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Design and evaluation of heat utilization systems for the high temperature engineering test reactor

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FOREWORD

The Co-ordinated Research Project (CRP) on Design and Evaluation of Heat Utilization Systems for the High Temperature Engineering Test Reactor was established by the IAEA to foster international co-operation in the research and development of applications for nuclear process heat from the high temperature gas cooled reactor (HTGR). This CRP was initiated following the recommendation of the International Working Group on Gas Cooled Reactors (IWGGCR).

The IAEA has co-ordinated an extensive programme addressing the technical development of advanced gas cooled reactor technology. This CRP on the application of high temperature nuclear heat complements other recent CRPs which focused on research on the safety of the HTGR. The technical areas within these CRPs included determining the ability of advanced HTGR designs to dissipate decay heat by natural transport mechanisms, the neutron physics behaviour of the core and on the ability of ceramic coated fuel particles to retain fission products under accident conditions.

In support of this CRP, the Japan Atomic Energy Research Institute provided information on the High Temperature Engineering Test Reactor (HTTR) as the test facility for consideration regarding implementation of a test programme. The IAEA is grateful to Japan for providing this information for the purposes of allowing the Chief Scientific Investigators from Member States with national HTGR programmes the opportunity to evaluate the status of high temperature heat process technologies and identify associated research and development needs for future commercial application.

The following Member State national institutions participated in the CRP:

- Japan Atomic Energy Research Institute (JAERI), Oarai, Japan
- Forschungszentrum Jülich (FZJ), Jülich, Germany
- Russian Research Center Kurchatov Institute, Moscow, Russian Federation
- Institute for Nuclear Energy Technology (INET), Tsinghua University, Beijing, China
- National Atomic Energy Agency of Indonesia (BATAN), Jakarta, Indonesia
- Weizmann Institute of Science, Rehovot, Israel
- General Atomics (GA), San Diego, California, United States of America

Development of this report was co-ordinated by T. Nishihara, S. Shiozawa and M. Ogawa, JAERI, with final compilation by H.L. Brey. The IAEA staff member responsible for this publication was J.M. Kendall of the Division of Nuclear Power.

CONTENTS

Chapter 1

INTRODUCTION

Among the nuclear power plants in existence or currently under design, the high temperature gas cooled reactor (HTGR) is unique in the ability to achieve core outlet temperatures approaching 1000°C. This capability opens a wider spectrum of industrial applications for the utilization of nuclear fission as a broad based energy source. Currently, nuclear energy produces approximately 17% of the world's total electrical generation. Overall, about 30% of the world's primary energy consumption is used for electricity generation, approximately 15% is used for transportation and the remaining 55% is converted into hot water, steam and heat. This indicates that the potential for applications of nuclear energy in the non-electric sector may be quite large, although currently only a few nuclear plants are used for non-electric applications [1].

It was with the intent to develop and demonstrate high temperature applications of nuclear power that the Japan Atomic Energy Commission recommended the construction of the High Temperature Engineering Test Reactor (HTTR). Construction of the helium cooled 30MW(th) HTTR began in March 1991, at the Oarai Research Establishment site of the Japan Atomic Energy Research Institute (JAERI).

It was during this time that JAERI offered the HTTR to Member States of the International Atomic Energy Agency (IAEA) as a test facility for international cooperation in the design and evaluation of heat utilization systems for demonstration while connected to the HTTR. Subsequently, in 1994, the IAEA established the Coordinated Research Programme (CRP) on Design and Evaluation of Heat Utilization Systems for the HTTR.

Member States participating in this CRP included China, Germany, Indonesia, Israel, Japan, Russia and the United States of America. The CRP addressed the following heat utilization systems:

- Steam reforming of methane for the production of hydrogen and methanol
- Carbon dioxide reforming of methane for the production of hydrogen and methanol
- Thermochemical water splitting for hydrogen production
- Combined coal liquefaction and steam generation
- High temperature electrolysis of steam for hydrogen production

JAERI's objectives for the HTTR included the establishment and upgrading of the HTGR technology basis, the conduct of innovative basic research on high temperature engineering and the demonstration of high temperature heat applications of nuclear energy [2]. The goals of this CRP complemented these objectives and included: a.) Defining the R&D needs remaining for the above mentioned heat utilization systems prior to coupling to the HTTR, b.) Defining the goals of the utilization systems for demonstration with the HTTR, c.) Preparation of design concepts for coupling selected systems to the HTTR and performance of associated safety evaluations, and d.) Checking of the licensibality of selected systems under Japanese requirements and conditions.

In addition to the heat utilization systems indicated above, testing of advanced intermediate heat exchangers and coupling the HTTR to the gas turbine for the generation of electricity were examined under the CRP.

This TECDOC documents the activities of Member State participants in the CRP and provides detailed analyses associated with each heat utilization system including:

- the determination of the priority for its connection to the HTTR[l];
- the description of the system process and engineering design, modes of operation and system safety requirements;
- identification of goals to be achieved from the demonstration with the HTTR;
- the experiences associated with the system by Member States in conjunction with their individual national gas cooled nuclear reactor programmes.

A critical analysis of the differences between the use of conventional heat sources such as fossil fuels and the HTGR related to the environment, economics, and other energy and technological considerationswas also conducted.

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Chapter 2

HTGR HEAT SOURCE AND UTILIZATION

The HTGR is the only nuclear energy source, which can produce temperatures higher than 950°C for different process heat applications, such as coal refinement and steam reforming of natural gas. Through refining the fossil energy carriers and thereby producing benign secondary energy carriers, e.g. hydrogen and methanol, with high process efficiency, it is possible to make a contribution for the atmospheric reduction of CO_2-emission and thereby to help address this global environmental issue. Moreover, the modular HTGR concepts under development offer the prospect of cost effective energy production at smaller unit size due to the simplifications arising from an emphasis on passive safety features. This supports a better match to local industrial process heat loads, and simplification of operational safety requirements makes the concept better suited to a process industry environment.

2.1. DIFFERENCES IN THE USE OF CONVENTIONAL AND HTGR HEAT SOURCES

The conventional heat sources for the chemical process applications are based on the use of furnaces, which burn the products of these processes or some other fossil energy carrier e.g. natural gas or oil. An example is the reformer of the ammonia technology of the UHDE-company in Germany (Figure 2.1), whereby the burners are arranged at the ceiling of the furnace and the flue gas heats the reformer tube along it's entire length primarily through thermal radiation. The temperature of the flue gas at the top of the furnace is ~ 1800°C and > 1,000°C at the bottom. Figure 2.2 shows the temperature profiles of the reformer tube as well as that of the process fluid along the heated length of the tube. The maximum heat flux in the range of about 65 kW/m^2 prevails near the inlet of the reformer tube. This is located in the upper part of the furnace and results in the production of the maximum temperature gradient in the reformer tube. However, during the part load conditions intensive cooling of the reformer tube via cold process fluid takes place in this area and a similar temperature profile also prevails. In the lower part of the reformer tube, where equilibrium of reaction is approached, the heat flux has been reduced to the value of about 15 kW/m^2. Some furnaces are designed with the arrangement of wall fired burners, instead of top fired, so that an average heat flux prevails along the heated reformer tube length.

Utilising the HTGR as the heat source provides hot helium from the core outlet, which flows through the reformer tube bundle in counter flow to the process gas flow. This allows for the transfer of its heat via endothermic chemical process through heat convection between the two fluids and through heat conduction in the reformer tube wall, