HTGR heat, this is provided between around 500°C and 850-1150°C (see Fig. 3-51). Such heat profile is not adequate, if the heat needed for the process is exclusively at too high temperatures.

The heat demand for sulfuric acid decomposition is – at a first glance – well suited with respect to the heat provided by an HTGR, which explains the great interest of nuclear engineers for such processes. Respective calculations, however, were made at thermodynamic equilibrium. The necessity of using a catalyst for SO₃ decomposition will increase the temperature level of the SO₃ by some 50-100°C which may perhaps be no longer compatible with the final design of the heat provided by the HTGR.

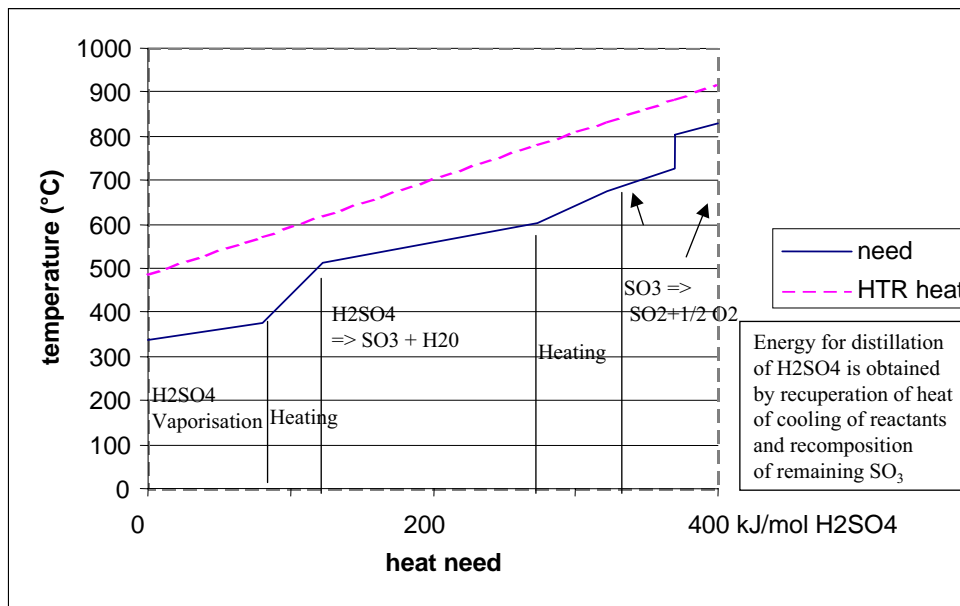


Fig. 3-51: Preliminary Heat matching calculations with heat source for sulfuric acid decomposition (source: CEA)

For high temperature electrolysis, difficulties are different: In HTE, only a small percentage of energy is needed as heat, which is around 90 kJ/mol for an electrolyzer operating at 950°C.

This splits up into ~ 60 kJ/mol of H_2 around 200°C to decompose steam and only 30/40 kJ/mol (maximal realistic value) at 950°C for the electrolyzer itself, if supposed that the heat of the products is recovered to heat up the reagents. Therefore the HTGR will only provide less than 10 % of useful heat energy to the electrolyzer, if compared to a solution where heat is provided at low temperatures only, resulting in an increase of efficiency from around 40 to 42 %.

The question is whether it is really worth coupling an HTE system to an HTGR and achieving such a low theoretical gain of efficiency:

- Hydrogen has to be kept separated from the nuclear plant to avoid hydrogen explosion hazard. This will be on the expense of losses in the heat exchanger, which will lower the global efficiency of the system, and of increasing investment cost for security reasons. It has been suggested to increase the number of water recirculating with hydrogen to avoid hydrogen explosion; but doing so will reduce the final efficiency of the process because the heat of vaporization of water can not fully be recovered.
- HTE systems are supposed to be very fragile, especially under thermomechanical cycling conditions. Repeated maintenance of HTGR systems will lower their lifetime and their availability.
- Allothermal instead of autothermal operation of HTE will increase the number of cells required, therefore directly impacting the already high investment cost.

The situation is different for countries where renewable sources of heat are existing. While biomass deserves special examination, two other types of possible sources of heat are geothermal and solar. Geothermal produces warm and low-cost steam in a reliable and continuous way. Even if the temperature of the geothermal steam is a few hundred degrees lower than the maximum temperature needed for most of the high temperature processes for producing hydrogen, the complement of heat can be obtained by Joule heating. This appears suitable for HTE because no

additional heat is needed for vaporization and Joule heating is used to increase the steam temperature to 950-1100 K. In this concept, the electricity needed for the higher temperature heating and for the electrolyzer itself could be produced by any alternative source: hydraulics or nuclear, and not necessarily from geothermal.

Solar is an intermittent source, therefore an intermediate storage (thermal and/or chemical) would be needed for a continuous process. It would be difficult to drive a thermochemical cycle with a critical phase change. However, solar can be well suited for hybrid cycles, if chemical storage at some interface is possible. The electrical part would be still driven by nuclear or another electricity source, while the thermochemical part is driven by solar. A good example is the hybrid S cycle with easy H_2SO_4 storage at the entrance of the sulfuric acid decomposition section. However, the S-I cycle can not be readily adapted for use with a solar heat source because one of the reactions, the Bunsen reaction, is carried out near the solidification point of iodine.

3.6.3. Heat Exchangers

It is often supposed that the heat requirement for the endothermal chemical reactions is the major heat load in thermochemical cycles. But a large heat demand is always necessary to heat the chemical reactants to the reaction temperature. This amount of heat can be provided by recovering heat as higher temperature materials are cooled. This is a real challenge because of corrosion issues.

The stoichiometry of the products impacts these energy exchanges. As the free energy change of each reaction is designed not to be too negative to avoid too large exergy losses, a very large recirculation rate can be needed, leading to large energy exchanges.

A typical example is the S-I cycle. First the product of the Bunsen reaction as proposed by GA included not only HI but also a large amount of water and iodine (2 HI with 10 H_2O and 9 I_2 in GA proposal). Furthermore, as the HI decomposition process has a positive free enthalpy, a large recirculation rate is necessary in the HI decomposition section. For example, in [Goldstein 2005], a recirculation rate of 3 is proposed. This is equivalent to around 30 H_2O and 27 I_2 circulating to produce 1 mole of hydrogen. Following the design proposed in [Goldstein 2005], we see that more than 1500 kJ must be exchanged to produce one mole of hydrogen (the high heating value of which is only 286 kJ/mol).

In order to keep the contribution of external energy within reasonable limits, it is necessary to recover a part of the heat rejected by exothermal reactions. This energy recuperation will be performed through heat exchangers whose efficiencies will be depending on their pinch values (temperature differences between hot leg and cold leg of a heat exchanger). The efficiencies are better, but the technical feasibility is much more difficult for low pinch values. Since more than twenty years, it is known that this should be much more of a penalty for the HI decomposition than for the H_2SO_4 decomposition: A flow sheet elaboration presented in [Schepers 1984] has shown that for the S-I cycle, 56 % of the heat rejected by exothermal reactions of the H_2SO_4 decomposition part could be possible with pinch values not higher than 50 K for the high temperature part, while 95 % of the heat rejected by exothermal reactions of the HI decomposition part, with pinch values not higher than 10 K would be necessary.

For high water temperature electrolysis, heat exchanges are less severe. The trouble is, as the efficiency difference between high temperature electrolysis and alkaline electrolysis is quite low, it is necessary to be sure to recover the heat of products, which is problematic with pure oxygen at high temperature.

A technical-economic optimization of heat exchangers networks for HTE, performed by [Mansilla 2005] has shown that for a coupling with an HTGR and a rate of 30 % at the outlet of the electrolyzer, a minimal amount of 60 % recovering of the heat contained in the gas at the outlet of the electrolyzer, is necessary with pinch values not higher than 37 K.

3.6.4. Final Remarks

From the large list of proposed thermochemical cycles in the past (over 300 worldwide), only a few were left and deemed to have sufficiently promising potential for further detailed investigations. Among them are most certainly the two-reaction hybrid S (Westinghouse) cycle and maybe the S-I cycle which has probably the greatest efficiency potential, but for which heat exchange and raw material cost may be questionable. For solar heat, the two-reaction cycles (like ZnO_2 decomposition) seem to have the best potential. Raw material investment and maintenance, as well as energy recovery appear to be key issues for the thermochemical cycles.

Concerning high temperature electrolysis, specific attention must be addressed to the heat exchange to insure its potential competitiveness. A cheap heat source around 200°C seems the main opportunity for high temperature steam electrolysis. As described above, the coupling to an HTGR should be reconsidered.

All these innovative processes have to be finally compared to the low cost alkaline electrolysis to insure their potential viability.

3.7. Nuclear Hydrogen R&D Projects

The Generation IV International Forum“ (GIF) is a joint initiative by several countries including the EURATOM to develop the next (fourth) generation of nuclear reactor by 2030, which is, apart from being safer, more reliable, more economic, more proliferation-resistant, also expected to penetrate non-electrical markets like the supply of heat or hydrogen on a large scale. One of the most promising “Gen-IV” nuclear reactor concepts is the VHTR (Very High Temperature Reactor) with its characteristic features of direct cycle gas turbine plant for high efficiency and coolant outlet temperatures of $> 1000^{\circ}\text{C}$ for hydrogen production. Top candidate production methods are the sulfur-iodine thermochemical cycle and high temperature electrolysis, considered presently by various countries.

A parallel initiative has been started by the International Atomic Energy Agency, IAEA, called INPRO, the “International Project on Innovative Nuclear Reactors and Fuel Cycles”. While the GIF, in which both IAEA and OECD/NEA are participating as observers, is serving essentially as a designers’ initiative, INPRO includes also other countries and has a longer time horizon. It incorporates IAEA safeguard considerations more directly encompassing both designers and end-users and their requirements.

Various countries have initiated ambitious programs with the goal to bring nuclear hydrogen production to the energy market. In contrast, the European Union does not have a dedicated nuclear hydrogen production program. A respective engagement by research, industry, and policy is here mainly given by the participation in activities within the current 6th Framework Programme of the EU. With CHRISGAS, SOLREF, HYTHEC, and H₂H₂, projects have started dedicated to the hydrogen production by biomass gasification, steam reforming, thermochemical cycles, high-temperature electrolysis. On the nuclear side, there is the RAPHAEL project, acronym for „Reactor for Process Heat Hydrogen and Electricity Generation“ which started in April 2005. This EU Integrated Project will treat the pertinent aspects of material development, HTGR fuel technology, nuclear waste management, and coupling to hydrogen production technologies.

3.7.1. France

Based on principles of the HTR Modul concept and evolving from the US GT-MHR concept, AREVA-NP (formerly: Framatome-ANP) is developing the conceptual design of ANTARES, an indirect cycle power conversion system which can be adapted to different cogeneration schemes. In the VHTR concept for electricity generation, the block-type core generates a power of 600 MWt with a helium outlet temperature of 850°C at a pressure of 5.5 MPa (see Figs. 3-52 and 3-54) [Copsey 2005]. Another version of ANTARES dedicated to hydrogen cogeneration by either the S-I cycle or HTE, the reactor coolant outlet temperature is 1000°C . Heat is transferred in the IHX to the secondary coolant, a mixture of He and N₂ raising its temperature to 950°C (see Fig. 3-53). Process heat supply to the chemical process is finally at a temperature level of 925°C [Gauthier 2004]. A preliminary schedule foresees the conceptual design of ANTARES to be completed by 2008 and the final design to be completed by 2012.

The IHX is the only novel component that needs further development for the considered operation range. Options are, besides the “conventional” tubular concept (as a fall-back solution), the plate machined heat exchanger and the plate fin heat exchanger designs, which should be capable of sustaining also higher temperatures typical for direct cycle applications. An extensive R&D and testing program is necessary, and has started already, to investigate appropriate materials [Copsey 2005, Breuil 2006]. Design selection is planned to be made by 2008.

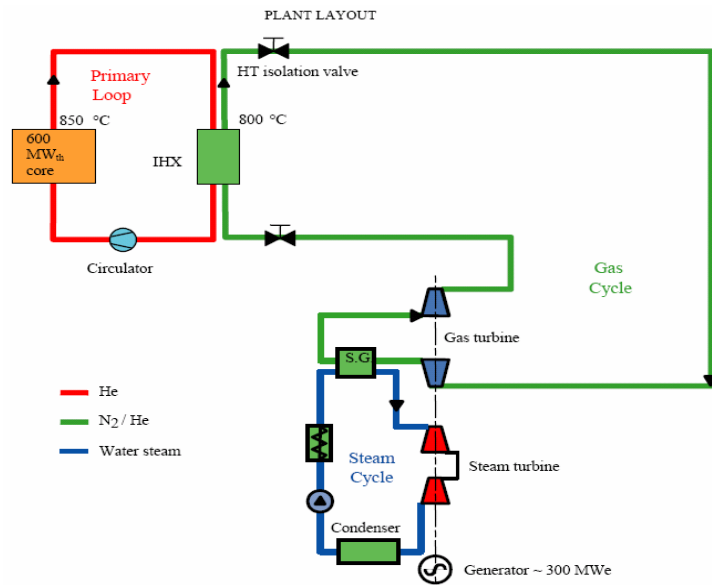


Fig. 3-52: Principle of the AREVA-NP combined cycle cogeneration HTGR [Copsey 2005]

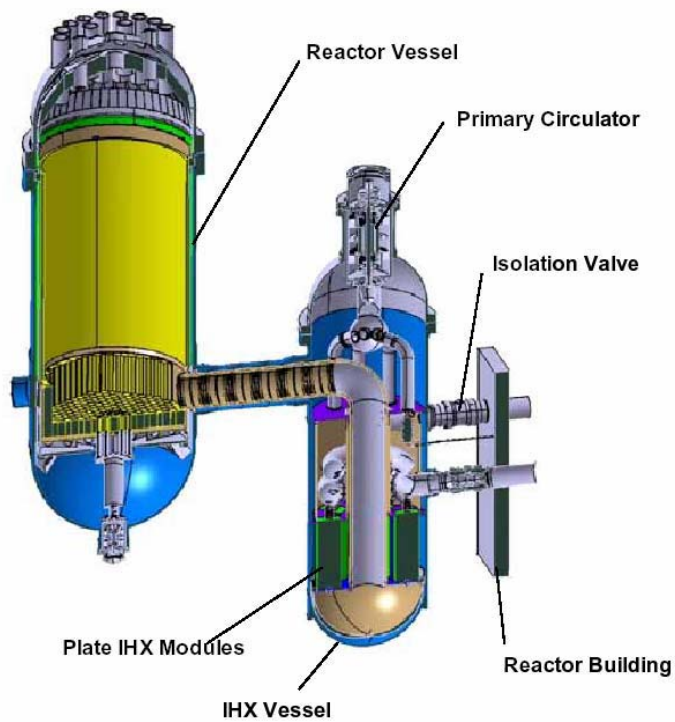


Fig. 3-53: AREVA-NP's VHTR nuclear heat source ANTARES (source: AREVA-NP)

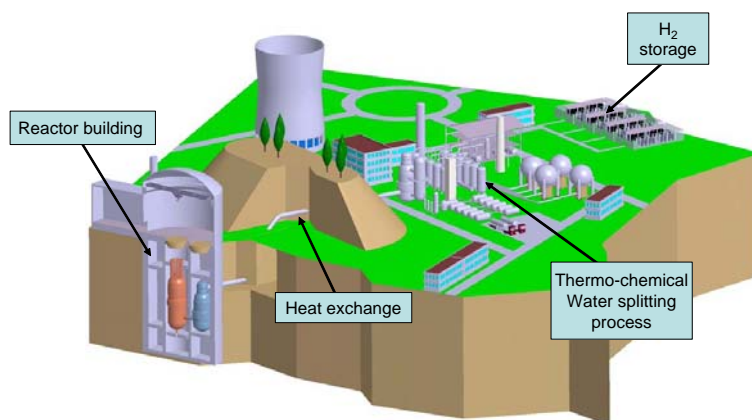


Fig. 3-54: Potential arrangement of a dedicated 600 MWt VHTR for H₂ production at a rate of 1 kmol/s [Anzieu 2005]

3.7.2. Japan

The “Basic Plan for Energy Supply and Demand” of October 2003 as part of the “Basic Law on Energy Policy Making” explicitly states that hydrogen is a clean energy carrier and that a commercialization of hydrogen production systems by means of nuclear, solar, and biomass, but no fossils, is desired. With the construction and operation of the 30 MWt HTTR (first criticality in 1999), JAEA has laid the basis for utilization of nuclear process heat for hydrogen production. The reactor allows a coolant outlet temperature of 950°C to provide process heat at 905°C outside the reactor vessel, which was demonstrated in 2004 for the first time in the world. Over several years, steam reforming of methane (SMR) was considered top candidate method to be connected to the HTTR and to generate the world’s first nuclear hydrogen. However, all efforts are now concentrated on the S-I thermochemical cycle as a CO₂ emission free H₂ production method representing currently the largest hydrogen project in Japan.

3.7.2.1. Hydrogen Production with High Temperature Reactor

The reference concept for nuclear hydrogen production in Japan is based on the GTHTR300C (C = Cogeneration) reactor [Kunitomi 2004] connected to an S-I thermochemical water splitting process (see Fig. 3-55). The direct cycle block-type HTGR with a thermal power of 600 MW provides a coolant inlet/outlet temperature of 594/950°C. In the IHX, a part of the thermal power, 168 MW, is transferred to the H₂ generation process with the remaining power to be used for electricity generation of 202 MWe. Assuming an efficiency of 50 % and an availability of 90 %, the average amount of hydrogen is 24,000 Nm³/h corresponding to the supply of some 100 refueling stations (assuming ~6000 Nm³/d) to keep a total of about 160,000 FCV (~3.6 Nm³/d) being operated [Takeda 2005].

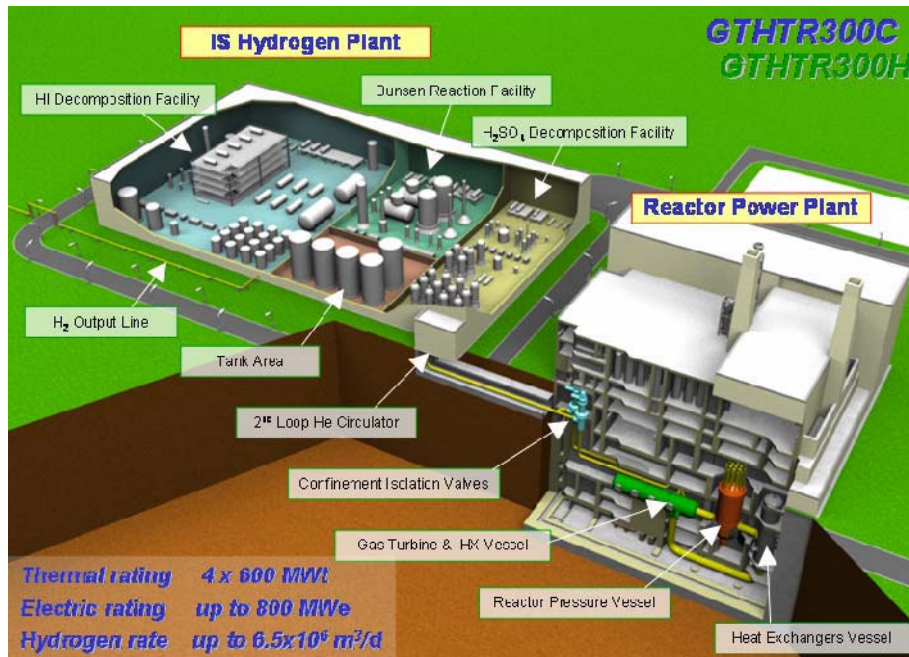


Fig. 3-55: Japan design [Yang 2005]

3.7.2.2. Hydrogen Production with Fast Reactor

FR-MR Concept

The so-called “FR-MR” concept has been suggested in [Hori 2004, Tashimo 2005] to perform methane-steam reforming at much lower temperatures, $\sim 550^{\circ}\text{C}$, by employing membrane reformer technology. Thus it would be possible to utilize the nuclear heat of sodium or lead cooled FBR or of supercritical water cooled reactors. The FR-MR is designed to be a sodium cooled fast reactor with a thermal power of 240 MWt and coolant inlet/outlet temperatures of $500/580^{\circ}\text{C}$, at an advanced stage $450/600^{\circ}\text{C}$. Heat is transferred via an IHX to the membrane reformer at a temperature 565 and 580, respectively. H₂ production is assessed to be 200,000 Nm³/h using a feed stock (CH₄) input of 50,000 Nm³/h. Nuclear heated membrane reforming can also be applied in the refining industries helping to save hydrocarbon feedstock. Furthermore it would allow coal gasification at reduced temperatures [Hori 2004].

Concept of Fast reactor with JNC Process

JAEA is also studying the concept of a sodium cooled reactor to produce H₂ in the so-called “Thermochemical and Electrolytic Process” or JNC process (see section 3.4.5.5.). Since the maximum temperature remains below approx. 500°C because electrolysis is applied, a fast reactor can be employed (see Fig. 3-56). It is designed for a power of 395 MWt to provide primary sodium at 550°C and secondary sodium at 540°C , respectively. The electrolytic steps require an electric power of 18 MW for the SO₃ electrolysis and 56 MW for the SO₂ electrolysis. Hydrogen is yielded at a rate of 47,000 Nm³/h [Chikazawa 2005].

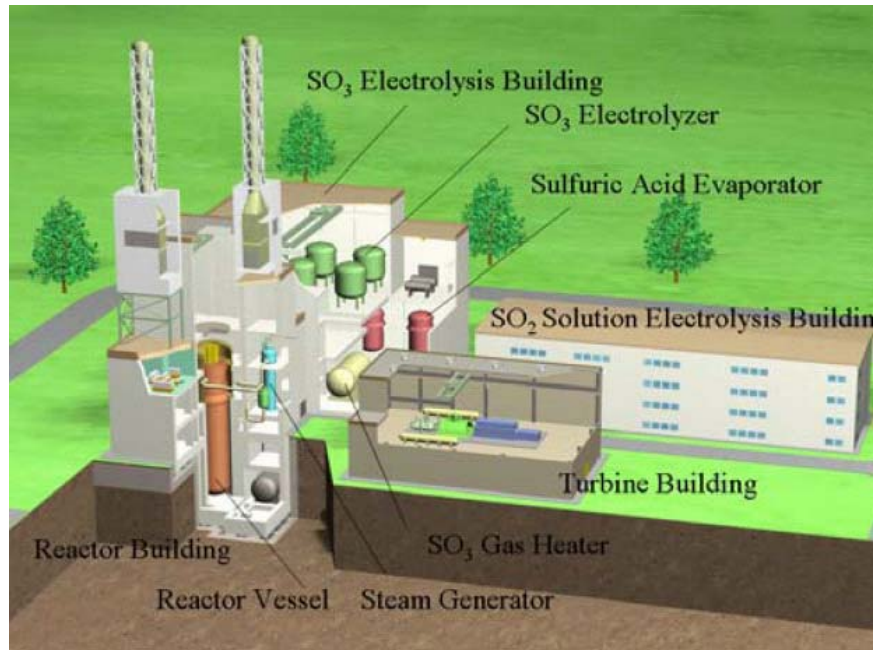


Fig. 3-56: Concept of sodium-cooled fast reactor with H₂ production by JNC process [Chikazawa 2005]

3.7.3. Korea

In March 2004, the Ministry of Science and Technology (MOST) in Korea has started the Nuclear Hydrogen Development and Demonstration (NHDD) project. Its main goal is the completion of the development and demonstration of the nuclear based hydrogen production technology by the year 2019 and the achievement of commercial nuclear hydrogen production by the middle of the 2020s to cover 20 % of the total vehicle fuel demand corresponding to 3.3 million t/yr of hydrogen [Park 2005]. While the government is leading the program in the design phase, industrial participation is required from early on to later take the lead in the construction of a demonstration nuclear plant scheduled to start operation after 2017.

The nuclear reactor is planned to be a VHTR with either a block type core to produce 600 MWt or a pebble bed core to produce 400 MWt and with coolant inlet/outlet temperatures of 490/1000°C (see Figs. 3-57 and 3-58). The reactor pressure vessel will be based upon the Korean Advanced Power Reactor APR-1400 vessel which is the largest that can be manufactured today.

Three alternative H₂ production methods are being pursued in Korea, apart from the two shown in Fig. 3-57 (S-I cycle, high temperature electrolysis) also the MMI cycle.

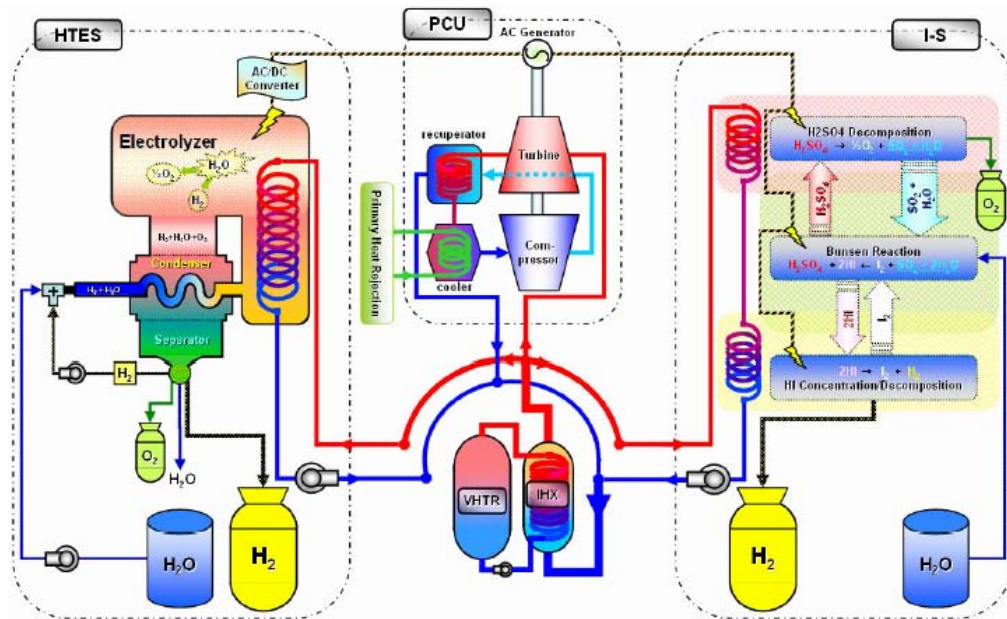


Fig. 3-57: Layout of the Korean NHDD plant [Lee 2005]

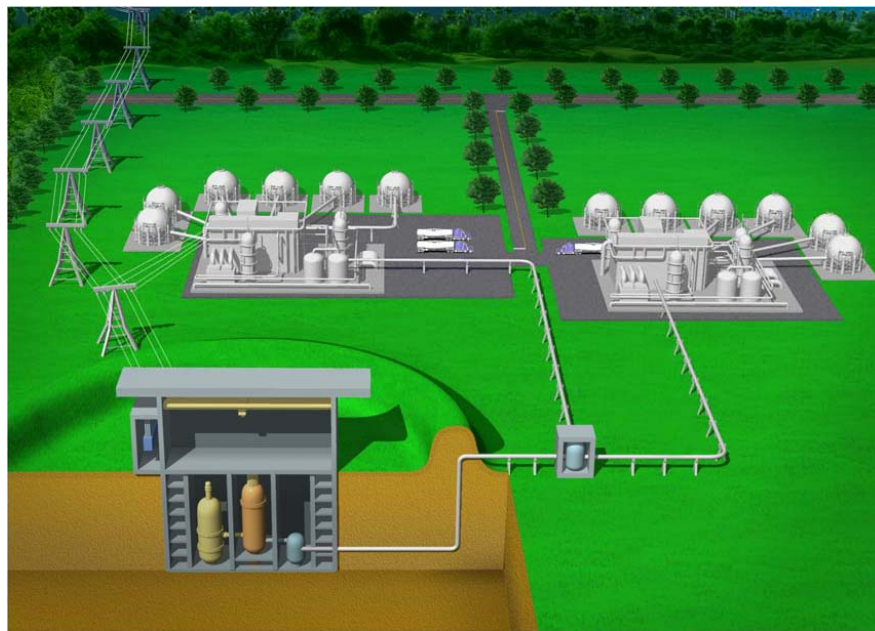


Fig. 3-58: Korea design [Shin 2005]

3.7.5. Russia

This section describes the main principles of technology developed in the Russian State Scientific Centre Institute for Physics and Power Engineering in the frame of activity on non-electricity applications of advanced liquid metal cooled fast reactors realized since 1990 [Gulevich 2005]. One of the proposed applications is the hydrogen production by methane thermal disintegration through direct contact heat transfer from the liquid metal coolant, Pb-Bi, to a hydrogen containing gaseous raw material such as methane. As is shown in the schematic in Fig. 3-59, the Pb-Bi (secondary) coolant is heated up from the primary reactor coolant (Pb-Bi, Pb, or Na) in a metal-metal IHX and circulating through a reactor vessel. Methane is given into the lower part of the vessel, where it bubbles through the hot coolant and is partially decomposed (pyrolysis). A separation device is then separating the hydrogen from the solid carbon and still un-decomposed methane. The method foresees to use a liquid metal heated by reactor coolant up to 700 °C and methane under pressure of 0.1 MPa. The assumed yield of hydrogen in this process is up to 95%.

In a similar process, instead of pure methane, a mixture of methane and water vapour enters the reactor vessel. In this case, H₂ is generated via the reforming reaction while bubbling through the hot secondary liquid metal coolant. At a temperature of 700°C, a methane partial pressure of 0.05 MPa, and a steam-methane ratio of 2, the H₂ yield was estimated to be ~ 85 %.

The above processes are based on the inherent coolants characteristics presented in Table 3-14 allowing to realize this method.

Table 3-14: Pb-Bi, Pb coolant properties involved in H₂ production from CH₄

Coolant Characteristic	Consequences
High density of liquid metal $\rho_{\text{Pb-Bi, Pb}} \approx 11,000 \text{ kg/m}^3$	Possibility of separation: liquid metal coolant and contacting liquid or gas
High boiling point $T_{\text{Pb-Bi, Pb}} \approx 1700^\circ\text{C}$	High temperature to decompose methane molecule: $(\text{CH}_4) \rightarrow <\text{C}> + (2 \text{ H}_2)$
Absence of chemical interaction between Pb-Bi, Pb and contacting liquid or gas	Possibility of phases separation

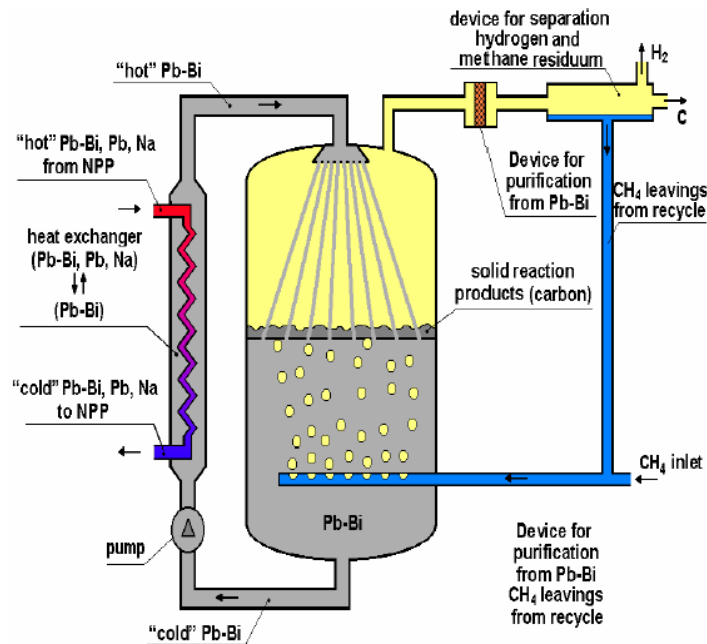


Fig. 3-59: Schematic of H₂ production by methane thermal disintegration in the reactor coolant [Gulevich 2005]

3.7.6. USA

The United States are currently designing a “Next Generation Nuclear Plant” (NGNP). This government-sponsored demo program is based on a 400-600 MWt full-scale prototype gas-cooled reactor to provide electricity and process heat at 900-1000°C. From the total thermal power, a part of 100 MW are planned to be consumed for hydrogen production using the S-I process as reference method, alternatively high-temperature electrolysis.

3.7.6.1. The H₂-MHR

The goal of the “Nuclear Energy Research Initiative”, NERI, is to demonstrate the commercial-scale production of hydrogen using nuclear energy by 2017. The Modular Helium Reactor (MHR) has been suggested as the Next Generation Nuclear Plant, NGNP, Gen IV reference concept for nuclear hydrogen generation on the basis of either the S-I thermochemical cycle or high temperature electrolysis. The so-called H₂-MHR based on the principle of the GT-MHR is a helium cooled, graphite moderated, thermal neutron spectrum reactor directly coupled to a Brayton cycle power conversion system with a thermal power of 600 MW (Fig. 3-60). It is designed for averaged coolant inlet/outlet temperatures of 590/ 950°C working with an efficiency of 48-52 % for electricity production [Richards 2005b].

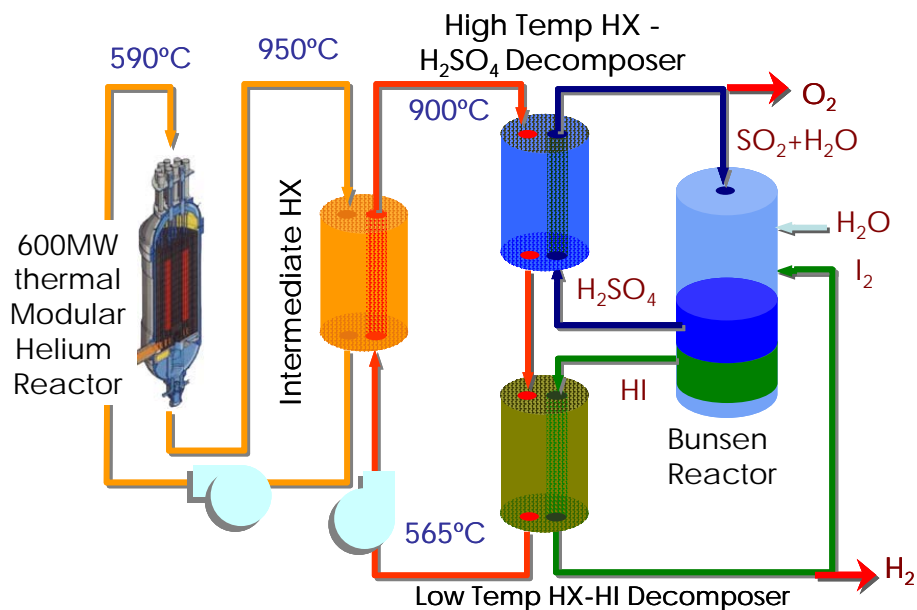


Fig. 3-60: Concept of the US H₂-MHR combined with the S-I cycle



Fig. 3-61: US view of H₂-MHR combined with both S-I and HTE systems

In the HTE based H₂-MHR, 68 MW of heat is transferred through a printed circuit IHX to produce superheated steam, while the remainder power is taken for electricity production. The concept of the HTE section is to have 12.5 kW(e) 500-cell stacks with 40 stacks to be composed to a module. Eight modules would occupy a size that fits a trailer. For a full-scale power plant with four 600 MWt MHRs, some 300 of these 8-module trailer units (each 4 MWe) would be needed.

In the S-I cycle based H₂-MHR (see Figs. 3-60 and 3-61), most of the heat is provided through the IHX to the thermochemical cycle at temperatures > 800°C, whereas the remaining heat is used for electricity production to operate pumps and compressors on the H₂ production side. The primary helium loop and H₂SO₄ decomposition unit will be operated at a 7 MPa system pressure, the secondary helium loop at a slightly higher pressure. The option to run the H₂SO₄ and HI decomposition reactions in series is favored to the option of parallel operation. With respect to the HI decomposition, it is foreseen that reactive distillation will be performed in the long run. Overall efficiency is expected to be 45 %.

3.7.6.2. The Molten-Salt Cooled Advanced High-Temperature Reactor

A new reactor concept, the “Advanced High Temperature Reactor”, AHTR, has been suggested in [Forsberg 2004] incorporating essential features of the “classical” HTGR such as the coated particle fuel, coolant outlet temperatures between 700-1000°C, or passive safety systems. The reference design (see Fig. 3-62) considers a large-size annular core of 2400 MWt with a power density of 8.3 MW/m³ and cooled by a liquid fluoride salt at near atmospheric pressure. The large power size which could be selected up to 4000 MWt represents the main difference to a gas-cooled reactor whose decay heat removal capability limits its size to ~ 600 MWt. In case of a loss-of-forced-convection accident, a liquid coolant can transport a much larger quantity of heat by natural convection limiting the maximum fuel temperature to an estimated 1160°C in such a case. Another difference is the small difference of the coolant between core inlet and outlet of approximately 100°C. Heat is transferred via an IHX to a secondary circuit which also uses a liquid salt coolant, before it is utilized for electricity or hydrogen generation.

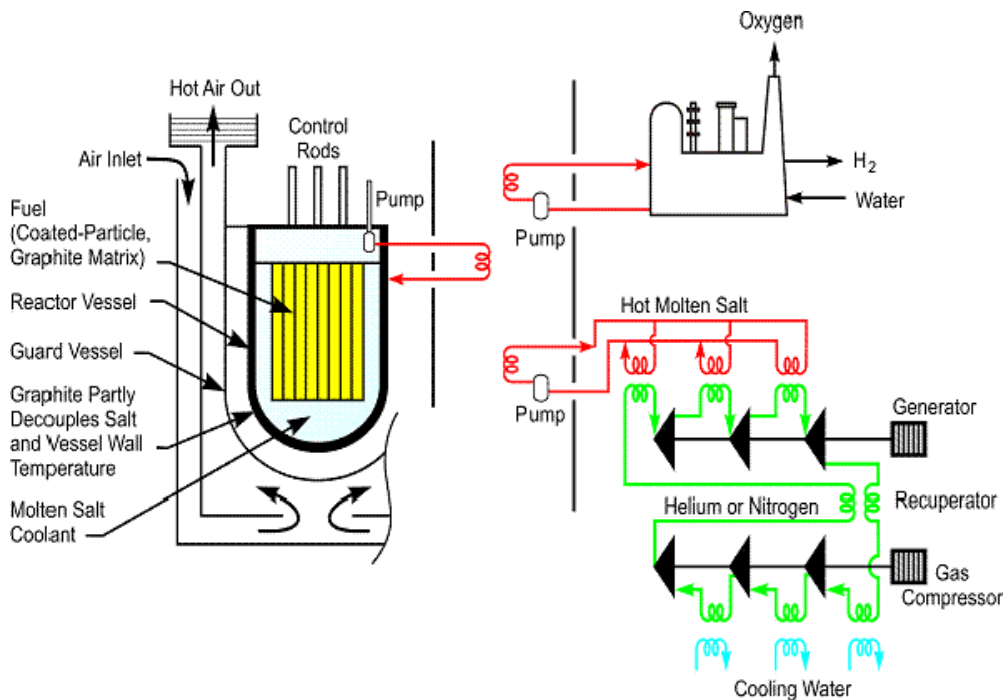
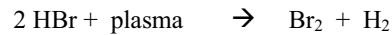


Fig. 3-62: Schematic of the AHTR for electricity production [Forsberg 2004]

3.7.6.2. The STAR-H2 Reactor

Another project as part of the US-DOE NERI is the “Secure Transportable Autonomous Reactor Hydrogen”, STAR-H2, project (see Fig. 3-63). It is designed as a heavy liquid metal cooled, mixed U-TRU-nitride fueled fast reactor with a power of 400 MWt supplying heat at max. 800°C, and with passive safety features. The primary coolant, lead, circulates by natural convection and transfers its heat to a molten salt coolant (FLiBe = containing F, Li, Be) in a low-pressure intermediate circuit, which then transfers heat to a hydrogen production system based on a variant of the UT-3 thermochemical cycle. It operates at atmospheric pressure employing a single-stage HBr dissociation step.



This production process rejects heat at ~ 600°C which – plus some heat from the flibe – is used for electricity production in a supercritical CO₂ Brayton cycle. Rejected heat from the Brayton cycle (< 125°C) finally can be used in an MED desalination plant with a capacity of 8000 m³/d of potable water [Wade 2003]. The operation of a liquid metal reactor above 600°C poses extensive material problem to be solved.

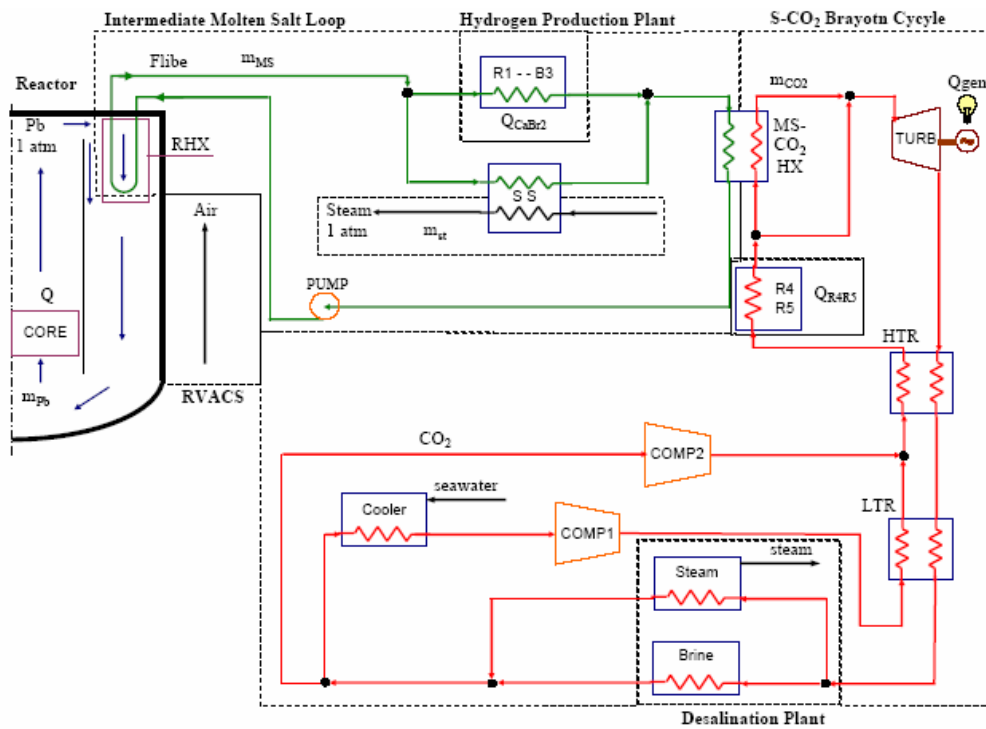


Fig. 3-63: STAR-H2 [Moisseytsev 2003]

3.8. Outlook

It was indicated already previously that for the selected H₂ production processes, the requirements related to the nuclear heat source are different. Table 3-15 shows an assessment on which of the processes could be realized by 2015; Table 3-16 summarizes the main technical data for different H₂ production technologies. Both for the reactor and for the SR and for the IHX, the basic technical fundamentals are available to build large components for commercial deployment until 2015. The extrapolation from 10 MW components to 100 MW in case of steam reformer and 170 MW in case of IHX includes some risks. However, the main questions have been solved by successful operation of 1:1 scale reformer tubes, the 1:1 scale hot gas header for the IHX and the specific design of IHX bundle and other components of IHX-cycle.

Table 3-15: Some aspects for comparison and evaluation of processes of nuclear hydrogen production

Process	Necessary power / output	Reference / status	Character. data of reference	Aspects of extrapolation	Evaluation
HTGR as heat source	300 MW or 2 x 170 MW	AVR HTTR	T _{He} = 950°C	Power, flow direction, hot gas ducts, valves	Possible by 2015
SR	~ 100 MW	EVA-I plant EVA-II plant JAEA mock-up	1-10 MW T _{He} = 950°C 30 tubes	Diameter of bundle (3 m)	Possible by 2015
IHX	170 MW	KVK HTTR	10 MW T _{He} = 950°C	Diameter of bundle (2.5 m)	Possible by 2015
Conventional electrolysis	10 ⁵ Nm ³ H ₂ /h	Alkaline process, diff. concepts	3*10 ⁵ Nm ³ H ₂ /h	Modular arrangement	Possible today
HTE	10 ⁵ Nm ³ H ₂ /h	Small-scale laboratory experiments	< 1 Nm ³ H ₂ /h	Materials, engineering	Possible after 2015 (?)
S-I cycle	10 ⁵ Nm ³ H ₂ /h	Small-scale laboratory experiments	< 1 Nm ³ H ₂ /h	Materials, process techn. engineering	Possible after 2015 (?)

Table 3-16: Main technical data for comparison of processes of hydrogen production

Aspect	Conventional electrolysis	High temperature electrolysis	Steam-methane reforming (no IHX)	Steam-methane reforming (with IHX)	S-I thermo-chem. water splitting (with IHX)
Thermal power of reactor [MW]	300	2 x 170	2 x 170	2 x 170	2 x 170
Process behind reactor	GT	GT	SR + SG	IHX	IHX
Process of hydrogen production	low- temp. electrolysis	HTE	SMR	SMR	S-I cycle
Max. He temperature from reactor [°C]	700 - 900	900 - 1000	900 - 950	950	950 - 1000
Max. process temperature for H ₂ prod. [°C]	80	800 - 900	750 - 800	750 - 800	850
Max. process pressure for H ₂ prod. [MPa]	3	4	4	4	4
Input of fossil energy [Nm ³ CH ₄ / Nm ³ H ₂]	-	-	0.25	0.25	-
Efficiency of electricity production [%]	35	45	-	-	-
Energy requirement for H ₂ production [kWh(e) / Nm ³ H ₂]	4.5 - 4	2.6 - 3	-	-	-
Energy requirement for H ₂ production [kWh(th) / Nm ³ H ₂]	-	2	6.8	6.8	7
H ₂ production [Nm ³ /h]	3.1*10 ⁴	4*10 ⁴	2*10 ⁵	~ 2*10 ⁵	~ 2*10 ⁵

Conventional electrolysis could be operated with the steam turbine process already, whereas for the hot electrolysis reactors with 950-1000°C, it will be necessary to provide the hot steam. Processes using steam reformers can be heated with helium temperatures between 900 and 950°C effectively related to heat fluxes of about 60 kW/m². If an IHX is used for this application, a higher temperature of the helium by at least 50 (up to 100) °C is preferable to get similar areas for heat transfer.

Thermochemical cycles for water splitting need very high helium temperatures at the reactor outlet because an IHX will be necessary for separation of the nuclear reactor and the hydrogen production process. Due to the temperature drop across the IHX, the reactor outlet temperature should be at least about 950-1000°C.

For the HTE, a broad R&D program is necessary so far as the process technology itself, the materials and the design and testing of larger plants is considered. Although this technology may

inherently benefit from the development of high temperature fuel cells, it still remains a longer term option, and it is doubtful if all open questions can be answered before 2015.

Thermochemical cycles for hydrogen production need a further period of development too. Again the process technology, the materials, the design and testing of larger plants are required before all serious answers on the prospects of such processes really can be given. In any case, the time schedule for this development is relatively long and the chance that these processes can be applied on an industrial basis before 2015 is very low.

The overall evaluation of the alternative processes is as follows:

- conventional electrolysis with electrical energy from gas-turbine HTGR: until 2010
- steam reforming of methane with or without IHX: until 2015
- high temperature electrolysis: after 2015
- thermochemical cycles: after 2015

4. SAFETY ISSUES AND REGULATORY FRAMEWORK

4.1. Safety Considerations in a Process Heat HTGR

4.1.1. Identification of Hazard Sources

Safety items can be categorized into several classes. The items associated with the accidental release of a large amount of radioactive materials and core damage from thermal turbulence are categorized into the class with the largest hazards. In relation to these items, the system must be designed with high reliability and redundancy to avoid the loss of safety functions. On the other hand, the items associated with continuous normal operation are categorized into the class with lowest hazards, for which such a high level of reliability and redundancy is not required.

The hydrogen production system connected to a nuclear plant will most probably not be designed as a nuclear grade system. Therefore, particular safety items other than the “conventional” safety features will not be provided in the chemical part of the combined system. Much of the following has been evaluated for the Japanese HTTR to be connected to a H₂ production system based on steam reforming of natural gas. Many aspects, however, will be similarly applicable to other nuclear production methods for hydrogen. There are three areas of concern associated with the connection of a hydrogen production system to a nuclear heat source:

- Hydrogen production system is the final heat sink for the nuclear reactor;
- Flammable substances are present in the system;
- The product hydrogen is handled outside the nuclear plant.

Potential hazardous events in connection with the steam reforming system are

- Tritium transportation from the core to the product hydrogen and methanol;
- Thermal turbulence induced by problems in the chemical system;
- Fire and explosion of flammable mixtures with the process gases.

The IHX structure forming the boundary between primary and secondary helium shall withstand a creep buckling load as the result of an accidental pipe rupture within the secondary helium piping system. This accident is considered the severest to these structures.

Maximum allowable change rates for the reactor coolant temperature under normal operation depend upon the temperatures of the reactor coolant and the metal structures in contact with the coolant. An acceptable stress is limited to the lower level at higher temperatures due to creep damage on the structures. Based on a parametric analysis for the HTTR on the structural integrity of the IHX hot header and reducer, the maximum allowable change rate for the coolant temperature is limited to 15 °C/h at temperatures ≥ 650°C.

A safety related issue is the operability of the production process system during a nuclear reactor scram. According to the actual safety regulations, the ultimate heat sink of an NPP is limited to water and/or air, and cannot be electricity or chemical energy as the result of a conversion process. Therefore the production process system is not designed to take over safety functions for the nuclear system; these are exclusively left to the reactor cooling system.

In case of a scram of the HTTR, the power output immediately falls to such a low level that the reactor safely shuts down and remains in a sub-critical state. Concerning the steam reforming system, the abrupt cut in heat input is usually followed by an instantaneous disconnection of the feedstock supply and filling of the steam reforming loop with nitrogen to prevent carbon deposition on the catalyst. Also the large heat capacity of the catalyst allows for a reduced rate of temperature decrease [Hada 1994].

4.1.2. Tritium Transportation

Before hydrogen or any other product from a nuclear process will be usable as a normal commodity, it must have a tritium contamination below the tolerated limits specified by the national legislation. Therefore, one safety requirement is to correspondingly reduce tritium penetration into the products. In this case, the required safety items are not directly related to the reactor safety and thus can be classified into the lowest safety level.

Tritium is produced in the reactor core of an HTGR during normal operation as a ternary fission product [U-235 (n,f) T] and by activation reactions of lithium [Li-6 (n, α) T] and boron [B-10 (n, 2α) T] in the graphite components and control rods. In addition, the He-3 fraction in the helium coolant [He-3 (n,p) T] is a significant tritium source in the HTGR. The tritium produced in a fission process will be retained within the fuel particles, which have an intact coating; only a small fraction originating from fuel particles with a broken coating or from uranium contamination of the core graphite is expected to escape into the coolant. On the other hand, tritium produced in the graphite can rapidly diffuse through the graphite components into the coolant. Most impurities including tritium in the coolant are removed by the helium purification systems. There is, however, a small amount of tritium that can be transported to the process side by permeation through the heat exchanger tubes into the products hydrogen or methanol.

For the example of the 170 MWt process heat HTR-MODUL, the reactor internal tritium production and release into the helium coolant was assessed in a conservative way as summarized in Table 4-1 [Eichler 1985].

Table 4-1: Tritium production and release for the 170 MWt process heat HTR-MODUL

Tritium source	Tritium production rate [10 ³ Bq/s] ([%])		Tritium release rate into coolant [10 ³ Bq/s] ([%])	
	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)

Under the operating conditions of a process heat HTGR with its high coolant exit temperatures, hydrogen and tritium are highly mobile resulting in permeation through the walls of heat exchanging components.

There are three approaches to reduce the tritium concentration in the products:

1. Oxide layers on the heat exchanging surfaces were found to effectively decrease the amount of tritium permeating, whereas the permeability is high for clean surfaces. Under steam reforming conditions, an oxide layer will rapidly develop on the tube surface.
2. A gas purification system can be an effective and powerful means to remove tritium from the primary circuit. Additional components could serve as getter materials for H₂ and T. For the HTTR, however, the T is not completely removed from the circuit in the coolant purification system, since only a small fraction of the coolant is bypassed through the purification system.

3. An intermediate circuit purified by a sweep gas flow serves as an additional boundary between primary and secondary circuit. If oxygen or steam are added to the sweep gas, part of the T could be bound as tritiated water HTO and would no longer be available for permeation.

Another possible path for a transition of radionuclides into the product gas is by leakage in the heat exchangers. A leakage of radioactivity from the primary to the secondary circuit requires a pressure drop from the primary towards the secondary loop, in contrast to the plant's condition during normal operation. A measure to prevent a major risk is the installation of gas supervising systems to trigger the disconnection of the product gas lines from the grid.

In the opposite direction, hydrogen produced on the process side can permeate through the tube walls into the primary circuit causing corrosion reaction with the graphite structures. Additionally, by transport of carbon in helium circuits, carbon deposition surfaces of high temperature alloys could result in changes of material properties. Therefore, measurements of hydrogen permeation rates have been carried out, dependent on wall temperature, type of material, wall thickness and partial pressure of hydrogen. Especially the influence of steam is important, because a layer of oxide is formed on the surfaces of the metallic walls on the process side.

4.1.2.1. Activity in Germany

Within the frame of the PNP project in Germany, experimental investigations were made on the permeation process in high temperature alloys. Test facilities allowed both long-term (1000-3000 h) at temperatures up to 1000°C and pressures up to 3.2 MPa (Fig. 4-1). Short-term analyses were used for pre-selection of materials. Results have shown that in-situ oxide layers show a large inhibition of permeation at temperatures above 650°C. Still the uncertainty is relatively large at lower temperatures and also if looking at respectively measurements from operated HTGRs.

The permeation rates of tritium through the walls of high temperature heat exchangers have been measured as a function of temperature, type of materials and the process conditions of steam reformer, steam generator, and intermediate heat exchanger. Selective filter systems to take up tritium from helium circuits have been developed, which consist of hydride forming materials such as Ti, Ce, or Zr or cerium mixed metals. Oxide layers on the heat exchanger surfaces were found to reduce the tritium transport through the walls significantly. In-situ oxide layers have shown a large inhibition of permeation in the temperature range of interest ($T > 600^\circ\text{C}$).

By means of a gas purification system in the IHX cycle of the PNP reactor, the release rate of the process was estimated to be less than 0.2 GBq ($5 \cdot 10^{-3}$ Ci) per MWt. In connection with the yearly hydrogen production of the plant, it results in a tritium contamination of less than 0.37 Bq (10 pCi)/g of H_2 . This was assumed to be tolerable in the PNP-project in comparison to other allowed levels of radioactive contamination.

For the H_2 transportation process in the opposite direction, as a typical result, around 50 ml /($\text{m}^2 \text{ h}$) of H_2 were measured at 900°C for typical steam reformer applications (see Fig. 4-2). These quantities of hydrogen could be easily removed by the gas purification plant. Of course, the stability of oxide layers during transients and other loads have to be considered and may influence the data.

In summary and as a result of the PNP-project, it was found that hydrogen permeation for steam reformer application represents a problem that can be solved. For processes using an IHX the permeated quantities of hydrogen can be reduced even more. As long as the heat exchangers remain intact, even under conservative assumptions, the expected radioactivity on the secondary side remains small compared with the tritium concentration by permeation [PNP 1981].



Fig. 4-1: Test facility for measurements of H₂ permeation (source: FZJ)

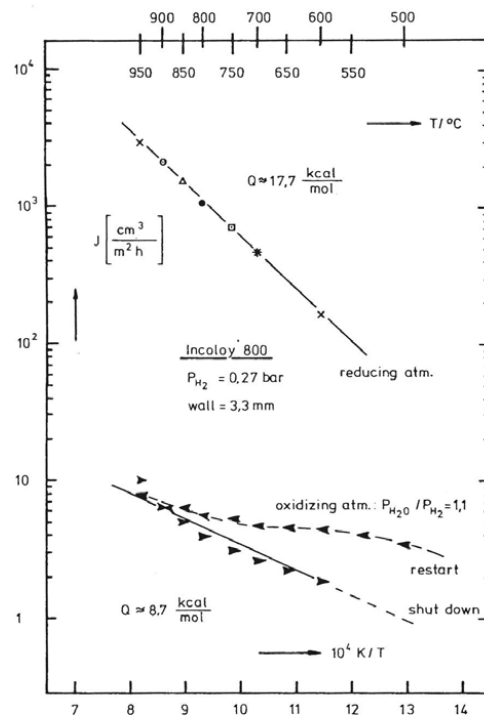


Fig. 4-2: Arrhenius diagram of H₂ permeation for clean and oxidized surfaces [Röhrig 1975]

According to the German Preventive Radiation Protection Ordinance, neither licensing nor announcement is required for the use of fossil products refined by nuclear process heat, whose tritium content does not exceed 5 Bq/g. This special case is the exception from the rule, where for any fabricated product, the specific radioactivity limit is lower by a factor of 10 compared to the above figure, i.e., 500 mBq/g [German. Fed. Gov. 1989]. The background for this special rule resulting from discussions in the context of the PNP project is the fact that, depending on the origin of the feed natural gas, the natural activity content would often have reached already the free limits given by the law.

4.1.2.2. Activity in Japan

For the case of the HTTR in combination with a steam reforming system, the flow paths of the tritium ("HT") and hydrogen ("H₂") have been identified as shown in Fig. 4-3.

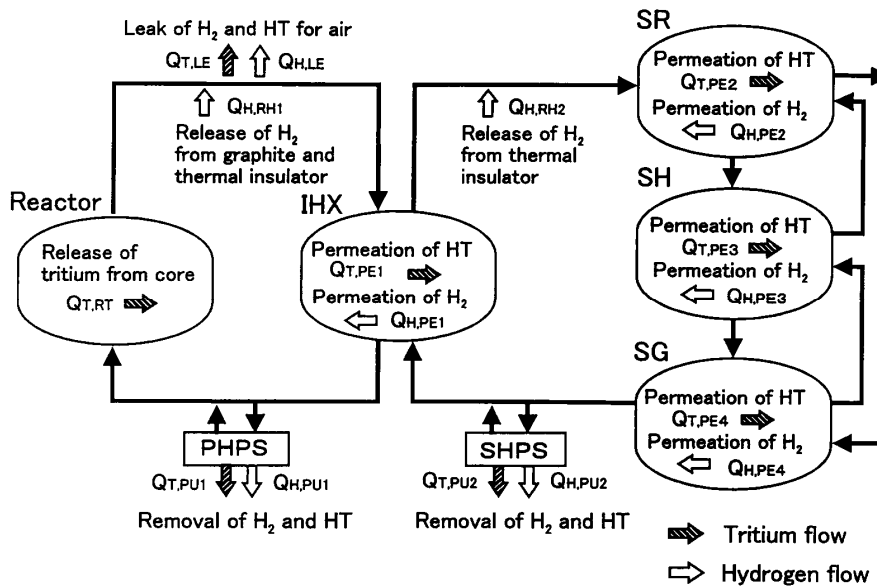


Fig. 4-3: Tritium ("HT") and hydrogen balance in HTGR H₂ production system

Tritium removal from the helium coolant is only partially effective, since in the HTTR, the purification systems are installed in the bypass loop and their flow rates are selected based on the concentrations of other impurities.

Preliminary calculations have been performed to determine tritium concentration at steady state for the HTTR steam reforming system. These results show that the combination of the self-grown oxide layer effect and a reasonable flow rate through the helium purification systems are sufficiently effective to restrict the tritium concentration in the product gas to an acceptable level.

Results have been acquired in a small-scale apparatus using test pipes made of the high temperature alloys Hastelloy-X and Hastelloy-XR, the designated materials for IHX heat exchanger tubes and SR pipes. Test tube dimensions were 1000 mm in length, 31.8 mm outer diameter, and 3.5 mm wall thickness. Test conditions were tube temperatures of 600-850°C and H₂ partial

pressures of 0.1-4 kPa (or vol%) in helium gas. Flow rate was 0.1 Nl/min of the mixture gas. The test series also confirmed the phenomenon of oxide layer formation which reduces the permeation rates [Takeda 1999].

In another experimental series, the effect of counter-permeation of deuterium (to simulate tritium) and hydrogen, i.e., the influence on permeation if hydrogen is present at the outside, was investigated. The test tube here consisted of Inconel 600 with an inner diameter of 7 mm and a thickness of 1.2 mm. Mixtures of argon with hydrogen and deuterium, respectively, were flown at constant rate and constant pressure through the test pipe and the outer so-called measurement tube (inner diameter: 50 mm). With deuterium flowing inside and hydrogen outside the test tube, it was found that for low partial pressures of deuterium < 100 Pa, its permeation rate to the outside is decreasing, if the hydrogen partial pressure is > 10 kPa. This is due to the fact that the dissolved H atoms are saturated on the surface. For the real system of HTTR combined with SMR, hydrogen partial pressures in the catalyst pipes will be about 2 MPa, therefore a comparatively low amount of tritium is expected to be transferred from the primary circuit to the hydrogen production system [Takeda 2004].

The reduction of permeability at the presence of an oxide film layer was again confirmed, the observed factor of 100-1000 enhancing with time. The ratio of hydrogen over deuterium permeability was measured to be 1.32 at 670°C; it decreases with increasing temperatures.

In the meantime, first experience has been gained from the HTTR operation at 950°C with regard to hydrogen behaviour in the core [Sakaba 2005a]. In this case, the sources of hydrogen are in the primary circuit the oxidation processes of moisture in the graphitic core, in the secondary circuit water liberated from insulation material. Since the latter produced a higher water concentration, the H_2 transport was actually from the secondary to the primary circuit. Fig. 4-4 shows the H_2 and H_2O behaviour during the first 950°C operation period of the HTTR.

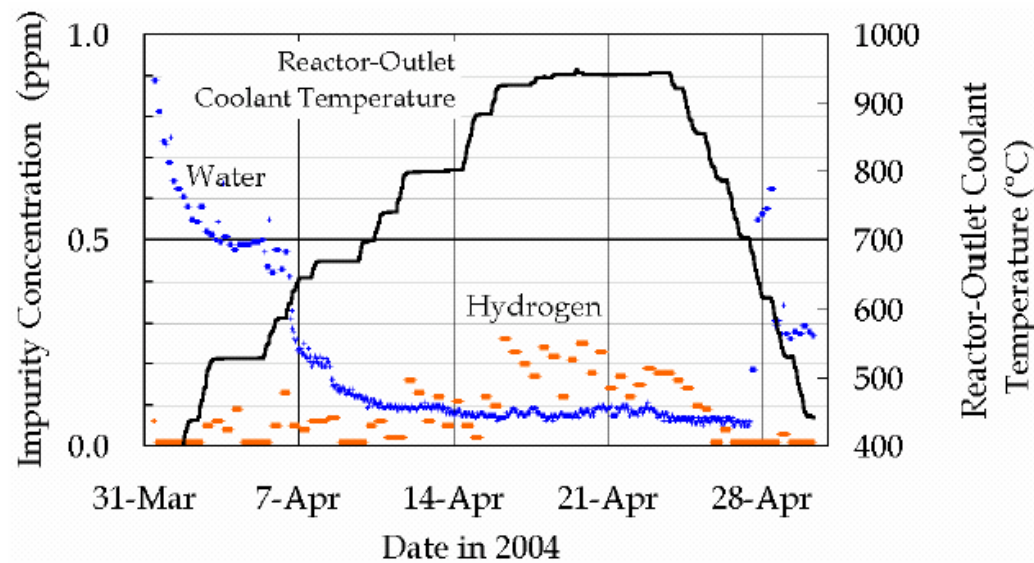


Fig. 4-4: Hydrogen and water behaviour in the primary cooling system during the first operation of HTTR at 950°C [Sakaba 2005a]

Assuming an Arrhenius-type relationship for the permeability and taking the data of activation energy and pre-exponential factor for high temperature alloys from the literature, the hydrogen permeability through Hastelloy XR was assessed as shown in Fig. 4-5. The new data indicate a somewhat lower permeability compared to previous works presumably due to formation of an oxide layer on the IHX tube surfaces [Sakaba 2005a].

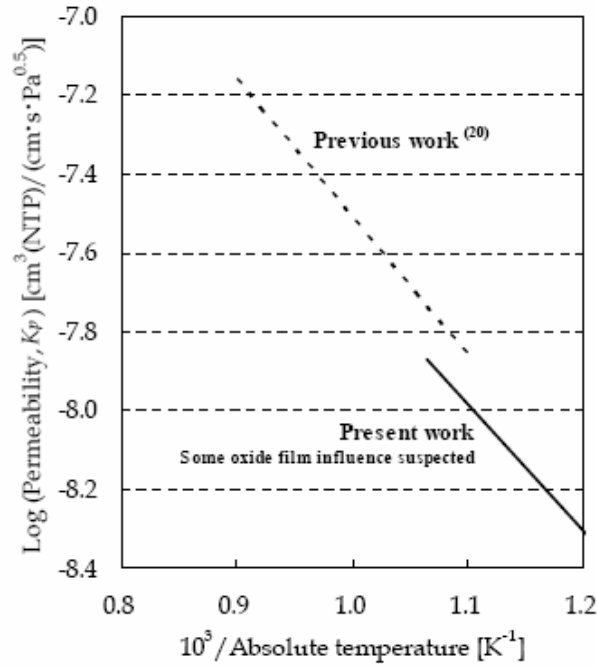


Fig. 4-5: Comparison of present and previous work [Takeda 1999] of hydrogen permeation in Hastelloy XR [Sakaba 2005a]

4.1.2.3. Activity in the USA

In the USA, the TRITGO computer model is being used for calculating tritium transport behaviour from its origin in the reactor core to the product hydrogen. Sources considered are the He-3 in the coolant, Li and B impurities in the graphite, and ternary fission. Removal mechanisms are radioactive decay, He purification, and sorption on graphite.

The assessment of tritium contamination was made for the Heatric-type IHX as part of the H2-MHR concept (see chapter 3.2.4). Assuming a linear temperature profile from 1000°C to 636°C of the IHX wall and a tritium concentration of 3.7×10^5 Bq/m³ (10 µCi/m³) in the primary coolant, the tritium permeation rate was calculated to be approximately 11×10^5 Bq/s. Assuming further that all tritium transferred will go into the product H₂, the contamination of the H₂ gas would amount to 0.8×10^5 Bq/Nm³ (or ~ 890 Bq/g), which is about one order of magnitude higher than the maximum limit specified in the US regulations. A more thorough analysis will be conducted, when a more detailed design of the total plant is available [NERI 2003].

4.1.3. Thermal Turbulences in the Steam Reforming System

A system with an endothermal chemical reactor connected to an HTGR exhibits thermal dynamics, which differ significantly from those of the nuclear reactor itself. In the Japanese HTTR core, the coolant temperature is primarily controlled by the reactor power, not by the coolant flow rate. Nuclear heat is transferred to the helium gas with the result of a linear relationship between reactor power and helium temperature. On the other hand, in the chemical reactor where endothermal reactions take place, the heat input necessary to cause the reaction tremendously increases with increasing reaction temperature due to the Arrhenius type temperature dependence of the reaction rate. The development of a new control technology is required in order to balance the difference in the thermal dynamics between the nuclear and chemical reactor. For the HTTR, the selected design and arrangement of the steam generator is expected to fulfill this control function.

The operating procedures for startup and shutdown are similar, but reversed. Before startup, nitrogen is supplied at a pressure of 2.2 MPa. The HTTR is then started. When the secondary helium gas is heated to above 500°C and the steam generator is controlled at the rated pressure of 5.0 MPa, steam is gradually supplied to the system and nitrogen is released into the environment with this steam by switching the flow line. With the steam flow rate constant at rated conditions and the helium gas temperature at the inlet of the steam reformer increased to 700°C, methane feed gas is supplied to the system. Even during low startup system operation, a stepwise increase in the feed flow rate by 10 % (as it is difficult to control the feed gas at low flow rate levels), results in a stable helium gas temperature level at the inlet of IHX due to the influence of the steam generator. After 60 h, the helium gas temperature reaches 950°C and the entire system can be operated automatically.

A higher probability of malfunction or failure is expected on the process side rather than on the power generation side. Safety measures are required to mitigate potential disturbances resulting from a malfunction or failure in the hydrogen production system to allow for a continuous reactor operation without reactor scram.

If the methane supply system is shut down due to a loss of electric power or a malfunction of the control system, the helium gas temperature at the steam reformer outlet will increase. The steam generator installed downstream of the steam reformer in the secondary loop (see Fig. 4-6), can cool down the hot helium gas to the saturation temperature of the steam, thus providing a stable controllability for any disturbance at the steam reformer due to the large heat sink capacity and preventing a reactor scram. However, if the feed water supply is interrupted, the steam generator cannot continue to operate. It is therefore proposed to re-use the generated steam as feed water after condensation in the radiator.

Fig. 4-6: Arrangement of steam generator (SG) in HTTR circuits [Ohashi 2005]

In order to prevent a scram of the HTTR due to a loss of feed water, the hot helium gas is cooled by the steam generator and the generated steam from the steam generator flows into a natural ventilation type radiator connected to the steam generator. The condensed water is then supplied to the steam generator as feed water. The steam generator can keep its water contents for normal operation. The total heat capacity is about 8.8 MW representing the heat capacity of both the steam reformer, the super heater and the steam generator. Furthermore, if a pressure drop is detected in the cooling system due to pipe failure or valve malfunction, and the water level in the steam generator is low due to interruption of the feed water flow, the valve in the steam line closes, while that in the radiator steam supply line opens passively through an automatic air supply system. Generated steam is supplied to the natural convection type radiator and is cooled down. Condensed water is recycled to the steam generator as feed water. This system does not require electric power nor feed water.

The steam reforming system is a ternary cooling system. A change of the flow rate of either the feed gas or the water to the steam reformer induces a thermal disturbance of the helium outlet temperature on the reformer due to the change of the amount of heat input for the reforming reaction. If the temperature of the helium returning to the IHX exceeds the allowable limit, the reactor will scram.

Static calculations of the cooling ability of the steam generator have been carried out showing that a reduction of the feed gas changes the outlet temperature of the reformer correspondingly. But the steam generator mitigates the temperature variation within 5°C. The continuous cooling of the hot helium gas by the steam generator allows the HTTR steam reforming system to continue at normal operation. A transient analysis assuming a stepwise decrease in process gas flow rate by 20 %, indicated that an increased heat input to the steam generator due to increasing helium inlet temperature only results in an increase in steam quality at saturation temperature due to boiling, but not in an increase of the steam temperature.

The function of the ternary cooling system is to remove heat from the core during normal operation. Since the reliability required for this system is not particularly high, problems may occur more often during operation lifetime.

The safety design of the nuclear plant is based on the defence-in-depth concept. Therefore in case of a reactor scram, the propagation of thermal turbulences should be stopped in the secondary loop. The safety requirement for this event is to limit the secondary helium temperature variation within ± 15 K at the inlet of the IHX to prevent a reactor scram.

Performance tests have been conducted between October 2001 and February 2002 to check the ability of hydrogen production and controllability of the experimental facility. The test of the system controllability was made to investigate the transient behavior of the gas flows in case of a loss of chemical reaction, i.e., the disconnection from the methane feed [Ohashi 2004].

As illustrated in Fig. 4-7, nominal flow rates were 12 g/s of methane and 47 g/s of steam, when – at time 0 – methane feed, and thus hydrogen production, was shut off. The helium temperature at the SR outlet increased from 611°C and stabilized at around 800°C after 1.3 h. At the same time, the temperature at the inlet was raised from 531 to 762°C. Helium temperatures at the steam generator outlet were observed to fluctuate not more than within the range of -5.5 to +4.0 K, which is within the specified range of -10 to +10 K required for HTTR/SR operation.

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Fig. 4-7: Flow rates of methane, steam, nitrogen, and production rate of hydrogen during the system controllability test [Ohashi 2005]

4.1.4. Hazards in Electrolyzers

The risks of (alkali) electrolysis plants are given by [Bongartz 1996]

- a potential explosion of the hydrogen contained in the system;
- a leakage of electrolyte;
- a failure of the cooling water system possible connected with a mixing of H₂ and O₂;
- corrosion of parts of components with potential release and mixing of H₂ and O₂;
- mud formation in the gas piping and subsequent change of operational behaviour (e.g., increased operation temperature leading to enhanced corrosion);
- damaging of H₂ containing pipelines.

Initiating events are typically given by, e.g., component failure due to corrosion, a sudden disconnection from electricity supply where downstream components may not be shut down right in time, or human error (e.g., confusion of cell polarity).

4.1.5. Hazards in Thermochemical Cycles

The hazardous potential in thermochemical cycles is given by the H_2 and O_2 produced and by the presence of the chemical compounds during the cycle. All chemicals, interim products as well as the products from side reactions are mainly characterized by their corrosiveness and toxicity. Corrosiveness is particularly enhanced by the given high operation temperatures and pressures. To ensure safety and health of workers and protect the environment, a safety concept must concentrate on minimizing the quantities released in case of inadvertent pipe ruptures, system spills or leaks.

Multiple step thermochemical cycles produce hydrogen and oxygen in separate reactions, which can be realized at different locations, thus isolating the H_2 containing systems and reducing the chance for explosions. Lower-temperature thermochemical cycles operate at temperatures lower than the autoignition temperature of hydrogen. In case of an air ingress, no clean operation can be achieved.

4.1.6. Explosion Hazards

Fire and explosion hazards resulting from the leakage of flammable materials such as methane, hydrogen, and carbon monoxide should be considered because they have the potential of causing significant damage to safety components. Therefore these components should be designed against fire and explosion according to the highest safety level.

General safety requirements against fire and explosion hazards have been established in the IAEA SAFETY SERIES, according to which the amount of flammable materials in the plant and in the vicinity of the plant should be reduced to a minimum possible as a precaution measure. In the HTTR steam reforming system, however, the HTTR containment building does not have the capacity to withstand severe radiation heat and blast overpressure. Therefore it is necessary to minimize the risk of a huge fire or explosion event. One possibility is the separation between the accident source and the HTTR, which may be given by a safety distance or a fire-proof separation wall.

4.1.6.1. Release of Flammable Gases into the Containment Building

Fire and explosion events inside the reactor building may cause severe damage to nuclear safety systems. It is therefore required that the possibility of a flammable gas leak inside the reactor building should be low enough to avoid any fire and/or explosion at this location. The potential sequence of flammable gas ingress into the reactor building is the simultaneous failure of a secondary helium pipe inside the containment and a reformer tube in the H_2 production plant outside the containment (see Fig. 4-8). The only cause of such a failure of these components is conceived to be an earthquake. Therefore they are designed for a high seismic safety level.

Since the steam reforming system will be laid out as a non-nuclear facility, it is basically not equipped with additional safety systems. In case of an explosion event in the vicinity of the reactor building, the thermal load and blast overpressure may be strong enough to cause some damage to the containment. Therefore, it is required to prevent significant leakage of flammable gases in the vicinity of the reactor building. A double tube has been adopted in the HTTR steam reforming system to prevent leakage of flammable gas. Emergency shut-off valves are also provided to isolate the failure location of the pipe and, thereby, limiting the amount of leakage.

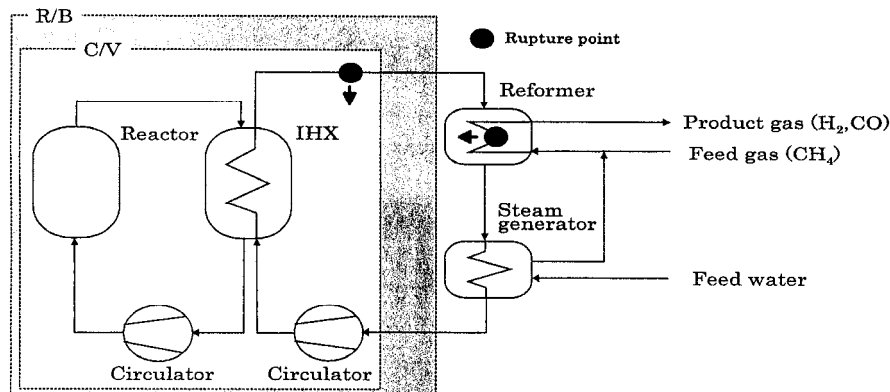


Fig. 4-8: Ingress of flammable gases into the reactor containment

4.1.6.2. Release of Flammable Gases outside the Containment Building

The danger of possible detonations or deflagrations of explosive gas clouds, which were released from tank ships or trucks or nearby chemical plants and which might travel towards a reactor building, has been subject of substantial theoretical and experimental investigation. This kind of threat is strongly dependent on the local environment and the future development plans for that area. Individual characteristics of the site and also the layout of reactor buildings will be relevant. Considerable basic aspects would be to avoid large flat surfaces, to surround the reactor building with auxiliary equipment buildings, to use underground construction for important connecting conduits.

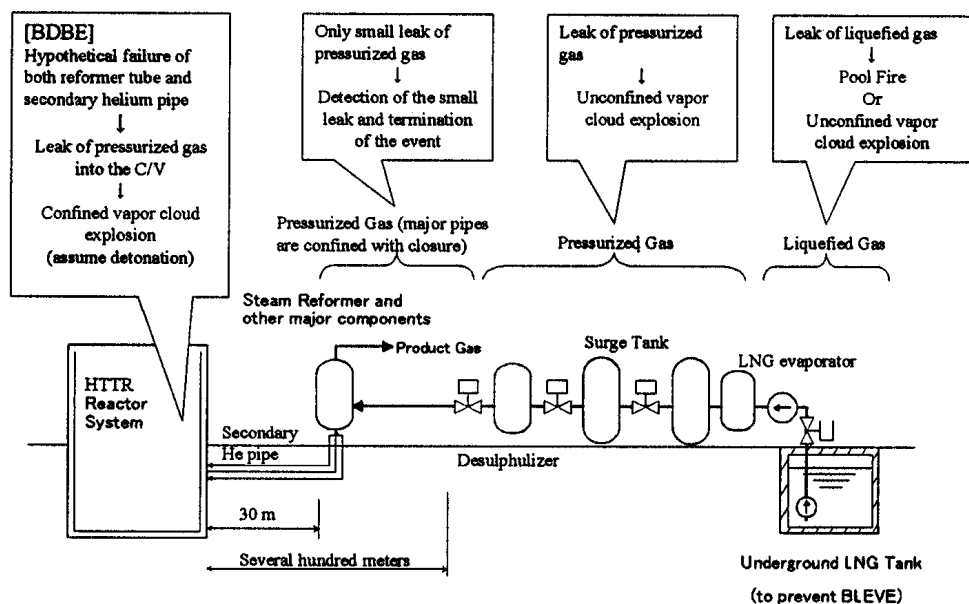


Fig. 4-9: Possible effects of fire/explosion accidents outside the reactor containment

Fig. 4-9 shows for the example of HTTR plus steam reforming system the potential fire and explosion hazards which may have an impact on the reactor containment. A safety distance can be selected to mitigate the effects of thermal load from fire and blast overpressure from explosion. Comparing these effects, the explosion pressure wave causes a greater damage than the fire itself. The explosion event is taken into account to estimate adequate safety distances.

A formula for the safety distance is generally acknowledged to have the form

$$R = k * M^{1/3}$$

where R is the safety distance in meters and M the mass of the flammable substance in kg. The relation may be modified by damping parameters, if some sort of protective measure is applied, e.g., wall or earth coverage. The k-factor depends on the building to be protected (from German recommendations: 2.5 - 8 for working building, 22 for residential building, 200 for no damage) and on the type of substance.

The guideline on the “Protection of Nuclear Power Stations from Shock Waves Arising from Chemical Explosions”, drafted by the German Federal Ministry of the Interior (BMI) in 1976 [BMI 1976] has defined for nuclear power plants a k-factor of 8 in the safety distance relation (see dashed line in Fig. 4-10), meaning that the detonation of a mass M results in a maximum overpressure of 30 kPa in a distance of r. The mass M is either unsaturated hydrocarbons or compressed gas assuming a premixed stoichiometric mixture of hemispherical shape and central ignition. If the flammable mass is a pressurized liquid, k is reduced to 6.3 (or 50 % of the mass). For cryogenic liquids or hydrocarbons under standard conditions, k is 3.7 m/kg^{1/3} (or 10 % of the mass).

With respect to the HTTR with steam reforming system, the k-factor becomes 3.7 (for LNG). If applied to the foreseen 400 m³ LNG tank, the result according to the above equation is a required safety distance of 205 m. Such a safety distance would actually be fulfilled, if the planned distance between reactor building and the LNG tank of at least 300 m be realized. What is not considered here is the flammable content of the steam reformer which is sited in the immediate vicinity of the reactor building and, thus, not in compliance with the BMI guideline which requires a minimum safety distance of 100 m.

The guideline is valid for NPP of present design; it is explicitly mentioned that “no statement can be given at present concerning its application to future nuclear process heat plants”. It is supposed to be a concomitant effort with the development of nuclear process heat plants to solve the problem of external vapor cloud explosions.

In the USA, it is judged according to the US-AEC Regulatory Guide 1.91 [US-NRC 1978] that 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. No additional measures need to be taken, if the equation

$$R = 18 * W^{1/3}$$

is met, where R is the safety distance [m] from an exploding charge and W is the mass of TNT (equivalent) [kg] of the exploding material (see solid line in Fig. 4-10).

For the LNG storage tank of the HTTR/SR system, the 400 m³ of LNG correspond to a TNT equivalent of 1859 tons which then translates into a safety distance of as long as 2.2 km. This approach appears to be unrealistic for the HTTR/SR system considering the fact that much larger stationary LNG tanks up to 200,000 m³ (→ R ≈ 18 km) have been established worldwide. The 1.91 guideline, however, offers additional options such as risk analysis for further reduction of the safety distance.

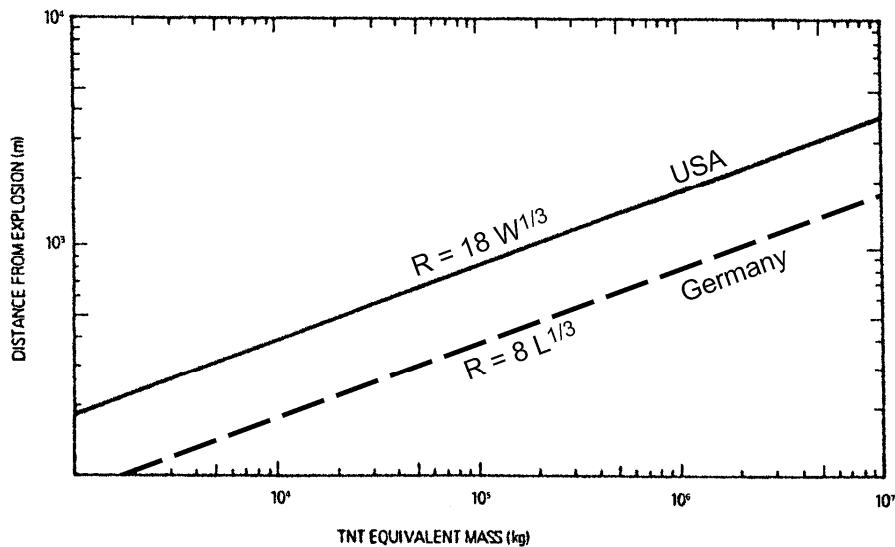


Fig. 4-10: Safety distance as a function of the quantity of released liquefied gas according to the BMI guideline and the US regulatory guide 1.91

In Japan, the “High Pressure Gas Safety Law” which applies also to hydrogen, defines safety distances for chemical plants containing < 10,000 kg of flammable materials – which would apply to the combined system of HTTR plus S-I process – as follows:

- 17.0 m to public buildings;
- 11.3 m to residential buildings;
- 8.0 m to facilities which are operating burners.

Other countermeasures considered are fire wall, berm (separating hill) as shown in Fig. 4-11 for the US H₂-MHR concept and in Fig. 4-12 for the US STAR-H₂ reactor concept, where the reactor modules are located in underground silos while the primary helium circulator, the IHX, the intermediate helium loop circulator and respective piping are located in an adjacent silo.

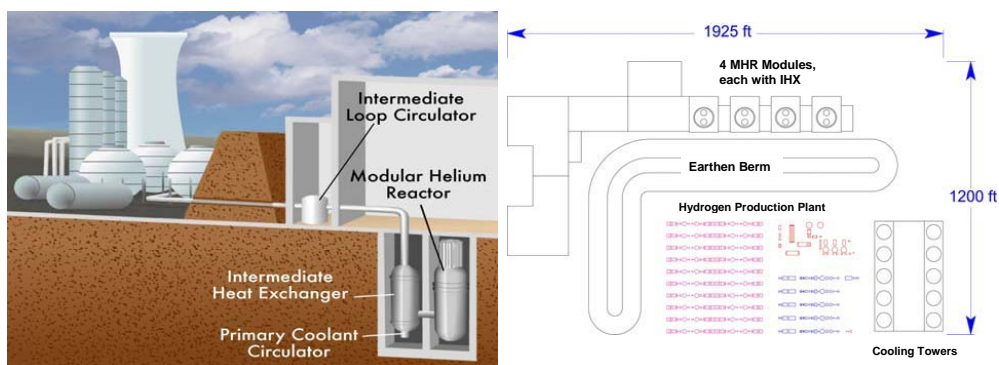


Fig. 4-11: Safe separation of nuclear and chemical section by earthen berm [Richards 2005b]

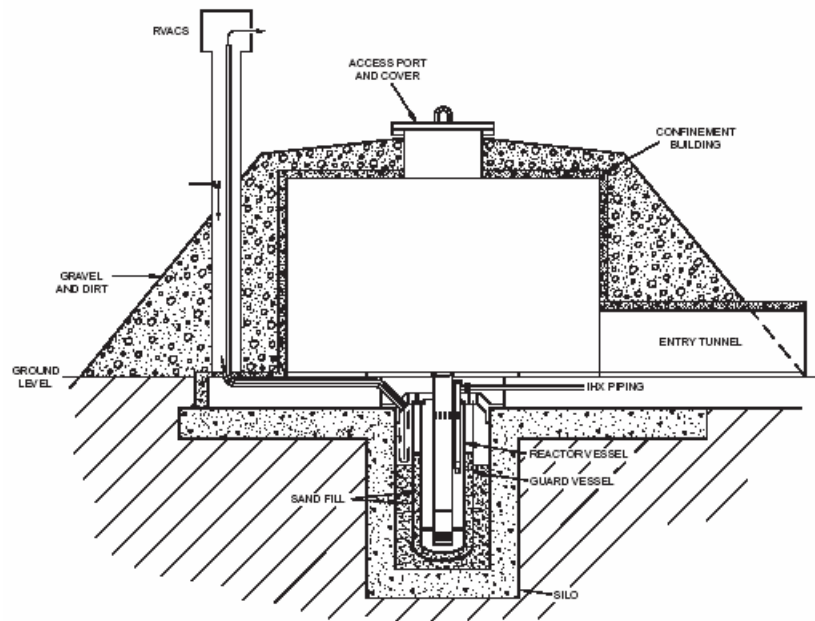


Fig. 4-12: Underground nuclear reactor STAR-H2 (Source: US-DOE)

4.1.6.3. Safety Concept for the HTTR Combined with SMR against Fire and Explosion

The basic safety concept as proposed by JAEA is to provide some safety barriers between the HTTR and the steam reforming system so as to prevent the anticipated operational occurrences for anticipated design basis events related to the steam reforming system. Design basis events related to the HTTR reactor system have already been considered. It is therefore important to discuss additional anticipated design basis events that could originate due to the connection of the steam reforming system with the HTTR. Additional safety design requirements and corresponding countermeasures for the hydrogen production system are given in Table 4-2 for the different operational conditions of the HTTR.

The hydrogen production plant is designed according to the same level of safety as requested in the domestic regulations for conventional chemical plants without any additional safety features. It obeys the defence-in-depth principle with its graduated steps of prevention of occurrence, prevention of propagation, and mitigation of consequences (see also Table 4-2 and Fig. 4-13):

- Prevent leakage and ignition;
- Prevent inflow into nuclear building;
- Detect leakage and disconnect natural gas feed line;
- Define safe distance.

Table 4-2: Safety design requirements for the HTTR hydrogen production system

Operational condition	Event	Safety requirement	Countermeasure
Normal operation	Tritium transport from core to product gas H ₂	Reduction of tritium radiation level in product gas	Restriction of permeation through tube walls Removal of tritium in the coolant by purification system
Anticipated operational occurrence	Thermal turbulence	Prevention of thermal turbulence to propagate to primary He loop	Mitigation of vibration of secondary helium temperature by steam generator installed downstream
Accident	Fire/explosion from leakage of flammable gases	Prevention of leakage inside and in the vicinity of reactor building	Upgrade design category of helium piping and reforming tubes Double-walled tubes Inerting of the compartment
		Mitigation of accident consequences	Safety distance

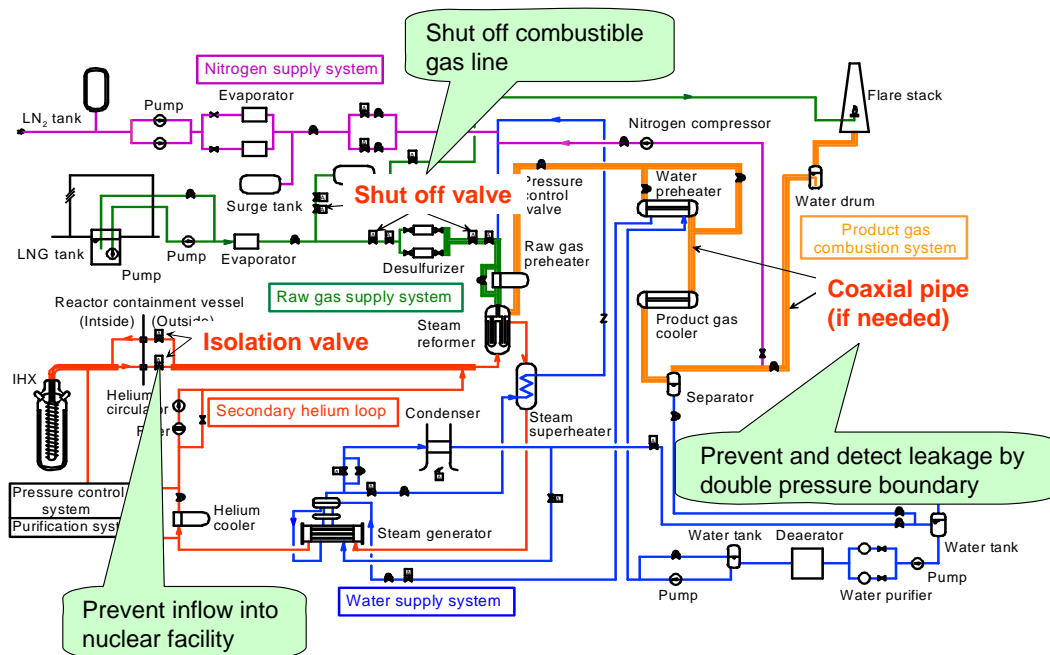


Fig. 4-13: Measures in the HTTR plus SMR system against fire and explosion

Adequate measures for occurrence of an event or its propagation are provided: pressure resistance design, combustible gas leak detection system, fire extinguishing system, emergency shutoff system. Incidents with small or medium leakages appear to represent no hazard to the nuclear system. In terms of mitigation, safety distance and alternatively an explosion- / fire-proof wall to allow shorter distances are considered (Fig. 4-14).

Explosions inside the nuclear containment resulting from the ingress and ignition of combustible gases may have a severe impact on safety-related systems of the reactor. To minimize explosion hazards inside, helium piping and chemical reactor should be designed according to the highest level of reliability and laid out against extreme (design) earthquakes. Furthermore, a combination of the containment vessel isolation valve installed in the helium piping and the emergency shutoff valve in the process feed line is planned.

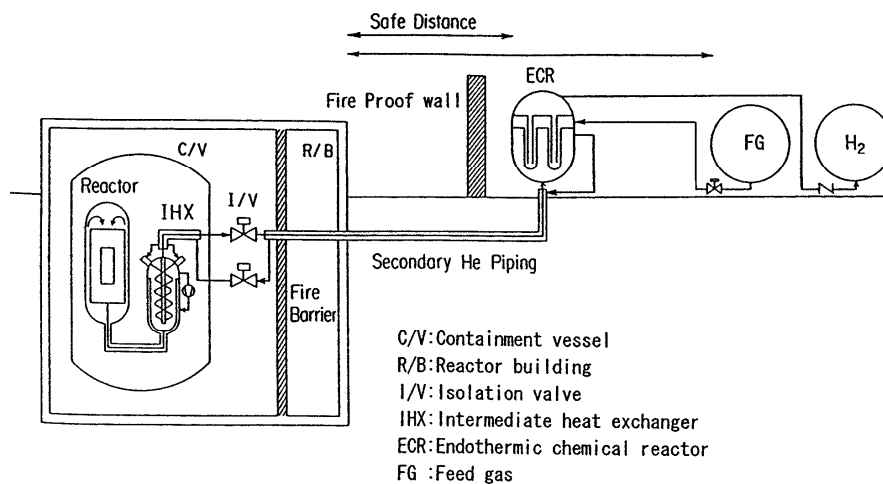


Fig. 4-14: Safety design concept against fire and explosion for the HTTR plus SMR

Concerning the concrete of the building, it is expected to exhibit no significant change of its mechanical properties over 24 hours at 175°C, and is designed to not exceed a wall-averaged limit of 175°C from radiation heat, in order to maintain the structural strength of the concrete.

Overpressure of the incidental blast wave is limited to 10 kPa (USA: 7 kPa, Germany: 30 kPa) to ensure no failure of the reinforced concrete and the steel frame components.

4.2. Licensing Requirements

4.2.1. Safety Features of HTGRs

The designs of high temperature gas-cooled reactors are very different from those of light water reactors, for which a known and stable regulatory environment has been established over many years. In LWRs, the safety philosophy has largely been driven by the possibility of core meltdown.

The unique design of a modular HTGR, however, uses inherent safety features which makes events leading to severe core damage unlikely. Its potential use in high temperature industrial processes and the possibility of using direct cycle gas turbines are two additional major aspects that set it apart from LWRs. For all these reasons, the existing known and stable regulatory environment established for LWRs is not well suited for the licensing of modular HTGR power plants and, therefore, a different approach is necessary.

The modular HTGR represents a fundamental change in reactor design and safety approach from the traditional LWR: safety is achieved through the combination of safety characteristics inherent to the HTGR design and a selection of features that maximise these inherent safety characteristics.

The modular HTGR has the capability to cool the reactor entirely by passive heat transfer mechanisms after accidental events without exceeding the temperature at which the coated particles begin to fail. This characteristic is achieved by limiting the core size and density and configuring the annular core in such a way that natural heat removal processes can maintain fuel temperatures within allowable limits. In order to obtain sufficiently high power, the core height is chosen as large as possible.

The safety features of modular HTGRs are based on the design condition that, even in the case of failure of all active cooling systems and loss of coolant (depressurization), the fuel element temperatures would not exceed the limits at which all radioactive fission products remain confined within the fuel elements. In the event that the active systems are unavailable, core decay heat is transferred by conduction to the pressure vessel and then by radiation from the vessel to the natural circulation Reactor Cavity Cooling System (RCCS). If the RCCS is also assumed to fail, heat is removed by conduction into the reactor cavity walls and surrounding environment. These mechanisms are sufficient to maintain the maximum core temperature below the design limit.

The core power density of less than 10 MW/m^3 and the core configuration are chosen to allow the passive decay heat removal from the core by heat conduction, thermal radiation and natural convection. These passive features of the modular HTGR ensure that even for extreme accidents with very low probabilities of occurrence, the cumulative fission products release at the site boundary remains within acceptable limits without the need for active powered systems or operator action. Furthermore the large heat capacity of the core graphite and the fuel matrix material makes core heat up very slow. A substantial period of time on the order of days is available to take corrective actions to mitigate abnormal conditions.

These HTGR design features are capable of making the concept inherently safe and allow reducing the measures to be considered for the situation of a degraded core.

4.2.2. Definition of Safety Approach

The objective is to define the main safety concepts and principles (e.g., defence-in-depth, physical barriers, event classifications principles, safety functions, target radiological release limits) applicable to modular HTGRs [Prison 2003].

For this purpose, the internationally accepted applicable standards, principles, methodologies (e.g., IAEA safety guides, OECD/NEA recommendations) and licensing materials available for former concepts like HTR-MODUL, THTR-300 in Germany or MHTGR in the US, and HTGR projects like PBMR in South Africa as well as the GT-MHR in the US can be used. The so-called EUR utility requirements, a document to provide design requirements for LWRs produced by utilities from nine European countries, should also be considered as a reference.

A tailored set of safety requirements delivered from the general consolidated principles of nuclear safety should be developed incorporating the specific characteristics of this kind of reactor.

Naturally, the general guidelines and principles must be adapted for the modular direct cycle HTGRs taking into account its inherent and unique safety features. In particular, the greatly reduced probability of accidents with severe fission product release must be considered.

As with other nuclear reactors, there are different levels of defence-in-depth to be considered in the design:

- Prevention of abnormal operation and of failures;
- Control of abnormal operation and detection of failures;
- Control of the design basis accidents;
- Control of severe plant conditions, including prevention of the accident progression and mitigation of the consequences of severe accidents with core damage;
- Mitigation of radiological consequences of significant releases of radioactive materials.

An important discussion topic for the HTGR is whether or not a “conventional” containment structure or some other mitigation system or process is required in order to provide adequate protection for the public. The decision to stipulate a containment has to be made based on technical judgement, taking into account the design features of the HTGR.

One significant advantage of the HTGR is the high resistance of the fuel against fission products release. The coated fuel particle with its successive protective shells constitutes a very effective confinement barrier. The requirements for confinement/containment provided by the third barrier will therefore be re-assessed considering the properties of the first barrier of HTGR fuel, the nature of the design basis accidents to be considered for licensing modular HTGRs and the complete exclusion of core melt situations for this type of reactor.

The confinement requirements will be closely related to the fuel barrier efficiency and fuel quality control process. The results of safety analyses and especially those related to fission product behavior are important data for the definition of quantitative confinement requirements. The integrity of the safety barriers will be also assessed in relation to external accidental events such as aircraft crash, chemical explosions, earthquakes.

4.2.3. Key Licensing Issues

It is important to identify the key safety aspects to which the Safety Authorities may be attentive in order to consolidate the developed safety approach and to anticipate the difficulties that may be encountered during the licensing process.

The following are some of the possible key safety issues that have already been identified for high temperature reactors:

- Fuel performance and qualification;
- Reactor pressure vessel (qualification of high temperature metal);
- Reactor vessel instrumentation;
- Plant shutdown phase (anti-reactivity margin);
- Completely passive system for ultimate heat sink and cavity cooling;
- Air ingress accident;
- Calculation of the source term / fission product transport;
- Absence of “conventional” containment;
- Accident management: benefit of the grace period without operator actions.

A major effort is needed to develop the safety case for HTGR designs that rely solely on the fuel to provide containment for the fission products. For the purpose of safety demonstration and licensing, these fuel capabilities have to be demonstrated in the different situations to be considered. Furthermore detailed safety analyses extended to all potential design basis events (design basis conditions and design extension basis conditions) that can be postulated to occur have to be performed to confirm that the design features are suitable and that the safety objectives are met.

In addition, the issues concerning fission product retention, transport, deposition, and remobilization, along with helium contamination and the thermal-hydraulic behavior of the reactors have to be analyzed. Achieving knowledge of retention factors of the fission products in the primary system components is important in order to evaluate as precisely as possible the helium contamination and release into the environment, during normal operation and during accidents.

5. MARKET POTENTIAL & ECONOMIC ASSESSMENT

5.1. Introduction

This section of the report on Market Potential & Economic Assessment identifies the potential markets for nuclear hydrogen production in the medium and long-term periods. Transition technologies with reduced CO₂ emissions are evaluated against zero emission alternatives with regard to saving fossil fuel reserves. The market potential of using off-peak electricity from existing LWR are addressed, too. The EPRI study [EPRI 2003] has been used as a point of departure in evaluating the economics of hydrogen production for Western Europe (WEU).

Current developments and statistics point towards a steady growth in the total energy use in WEU (Fig. 5-1), although the worldwide trend is expected to grow aggressively. This is due to the intensified economic growth from developing countries, especially in China and India, whereas on the other hand, much of WEU, which is a mature market economy, has trended away from more energy-intensive manufacturing industries towards less energy-intensive service industries.

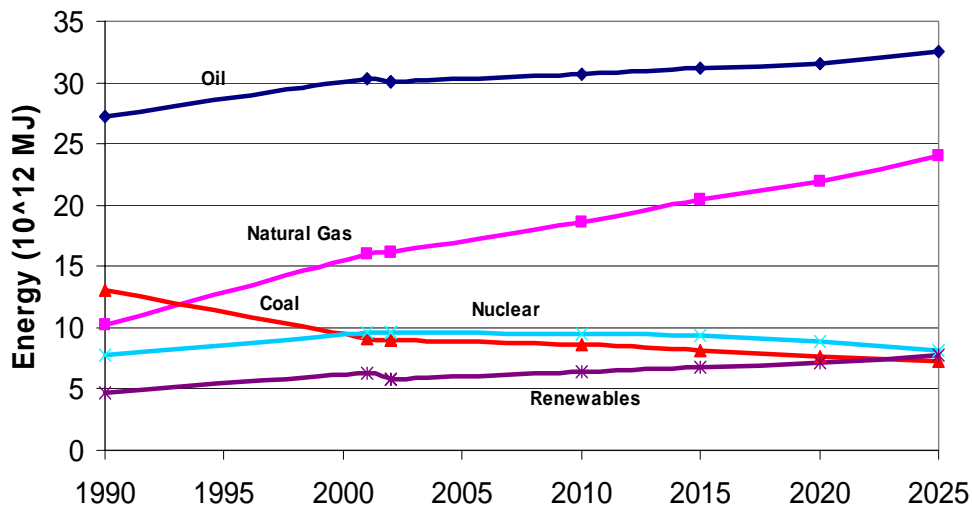


Fig. 5-1: Western Europe's projected energy consumption by energy type (1990 to 2025) [EIA 2005]

The nuclear industry as depicted in Fig. 5-1 can expect a steady decline of the region's energy share in the short-term period, if the status quo on national nuclear policies persists. However with the current pressing environmental issues on CO₂ emissions and concerns on energy security, as well as the prospects of a hydrogen economy in Europe, the use of nuclear energy would be favorable. Currently, oil constitutes the largest share (42 %) of the energy consumed in the WEU region. Nuclear reactors in the region contribute 14 % of the energy, which can be enlarged through the new avenues that the hydrogen economy can open. Hydrogen as an energy carrier can substitute oil in the transportation sector which presently makes up 57 % of all the oil used, and hence about 24 % of the regions total energy [EIA 2005]. All in all, oil and natural gas take up 65 % of the regions energy. This constitutes a potential market for the non-electricity nuclear energy of more than four times the current nuclear electricity capacity. For both short-term and long-term periods, nuclear hydrogen penetration into these markets will experience different resistances and challenges.

For the hydrogen economy to be realized, hydrogen production has to be technically feasible and affordable relative to current fossil fuel prices. This economy will also need to be introduced in strategic phases so as to manage technological readiness of all related hydrogen applications and the market demands and supplies.

5.2. Rollout Strategy for Hydrogen Production

The drivers for the non-electric use of nuclear in the hydrogen economy stems from the following premises:

- Nuclear energy can be used to produce hydrogen on a large scale to replace CO₂ emitting fossil fuels;
- By replacing fossil fuel, fossil reserves will be saved for later use in environmentally friendly applications (hopefully by then, the current carbon capture methods would have evolved economically);
- National energy security from extended fuel reserves and independence from foreign oil uncertainties.

However, the safety trust is still fragile, such that any other nuclear incident, even if there are no casualties, could have a large unfavorable impact on the future of nuclear energy. Waste and non-proliferation issues remain a major concern in the public's mind. These issues must be seen by the public – who's buy-in is crucial – to be fading with importance, while the benefits of using nuclear increases.

Fig. 5-2 shows the broader picture of where nuclear energy fits in into the major energy sectors with the aim of reducing CO₂.

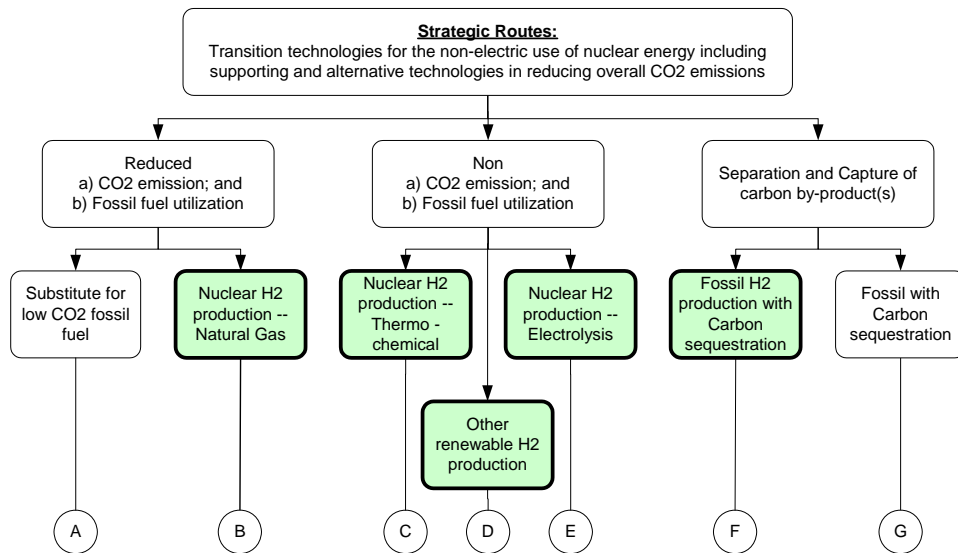


Fig. 5-2: Strategic routes for energy technologies for reducing overall CO₂ emissions (see also Table 5-1)

All these different technological routes will have to compete with each other and in certain cases augment each other in the process of opening up the hydrogen economy from now on. These routes are explained further in Table 5-1:

Table 5-1: Different strategic routes for energy technologies that reduce overall CO₂

Route	Description
A	Low CO ₂ fossil fuels primarily from coal and oil fuel should be replaced by “cleaner” coals and natural gas, respectively.
B	A nuclear system coupled with a steam-methane reformer can supply hydrogen mainly for the transport sector to replace oil fuels and to help meeting the initial hydrogen demand while other CO ₂ free technologies are getting ready. Steam-methane reformers coupled to a nuclear process heat plant will be ready when the HTGR designs become commercial. Other studies indicate a possibility of using LWR designs coupled to a dimethyl ether (DME) reformer to produce hydrogen [Fukushima 2003]. This is because they require energy input at low temperatures ~ 300°C.
C	Nuclear hydrogen production by thermochemical water splitting represents a more promising technology that will be available in the long term. This is mainly due to the large production capacity and energy efficiency with no CO ₂ emissions.
D	Hydrogen production by renewable technologies has to be included in the technology mix, although it will have limitations on large-scale operation. Other more promising technologies that are talked about are far from reaching commercialization.
E	Nuclear hydrogen production by electrolysis (conventional and high temperature operation) is also as promising as the thermochemical technology except that it operates at a lower theoretical efficiency (i.e., taking the efficiency of both the hydrogen plant and the energy plant). Electrolysis is a mature technology that can start being used together with current H ₂ production methods for supplying the hydrogen in a growing hydrogen economy.
F	Producing hydrogen from methane that is heated by natural gas can offer the H ₂ at low cost depending on the price of natural gas. If sequestration costs and CO ₂ penalties are added to the hydrogen production cost, the technology may become less attractive although it can certainly augment the supply of hydrogen in the initial phase of the hydrogen economy.
G	If sequestration technologies improve drastically in economics, some energy markets may become difficult to penetrate with the hydrogen production technologies. However, it may be harder to implement sequestration technologies in the transport sector, where major CO ₂ emissions take place.

There are indications that nuclear fuel resources are more abundant than fossil hydrocarbons to last for several hundred years at current and projected usage rates [EPRI 2003]. These reserves could be extended significantly by nuclear fuel recycling and even more, if breeder reactors were added. These reserves make it feasible to consider the endeavor of substituting fossil fuels with nuclear hydrogen.

Fig. 5-3 shows the CO₂ emission factors for fossil fuels compared with hydrogen, as well as the energy available for the respective fuel applications. For non-electric use of nuclear energy, fuel

oil (including diesel and gasoline) as well as natural gas and methane, are of interest, since their markets can be easily targeted to include hydrogen fuel produced from nuclear energy. In Fig. 5-4, the market share (by energy sector) for the consumption of fuel oil and natural gas is shown as a percentage of WEU's total energy. The transportation sector could be targeted first in introducing hydrogen applications (as is currently done with prototypes), and then the other sectors could follow in the long term as infrastructure hurdles are overcome.

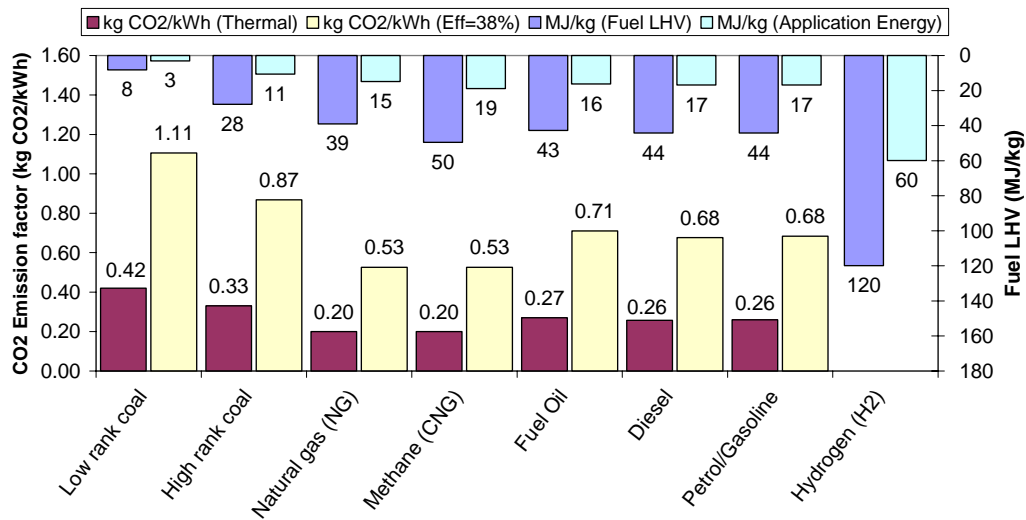


Fig. 5-3: CO₂ emission factors for different fuels

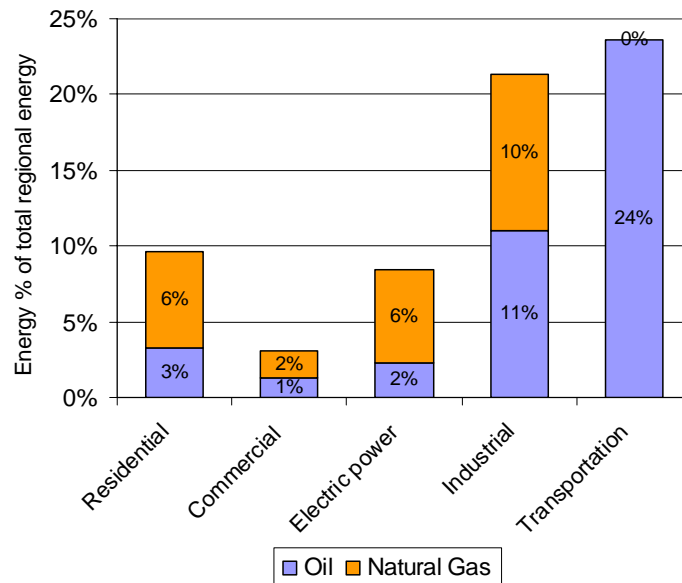


Fig. 5-4: WEU projected oil and natural gas energy market share (2002 – 2025) [EIA 2005]

An advantage that hydrogen applications have relative to fuel and oil applications is in the efficiency of energy conversion. Fuel cell applications which will be used in the long term, are able to extract more power from the same quantity of fuel if compared to traditional fossil fuel applications. This direct process results in a reduced amount of fuel being consumed and greater efficiencies, 30 % to 90 %, depending on the fuel cell system. In Fig. 5-3, the application energy is shown next to the fuel LHV, for which efficiencies of 38 % and 50 % were assumed for the fossil fuel and the fuel cell applications, respectively.

The different technologies for producing hydrogen can go through transitions as illustrated in Fig. 5-5 as they mature in the hydrogen economy.

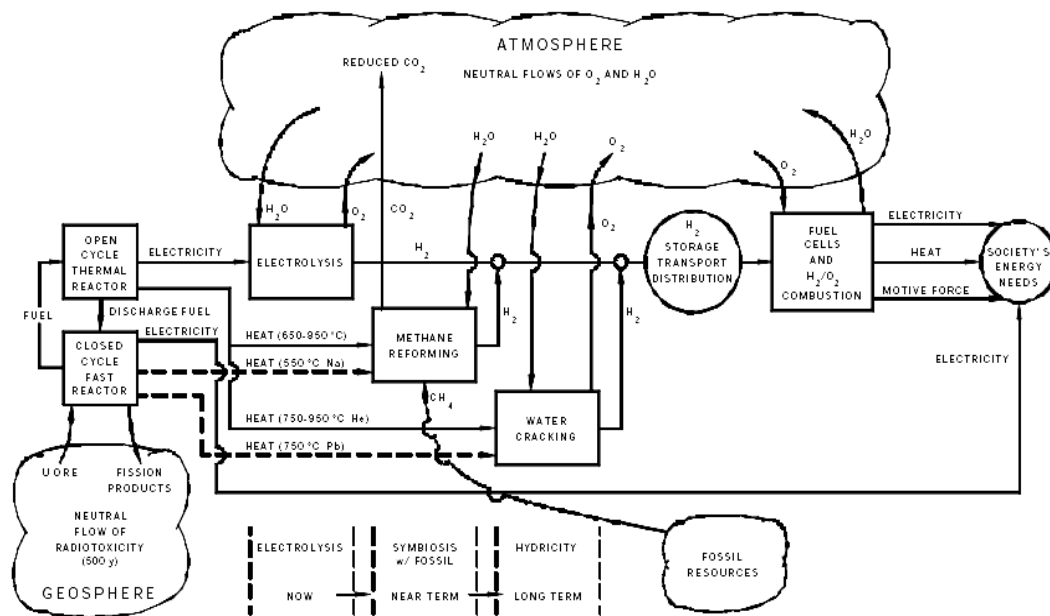


Fig. 5-5: Nuclear hydrogen transition to sustainability [Walters 2002]

The long-term goal is to produce hydrogen from thermochemical splitting of water by high temperature reactors ($\sim 900^{\circ}\text{C}$), which will be shown in the next section to be the best option for the non-electric use of nuclear energy. As the non-electric use of nuclear grows in the hydrogen economy, issues of ore reserves will become important. For this reason, fast breeder reactors would need to play a bigger role in providing high temperature process heat for thermochemical water splitting systems. This will help in the optimal use of nuclear fuels and will result in a more sustainable transition.

The transition technologies should be introduced in a step-wise fashion that matches supply and demand as the hydrogen economy develops, hence managing the “chicken-egg” issue. To this effect, an approach as is shown in Fig. 5-6 would resemble a guided step-wise introduction of hydrogen production and application in the growing hydrogen market. As the market volume increases, hydrogen cost reductions can be expected due to economies of scale and further improvements in hydrogen related systems.

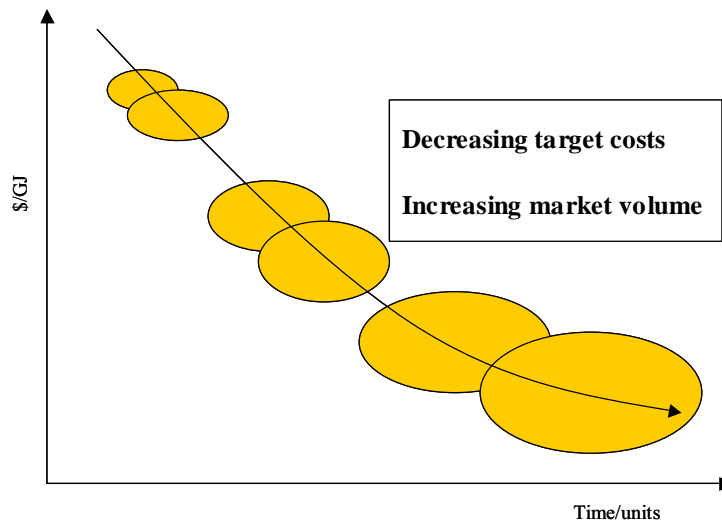


Fig. 5-6: Step-wise mass market entrance for H₂ production and applications [Ziolek 2005]

The objectives of a step by step to mass market are as follows:

- Validate technical and economic viability;
- Receive feedback from real environment;
- Guide further R&D activities;
- Overcome main legal and political barriers;
- Define codes and standards well in advance;
- Get public buy-in and that of other critical stakeholders early on.

The following Fig. 5-7 shows typical hydrogen applications that are currently being developed, and their readiness into the hydrogen economy.

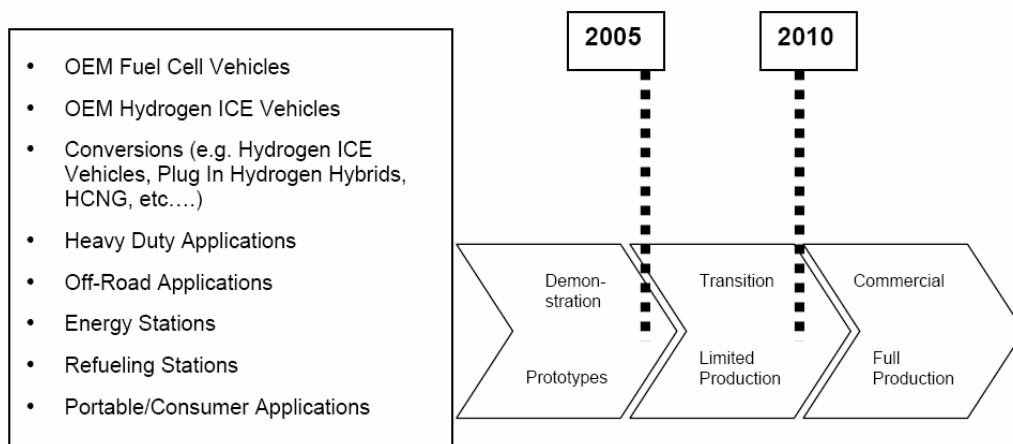


Fig. 5-7: Hydrogen applications under development [Hydrogen Highway Network 2005]

5.3. Economic Comparisons and Benefits of Nuclear Heat Utilization

The assessment used to compare and quantify the different nuclear heat utilization was based on a study by EPRI [EPRI 2003]. This assessment only focuses on different hydrogen production technologies with various configurations of HTGR plants and the possible use of current LWR plants. The hydrogen production plants and their respective nuclear plant couplings are shown in Table 5-2. Note that the nuclear utilities that provide electricity for hydrogen production are – for the purpose of this assessment – also regarded as falling under the category of nuclear heat utilization. Descriptions of these nuclear plants considered in this assessment are as follows:

- GT – Gas turbine HTGR made up of four 600 MWt units operating with a coolant outlet temperature of 850°C and an efficiency of about 48 %;
- PH 850– Process heat from HTGR made up of four 600MWt units with the gas turbine replaced by an IHX and a coolant outlet temperature of 850°C;
- PH 950– Process heat from HTGR made up of four 600MWt units with the gas turbine replaced by an IHX and a coolant outlet temperature of 950°C;
- PH-GT – Four 600MWt HTGRs, each transferring about 20 MWt of process heat and about 580 MWt to an indirect steam cycle with an efficiency of about 37 %, and a coolant outlet temperature of 850°C;
- LWR & Renewables – LWR and renewable (wind, hydro, etc.) plants sized to about 1152 MWe; and
- NG-SMR– Energy plant fueled by natural gas, this is included in order to compare with nuclear powered hydrogen production systems, because currently it is able to provide cheap hydrogen production, if CO₂ tax penalty is excluded.

Fig. 5-8 shows the effect of hydrogen plant sizing on the cost of hydrogen. A good sizing will be one with a throughput of more than 1 million Nm³/d. The figure also shows the effect of variance in electricity price on the cost of hydrogen for a specific throughput sizing. Hydrogen production methods using process heat do not vary with electricity price. For a dedicated GT-HTGR with fixed electricity price of about 3 US c/kWh, a hydrogen cost of \$15/MMBtu is achieved. Renewable electricity utilities like wind with high electricity prices will generate expensive hydrogen. For LWRs and other reactors producing electricity during off-peak hours, hydrogen can be produced cheaply, especially if these power plants are solely dedicated to be connected to hydrogen production facilities. Electricity from the grid during off-peak periods will probably still be higher than the price of electricity for a dedicated hydrogen power plant utility due to slight profits and taxes still charged at off-peak operation. The price of nuclear electricity in countries such as France will result in cheaper hydrogen production by conventional electrolysis.

Table 5-2: Comparison of leading hydrogen production methods [EPRI 2003]

Energy plant	Energy form	Power input	Energy plant capital cost [million \$]	H ₂ source	H ₂ process type	By-products	H ₂ plant throughput [10 ⁶ Nm ³ /d]	H ₂ plant capital cost [million \$]
GT	Electric	1152 MWe	1267	Water	Conventional electrolysis	O ₂	6.2	756
PH 850	Process heat	2440 MWt	887	Methane	SMR	CO ₂	60	2580
PH 950	Process heat	2440 MWt	1014	Water	S-I	O ₂	8.4	672
PH-GT	Process heat & electric	~ 80 MWt & 860 MWe	1276	Water	HTE	O ₂	7	700
LWR & Renewable	Electric	(Note 1)	(Note 1)	Water	Conventional electrolysis	O ₂	(Note 1)	(Note 1)
NG-SMR	NG heat	2440 MWt	61	Methane	SMR	CO ₂	38	1672

Note 1: The H₂ throughput and unit capital cost will vary with the electricity price at which the power is delivered. Typically, the hydrogen production facility will be sized with an energy plant of about 1152 MWe and above, which will correspond to a throughput of more than 6 million Nm³/d of H₂.

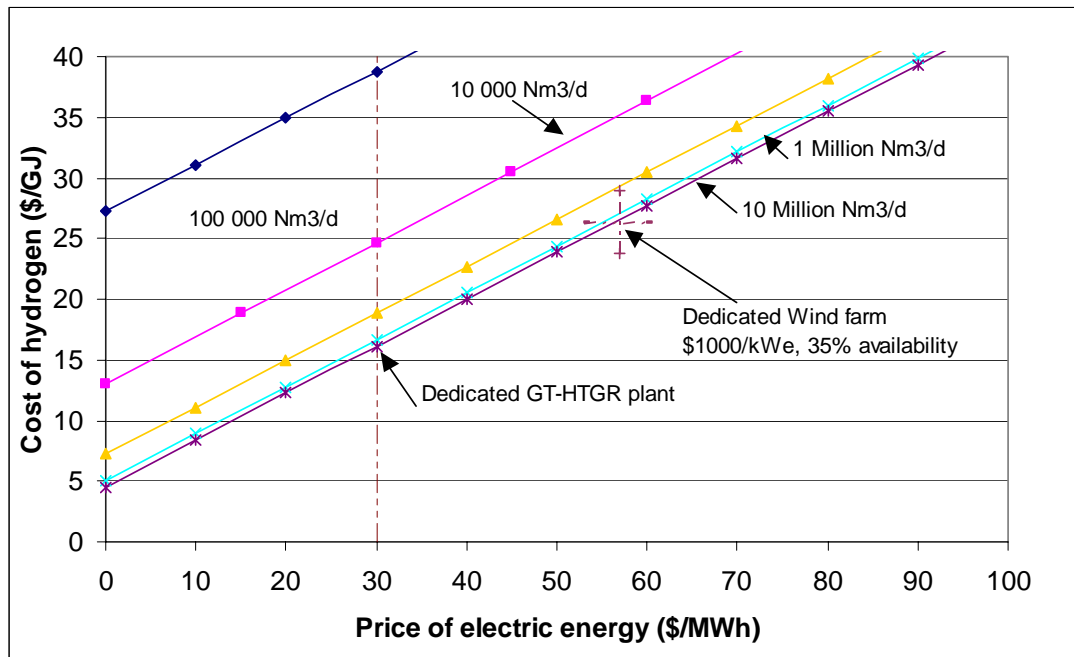


Fig. 5-8: Cost of hydrogen vs. electricity price for conventional electrolysis [EPRI 2003]

Furthermore it should be noted that if a hydrogen production plant were coupled to an energy system that is only available during electricity off-peak periods, then the reduced availability in the hydrogen system would have to be factored into the capital cost. This will have a bottom line effect on the price of hydrogen produced, although this change in cost price will not be quite significant, if the hydrogen production throughput is in the order of 10 million Nm^3/d . To illustrate this, it could be assumed that off-peak hours occur for half a day. This will imply that for a hydrogen production plant with a throughput of 10 million Nm^3/d , the actually throughput for half a day will be 5 million Nm^3 which, according to Fig. 5-8, will still result in good hydrogen cost price.

The cost of hydrogen and other cost assessments referred to in this section were based on assumptions stipulated in Table 5-3.

Table: 5-3: Assumed cost parameters based on US EPRI assessment [EPRI 2003]

Financial parameters	Value
Inflation rate	0 %/yr
Property tax and insurance rate	2 %/yr of capital
Discount rate	7 %/yr
Annual capacity factor (for both the power plant and the hydrogen production facilities)	90 %
Final hydrogen pressure	7.6 MPa
Assumed price for grid supplied electricity	\$30/MWh
Reference price for natural gas	\$5/GJ (~ \$5/MMBtu)
Natural gas combustor capital cost	\$25/kW(th)
Nuclear fuel (including enrichment and conversion)	\$7.4/MWh
Energy power plant O&M	\$3/MWh
Decommissioning costs (based on GT-MHR)	\$1/MWh
Cost of demineralized water	\$1.04/ m ³
Credit value for gaseous oxygen	\$20/t
Assumed CO ₂ emission tax	\$40/t
Cost to sequester CO ₂	\$40/t*

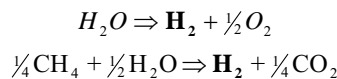
*Costs for CO₂ tax or sequestration were modified from \$30/t [EPRI 2003] to \$40/t. Present carbon capture technologies to reduce emissions are said to be between \$35/t of C and \$264/t of C. If it is assumed that a sequestration technology with an average carbon capture cost is to be used, then the cost will be \$150/t C (~\$40/t of CO₂).

This cost is not far out from other studies on the cost of future sequestration technologies [Riahi 2004].

A cost model with six cost components is used to calculate the hydrogen cost (expressed in \$/GJ or \$/kg) for the different hydrogen production technologies shown in Table 5.2.

1. Feedstock Cost Component

The main feedstock sources are water for electrolysis and thermochemical hydrogen production, as well as natural gas for hydrogen production by methane reforming. Natural gas is purified into methane at a cost of 5 % of the price of natural gas. From the stoichiometric ratios for the reactions of hydrogen production, as shown the equations below, the amount of feedstock needed to produce an annual hydrogen throughput is calculated. This annual total cost of feedstock is then divided by the total annual production of hydrogen to calculate a feedstock cost component in the hydrogen cost price.



2. Hydrogen Plant Capital Recovery and O&M Cost Component

The hydrogen plant capital cost is calculated by first multiplying the annual hydrogen throughput (Nm^3/d) for one of the hydrogen technologies shown in Table 5.2 with the hydrogen plant capital cost ($\$/\text{Nm}^3/\text{d}$) to get the overnight cost (including IDC). Then an annualized fixed charge rate (AFCR) is multiplied with the overnight cost to determine the annual hydrogen plant capital cost. The AFCR consists of the following cost rates:

- Discount rate for recovery of the cost of capital (7%/yr),
- Property tax and insurance rate (2%/yr), and
- Depreciation rate (depends on the recovery method used, for the EPRI analysis MACRS was used).

The annual hydrogen plant capital cost is then divided by the total annual hydrogen throughput to determine the hydrogen plant capital cost component on the cost price of hydrogen. The O&M total annual cost is calculated as 5 % of the overnight cost and then divided by the total annual hydrogen production to determine the hydrogen plant O&M cost component.

3. Power Plant Capital Recovery Cost Component

Like in the hydrogen plant capital cost, the overnight cost is calculated by multiplying the total available annual energy (MWt or MWe) with the specific energy cost ($\$/\text{kW}$ thermal or electric). Then the AFCR, with the same rates applicable as mentioned above, is multiplied with the total invested capital cost to determine the total annualized power plant capital cost. This total annualized power plant capital cost is then divided by the total annual hydrogen production to determine the power plant capital recovery cost component.

4. Power Plant Fuel, Decommissioning and O&M Cost Component

The power plant fuel, decommissioning and O&M cost is calculated separately from the capital recovery cost. These costs are calculated together by multiplying the total available annual energy (MW) by the respective costs (in $\$/\text{MWh}$) and dividing by the total annual hydrogen production in order to get their combined cost component on the price of hydrogen produced.

5. Hydrogen Pumping Power Cost Component

The annual energy required to pump the hydrogen gas to the final pressure of 7.6 MPa is multiplied by the grid supplied electricity cost. The product is then divided by the total annual hydrogen production to get its cost component in the cost of hydrogen.

6. By-Product Credit/Tax Cost Component

By-product credits and taxes are calculated in the same way as the feedstock cost components.

As an example, Fig. 5-9 illustrates the cost contribution of all the cost components explained above on the total cost of hydrogen produced by the different systems outlined in Table 5.2. A discount rate of 7 %/yr and a natural gas price of $\$4.6/\text{GJ}$ were used for calculating the cost price.

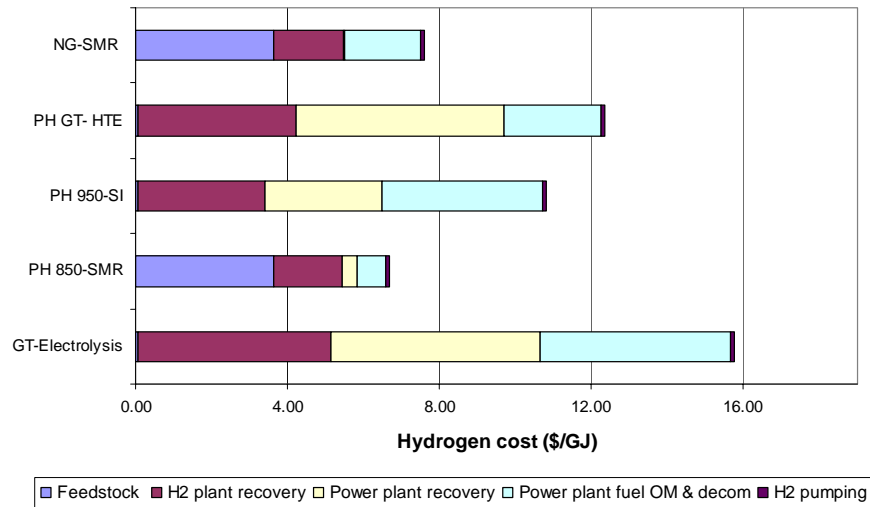


Fig. 5-9: Cost components of H₂ produced from different technologies (discount rate 7 %/yr)

Undoubtedly the price of natural gas will rise as more demand increases with time and the security issues on energy intensifies. In 2005, the gas price in Europe was at an average of \$7/GJ for the industry sector, as shown in Fig. 5-10 below.

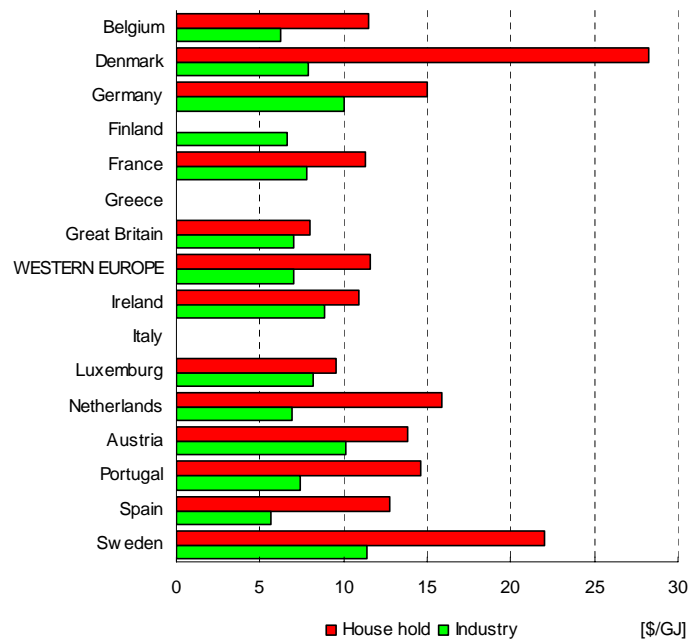


Fig. 5-10: International prices for natural gas in January 2005 (excl. VAT) [Energie.nl 2005]

Already at this price for natural gas, nuclear hydrogen production is more attractive economically, as can be illustrated in the Fig. 5-9.

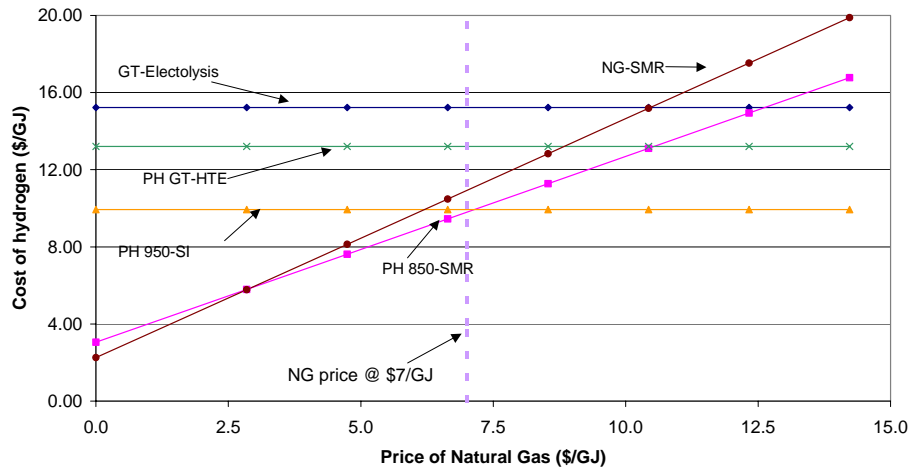


Fig. 5-11: Cost of H₂ vs. price of NG for leading technologies, without by-product credit and penalty costs (discount rate 7 %/yr)

The cost price for producing hydrogen is evaluated for the different technologies stated in Table 5-2, with varying prices for natural gas. Fig. 5-11 shows the cost of hydrogen production excluding by-product credit and penalty costs. It can be expected that in the short term, hydrogen can be produced without any penalties for emitting CO₂. In this case, hydrogen production by steam methane reforming, heated with natural gas, will be the cheapest option if NG price falls below \$7/GJ. For non-electric nuclear, hydrogen can be produced for 15 \$/GJ for an electric cost price of 3 US c/kWh from current LWRs. Electricity cost price projections by NEA (OECD/IEA NEA 2005), for the year 2010 onwards as shown in Table 5-4, can result in cheaper nuclear hydrogen cost prices mainly from LWRs.

Table 5-4: Projected electric cost prices 2010 onwards (US cents/kWh)

	Nuclear	Coal	Gas
Canada	2.60	3.11	4.00
Czech Rep	2.30	2.94	4.97
Finland	2.76	3.64	-
France	2.54	3.33	3.92
Germany	2.86	3.52	4.90
Japan	4.80	4.95	5.21
Korea	2.34	2.16	4.65
Netherlands	3.58	-	6.04
Romania	3.06	4.55	-
Slovakia	3.13	4.78	5.59
Switzerland	2.88	-	4.36
USA	3.01	2.71	4.67

From Fig. 5-8, the cost price for producing hydrogen by conventional electrolysis, at France's nuclear electricity price of 2.54 US c/kWh, will be 13 \$/GJ excluding credit cost reduction for selling oxygen by-product. It should be noted that during peak hours, the market electricity price will sell much higher, and during off-peak operation, the price will come down to values close to the cost price.

In the medium term, as the HTGR technology takes shape, hydrogen production by the Sulfur-Iodine method will yield a hydrogen cost price of about 10 \$/GJ from a completely "clean" energy source as shown in Fig. 5-11. High temperature electrolysis (HTE) at a cost price of 13 \$/GJ will do better than conventional electrolysis, if nuclear electricity cost price remains at \$30/MWh.

In the long term, it is expected that policy makers in developed countries will implement CO₂ by-product penalties. For this scenario, hydrogen cost prices would look like as shown in Fig. 5-12, which include oxygen credit cost reductions and penalties for producing or sequestration of CO₂.

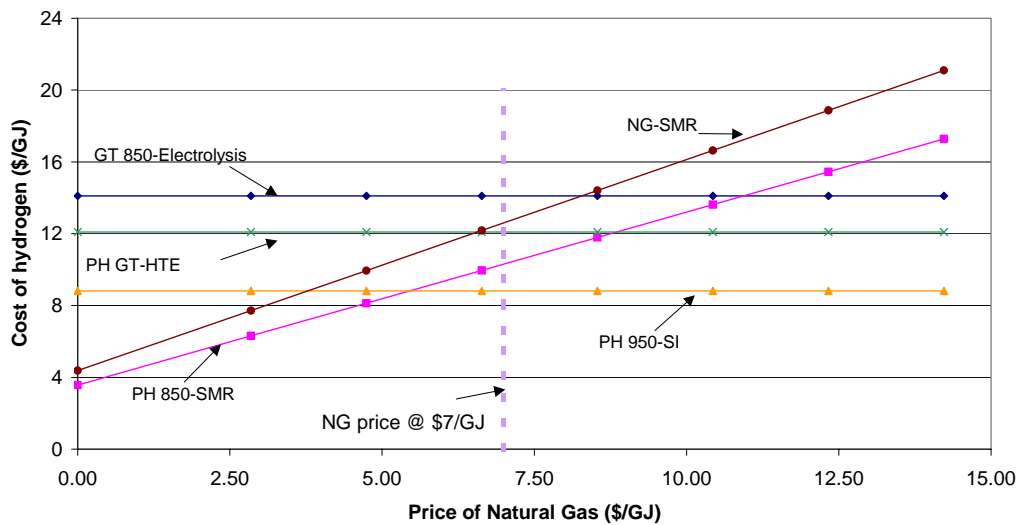


Fig. 5-12: Cost of H₂ vs. price of NG for leading technologies, with by-product credit and penalty costs

In the long term, this economic assessment indicates that nuclear hydrogen production by S-I for natural gas prices above \$6/GJ, will be the favorable technology with a hydrogen cost price of about \$8.8/GJ. Past the natural gas price of \$9/GJ, HTE with a hydrogen cost price of \$12/GJ, will be a cheap and clean alternative to hydrogen production using S-I. These prices do not take into account economies of scale, which could reduce the cost prices quite significantly. With this technology and its economic feasibility, the full benefits of a hydrogen economy can be realized, i.e., reduced overall CO₂ emissions, a cleaner environment, and energy security.

The cost items of the nuclear hydrogen production technologies shown thus far in comparison to steam methane reforming are shown in the figure below with by-product credit/taxes. The cost contributions to the total hydrogen cost were evaluated for different economic cases of discount rate and NG price.

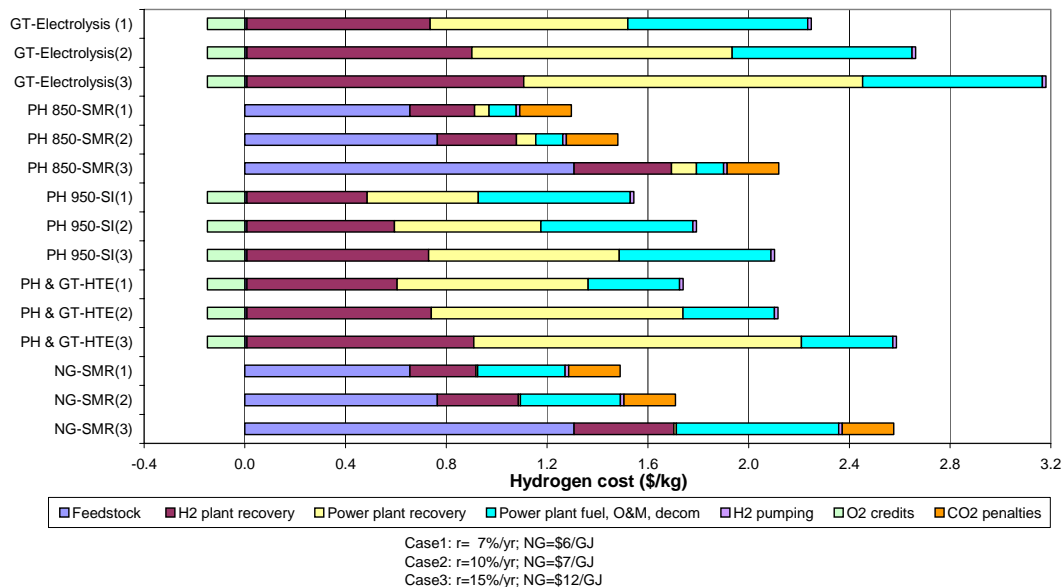


Fig. 5-13: Cost components of H₂ produced from different technologies and different economic cases

As would be expected from Fig. 5-13, high natural gas prices such as in Sweden according to Fig. 5-10, will have a high feedstock cost component on the SMR dependent systems, whereas on the other hand, high discount rates have a minor influence on them. The CO₂ free, nuclear hydrogen systems are heavily influenced by the discount rate charged on them to produce low cost hydrogen. If industrial natural gas prices stabilize at around \$7/GJ, while high investment returns and interest rates are expected, in a way that discount rates on capital sits at 15 %/yr, then the option for PH 950-SI(3) shown in the figure will not be able to compete with alternative SMR technologies {PH 850-SMR(2) & NG-SMR(2)}.

Hydrogen cost prices from literature, for hydrogen produced by other non-nuclear technologies, are also shown in Table 5-5 below. It seems quite reasonable that hydrogen production using an HTGR coupled to the S-I process, and production by SMR are the most economically suitable methods for the near future.

Table 5-5: Various non-nuclear hydrogen cost prices

H ₂ production process	H ₂ production cost [\$/GJ]	Source of cost
Natural gas reforming (decentralized with fuel price of 4-5 \$/GJ)	7-12 (1-1.7 \$/kg)	[Lucchese 2005]
Coal gasification	13-16 (1.8-2.3 \$/kg)	[Lucchese 2005]
Biomass gasification	17-22 (2.4-3.1 \$/kg)	[Lucchese 2005]
Electrolyser	18-25 (2.6-3.6 \$/kg)	[Lucchese 2005]
Electrolysis with hydro power	21-25 (3-3.6 \$/kg)	[Vandenborre 2004]
Electrolysis with wind power	28 (4 \$/kg)	[Vandenborre 2004]
Electrolysis with solar power	28	[Vandenborre 2004]

In 2005 the Department of Energy (DOE) in the USA, has revised its hydrogen cost price target from around \$1.5/kg to a maximum of about \$3/kg [DOE 2005]. Their strategy is to keep the cost of energy used from hydrogen the same as that of gasoline. This is because, from the consumer's point of view, it is much easier to adjust to a different technology that costs more or less the same as the old. Table 5-6 taken from the EPRI study [EPRI 2003] shows the normalized cost for the different fuels that hydrogen will need to compete with. With these cost prices, hydrogen will be a fuel to reckon with, given that the price of oil will continue to rise in the coming years.

Table 5-6: Fuel parameter comparison

Parameter	Diesel	Gasoline	Propane	LNG	NG	Hydrogen
Average chemical formula	$C_{16}H_{34}$	C_9H_{18}	C_3H_6	CH_4	CH_4	H_2
Density	851 kg/m ³	749 kg/m ³	503 kg/m ³	424 kg/m ³	0.651 kg/Nm ³	0.0899 kg/Nm ³
Normalized energy density [kJ/kg HHV]	45,400	46,450	50,600	55,500	55,500	142,770
Normalized cost [\$/GJ]	5.80	6.84	10.38	3.31	3.06	5.60-9.10
CO ₂ produced per kg of fuel combusted [kg]	3.52	3.35	2.99	2.75	2.75	0

6. ROADMAP

With the recent worldwide increased interest in hydrogen as a clean fuel of the future, Europe has also embarked on comprehensive research, development, and demonstration activities with the main objective of the transition from a fossil towards a CO₂ emission lean energy structure as the ultimate goal.

As a matter of fact, a major hydrogen economy exists already today and is expected to grow at a rate of 4-10% per year. Significant amounts of hydrogen are currently produced in the fertilizer industry for the manufacture of anhydrous ammonia. Hydrogen also plays a large and growing role in the refining of petroleum products, where the reserves of high quality light sweet crude oils are declining and the available crude stocks are becoming progressively heavier. These heavier crudes as well as the increasing share of “dirty fuels” (heavy oils, coal, oil shale, tar sands) require larger amounts of both process heat and hydrogen to produce cleaner burning end-point fuels with a higher hydrogen-to-carbon ratio.

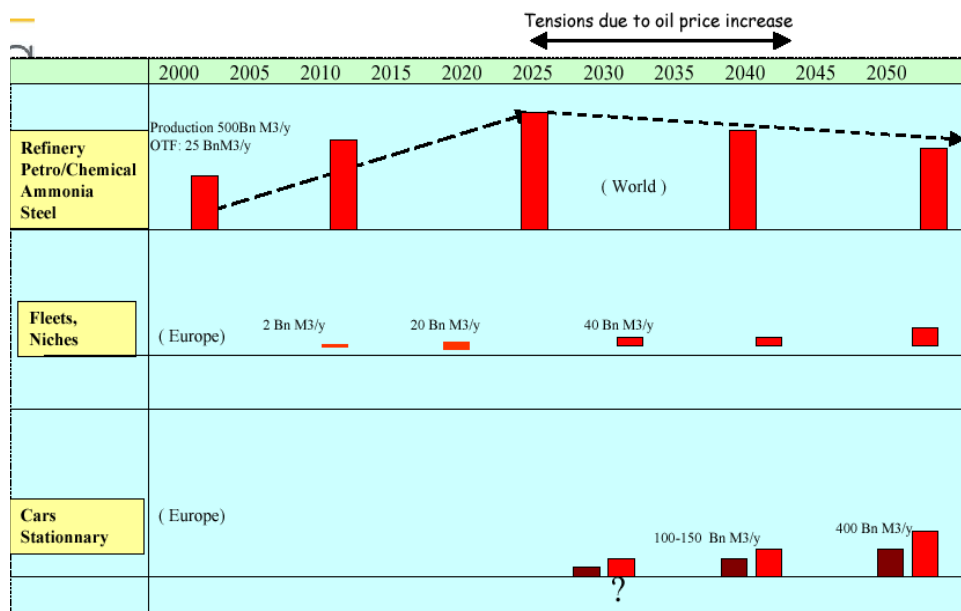


Fig. 6-1: Hydrogen production roadmap [EC 2003]

The projection of the European Commission High Level Group shown in Fig. 6-1 offers a realistic scenario about the hydrogen market and the application areas. It clearly shows that largest near-term markets will be the petrochemical industries requiring massive amounts of H₂ for the conversion of heavy oils, tar sands, and other low-grade hydrocarbons, as well as the fertilizer and steel industries. Furthermore it can be seen that the decrease of H₂ consumption in these areas after 2025 coincides with the market introduction of new H₂ applications such as in the transportation sector or as a distributed electrical energy source through the use of fuel cells.

If hydrogen is to play a major role in a future energy economy, the whole spectrum of primary energies (fossil, nuclear, renewable) for its production must be considered. Different hydrogen production methods are given in Table 6-1 listing benefits and barriers of the respective technologies.

Table 6-1: Benefits and Barriers of Hydrogen Production Technologies [EC 2003]

Technology	Benefits	Barriers
Steam Reforming: Splitting of hydrocarbons with heat and steam	well understood at large scale; commercially available with proven technology; widely available feedstock; highly economic at present; CO ₂ sequestration at large scale; ideal for centralized production.	small-scale units not commercial; CO ₂ emissions; H ₂ contains some impurities; primary fuel may be used directly; subject to NG price fluctuations; in distributed form not yet verified.
Gasification: Splitting of heavy hydrocarbons and biomass into hydrogen and other gases for reforming.	well understood at large scale; can be used for solids and liquids; abundance of (coal) resources.	less hydrogen-rich than methane; lower efficiency; large CO ₂ emissions from coal; feedstock requires pre-treatment; H ₂ requires cleaning prior to use; biomass gasification still at pilot plant scale; low energy density of biomass.
Electrolysis: Splitting of water using electricity	well understood; commercially available with proven technology; high purity hydrogen; modular; convenient for renewable electricity; ideal for distributed production.	electricity price strongly impacts cost of H ₂ ; efficiency of whole chain is low; need for development of durable HTE cells; competition with direct use of renewable electricity.
Thermochemical Cycles: Splitting of water using cheap high temperature heat from nuclear or solar.	potentially massive production at low cost; no GHG emission; high efficiency (~50% expected); international collaboration on R&D and deployment.	not commercial; aggressive chemistry; much R&D work still needed on process, and materials technology; high capital cost; high temperature nuclear reactor deployment needed.
Biological Production: Algae and bacteria produce hydrogen directly under certain conditions.	potentially large resource; no feedstock required.	slow hydrogen production rates; large area needed; low efficiency; appropriate organisms not yet found; still at R&D level.

The first step into the H₂ economy will always be based on existing technologies and established processes. In the near and medium term, fossil fuels are expected to remain the principal source for hydrogen. Natural gas as the “cleanest” fuel among the hydrocarbons is expected to have a couple of advantages as a starting point for the initial hydrogen market (transition phase) as a source of hydrogen in terms of environmental impact (highest H/C ratio), availability, and economy. In the long term, H₂ production technologies will be strongly focusing on CO₂-neutral or CO₂-free methods.

As is shown in the following Table 6-2, hydrogen is not always an ideal vector to carry energy from its place of production to the end-user, because a fairly high amount of energy is lost during handling, storage, and transportation. If the hydrogen is packaged in liquid synthetic fossil fuels, the overall energy consumption would be considerably lower [Bossel 2003].

Table 6-2: Energy Availability [Bossel 2003]

Hydrogen stages of application	Energy cost in % of HHV
Production: electrolysis	43
on-site production	65
Packaging: compression 20 MPa	8
compression 80 MPa	13
liquefaction	40
chemical hydrides	60
Distribution: road, 20 MPa, 100 km	6
road, liquid, 100 km	1
pipeline, 1000 km	10
Storage: liquid, 10 days	5
Transfer: 20 MPa to 20 MPa	1

Nuclear with its virtually no air-borne pollutants emissions appears to be an ideal option for large-scale centralized H₂ production (see Fig. 6-2). The future for hydrogen and the potential for nuclear generated H₂ will be driven by major factors such as

- production rates of oil and natural gas;
- societal and governmental decisions on global climate change gases and CO₂ emissions;
- need for savings of fossil resources for later use in environmentally friendly applications;
- energy security from extended fuel reserves and independence of foreign oil uncertainties;
- and, last but not least, the economics of large-scale hydrogen production and transmission.

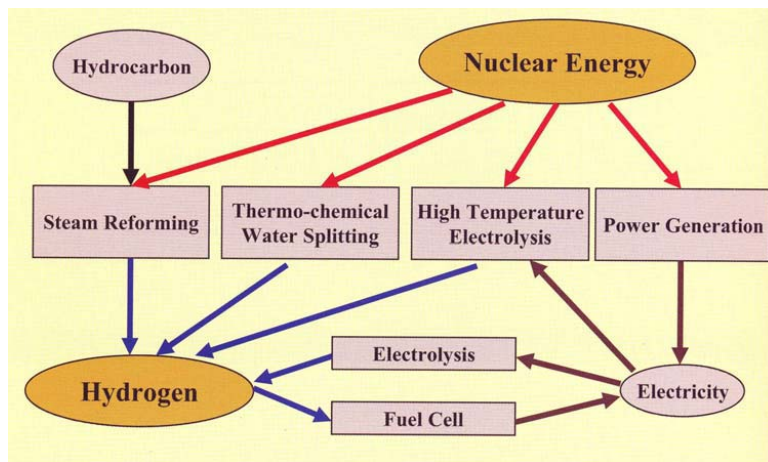


Fig. 6-2: Routes for nuclear hydrogen production [Tsukada 2005]

It is strongly recommended to extend the scope of future industrial activities on the market introduction of nuclear reactors, in particular, high temperature gas cooled reactors, within the spectrum of petrochemical processes and on combined heat and power generation. The main challenge at present is to include the production of hydrogen and combined heat and power applications by means of nuclear energy into the general strategies and to establish transition technologies from present industrial practice or emerging new resources (“dirty fuels”) in order to stabilize the cost for energy. The question of which energy source to be utilized, will be finally decided by the respective country with respect to its domestic resources, and methods on how to guarantee energy security.

If nuclear power as a substitute for the traditional fossil fuels is to be realized in future, further work is needed to accommodate nuclear technology with the various industrial needs. Apart from demonstrating the economics of production and end-use, requirements need to be met in terms of consumer’s perception of availability and convenience, and last but not least, social acceptance both by the population and by the industry itself. A nuclear reactor operated in the heat and power cogeneration mode must be located in close vicinity to the consumer’s site, i.e., it must have a convincing safety concept of the combined nuclear/chemical production plant. Both parts will be separated from each other by employing an intermediate heat exchanger (IHX) as an interface component between the primary helium circuit of the reactor and the heat consuming system, e.g., a steam reformer/steam generator facility. The intermediate circuit serves the safety related purpose of preventing primary coolant to flow through the conventionally designed heat consuming plant and, on the other hand, product gas to access the nuclear reactor building. For economical reasons, all necessary heat exchanger plants should be built, maintained, and repaired according to conventional, not nuclear, specifications.

Although the assessment is mainly devoted to hydrogen production methods, it must be noted that due to the complex interaction of the different chemical processes optimized to a very high degree, the potential supply of energy by nuclear power may not be dedicated to a specific process, but rather cover the overall cogeneration of process steam, process heat, and electricity. Any substitution of process heat in large-scale industrial productions by nuclear energy results in significant CO₂ reductions and in saving of fossil resources at competitive economics which are the dominant challenge for future energy systems. This will include the generation of process steam and sensible heat for a variety of applications beyond dedicated hydrogen production.

A near-term option of nuclear hydrogen production which is readily available is conventional low temperature electrolysis using cheap off-peak electricity from present nuclear power plants. This, however, is available only if the share of nuclear in power production is large. But as fossil fuel prices will increase, the use of nuclear outside base-load becomes more attractive. Main disadvantage is the very low efficiency from today’s reactors making it unattractive for H₂ production although it is a proven and clean technology. To demonstrate the order of magnitude: a 500,000 kg/d H₂ generation plant capable of serving around 1 million FCV, if based on nuclear power and low-temperature electrolysis, would require 500 of the largest electrolyzer units available today consuming a total of about 1 GW of electric power.

Nuclear steam reforming is another important near-term option for both the industrial and the transportation sector, since principal technologies were developed, with a saving potential of some 35 % of methane feedstock. Competitiveness will benefit from increasing cost level of natural gas. The HTGR heated steam reforming process which was simulated in pilot plants both in Germany and Japan, appears to be feasible for industrial application around 2015. It is compatible with a realistic evolution of actual industrial practice and could fully substitute the process heat requirements. The economics of this process will be much more favorable as compared to electrolysis as long as natural gas is available at reasonable cost. Although the process is not free of CO₂ emission, it is justified within a medium to long-term transition period towards a more puristic approach with inherently higher cost.

The know-how acquired from the German projects upgraded with new results and operating experience with the Japanese pilot plant will provide a sound basis for a commercial deployment even before 2015. A strong involvement of European organizations in the HTTR project and targeted complementary work on residual R&D items will be a precondition for this strategy.

A CO₂ emission free option is high temperature electrolysis which reduces the electricity needs up to about 30 % and could make use of high temperature heat and steam from an HTGR. It is evident that the optimization and adaptation of the electrolysis process in combination with HTGR will reduce the drawback of low thermal efficiency of the electrolysis path. This needs a much more complex economic evaluation as compared to dedicated hydrogen and dedicated electricity producing plants and opens room for significant cost reductions. R&D needs for HTE will surely capitalize from the solid oxide (high temperature) fuel cell development having some technology elements in common.

With respect to thermochemical water splitting cycles, the processes which are receiving presently most attention are the sulfur-iodine, the Westinghouse hybrid, and the calcium-bromine (UT-3) cycles. Efficiencies of the S-I process are in the range of 33-36 %, if operated at 950°C which is judged as a feasible upper temperature limit for the reactor and related heat transfer devices. Under the same conditions, the Brayton cycle may reach ~50 % and could be combined with electrolyzers having a conversion ratio > 90 %. Thus, this process will have an overall efficiency of > 45 % depending on the electrolyzer performance.

Process optimization and material qualification still require considerable R&D efforts beyond 2015 with regard to the potential of higher efficiencies and more compact chemical reactors to be optimized for commercial use. The S-I process will benefit from the medium-term progress in HTGR and materials technology as well as from non-nuclear activities on thermochemical water splitting (e.g., by solar energy) thus remaining a long-term option for commercial applications and a typical Gen IV R&D objective. Special R&D attention in Europe should be given to the high temperature reaction process of sulfuric acid splitting common to all cycles of the sulfur family with a first major milestone to develop technical specifications and construct a 10 MW H₂SO₄ decomposer thus supporting the Japanese development.

But also alternative processes should be taken into account in the future, e.g., those that make use of the steam reforming technology in a closed water splitting cycle (e.g., hydrocarbon hybrid cycle). The study of other heat transfer media like molten salts or liquid metals may help to adopt the chemical processes better to HTGR applications.

For the evaluation of efficiencies, both thermochemical processes and electricity generation in combination with electrolysis are finally governed by the Carnot law meaning that operational temperatures of the process and the heat source should be as high as technically feasible. It makes HTGRs more appropriate than other reactor types, because heat requirements ideally fit with heat available from the reactor.

Economic operation requests low electricity cost. Coupling to HTGRs is feasible and possesses optimization potential towards allothermal high temperature electrolysis and more efficient combined cycle HTGR designs. In industrial processes, energy supply is of utmost importance. With respect to the large variation in the quantity of energy demand and to the wide spectrum of operational parameters, small modular-type sizes and a flexible design are necessary to meet customers' requirements. Also the security of energy supply is essential demanding a very high degree of reliability and availability. Particularly in large establishments, the supply of heat/steam must be highly reliable with basically not more than a month of maintenance within a 5-yr operation period for the petrochemical and refining industries. This can only be ensured by sufficient reserve capacity and which makes a modular arrangement of electricity or heat producing plants appropriate. Also the demand of high reliability for desalination plants will be high, but lower compared to process heat. Methanol, as an example, is a convenient energy carrier to handle.

If the CO₂ needed for its synthesis is taken from other processes where it was released (“double use of CO₂”), CO₂ emissions could be further reduced.

The coupling of a nuclear reactor to a hydrogen production plant located in a chemical complex requires special attention with regard to safety, regulatory aspects, and licensing. There is a need for nuclear process heat reactors to have a common approach, within the EU and the GIF, respectively, of safety issues related with hydrogen such as explosions and fires, confinement and limits of contaminants (e.g., tritium), as well as reliable isolation of both nuclear and chemical plants. Current safety requirements, basically oriented to LWR nuclear plants, must be modified to Gen-IV and new reactors prone for hydrogen production which are inherently safe, reliable, and simple to operate. It also includes a set of new EU codes and standards applicable to the design, materials, fabrication, inspection, and quality of nuclear and hydrogen systems and components operating under these new conditions. This common EU regulatory frame work and standards will help the industrial deployment, coordinate licensing process, and reduce the uncertainties to obtain agreement by national and local authorities of the hydrogen/nuclear facilities.

Technical and economical feasibility, however, remains to be demonstrated, since production processes have not yet been tested beyond pilot plant scale. A new, perhaps revolutionary nuclear reactor concept of the next generation will offer the chance to deliver besides the classical electricity also non-electrical products such as hydrogen or other fuels (e.g., methanol). In a future energy economy, hydrogen as a storable medium could adjust a variable demand for electricity by means of fuel cell power plants (“hydricity”) and also serve as spinning reserve. Both together offer much more flexibility in optimizing energy structures (e.g., substitution of natural gas fired peaking plants by hydrogen). Prerequisites for such systems, however, would be competitive nuclear hydrogen production, a large-scale (underground) storage at low cost as well as economic fuel cell plants. The utilization of oxygen (rather than air) co-produced in the water splitting processes, would improve the efficiency of fuel cell operation significantly, thus further reducing cost.

In China, France, Japan, Korea, and the USA, ambitious programs have been started within the GIF initiative with the main objective to bring nuclear hydrogen production to the energy market. Numerous institutions are active in the first stage of demonstrating the viability of nuclear hydrogen production to be followed by the stages of performance testing and demonstration of the pursued technologies. The European Union does not have a dedicated nuclear hydrogen production project. Here, the respective engagement by research, industry, and policy is mainly given by the participation in activities within the Framework Programmes (FP) of the EU and several national R&D programmes.

A “Quick Start” initiative was launched by the EC resulting in 16 contracts on hydrogen and fuel cells with approx. 100 million Euro of EU funding (to be matched by corresponding private funding). Three of the awarded contracts are dealing with the production of hydrogen: HYTHEC has the objective to evaluate the potential of thermochemical processes focusing on the S-I and HyS cycles with nuclear and solar to be considered as the primary energy sources. The objective of CHRISGAS is the development and optimization of an energy-efficient and cost-efficient method to produce hydrogen-rich gases from biomass for the purpose of obtaining either commercial quality hydrogen or synthetic liquid fuels. In Hi2H2, it is proposed to develop a high temperature water electrolyzer with very high electrical efficiencies.

Another project is HYSAFE conceived as a Network of Excellence. The main objective will be to strengthen, integrate and concentrate existing capacities and fragmented research efforts aiming at the removal of safety-related barriers to the large-scale introduction of hydrogen as an energy carrier. By harmonizing methodologies for safety assessment, the focus is on studies of fire and explosion safety, mitigating techniques, and detection devices. In this way, the network contributes to promoting public awareness and trust in hydrogen technology.

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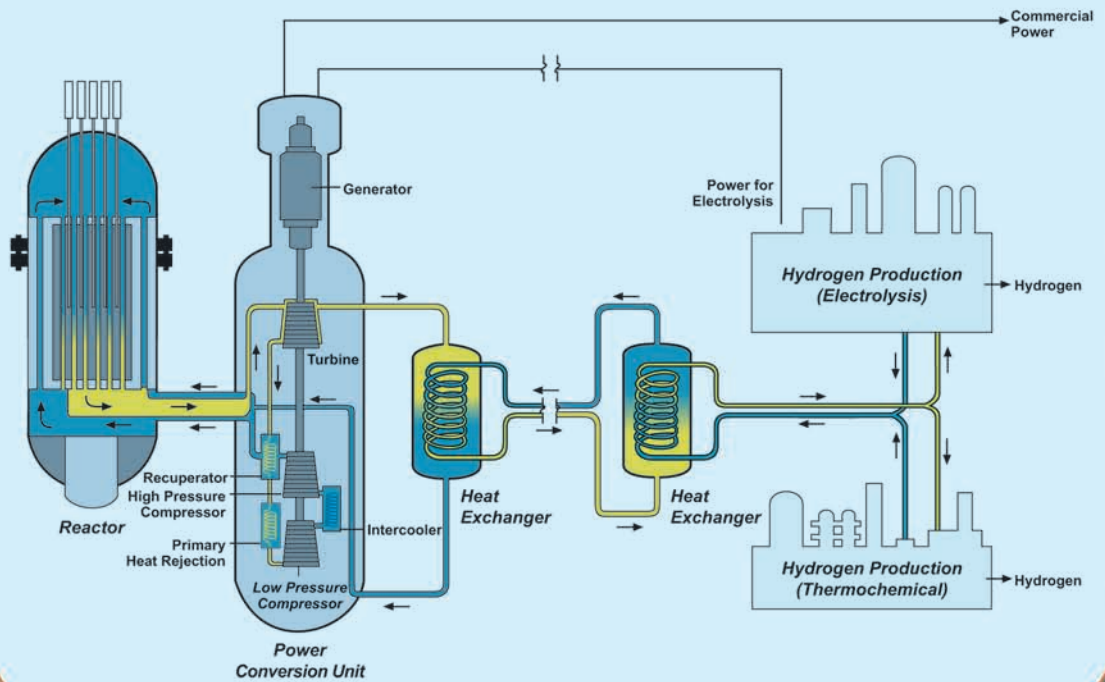
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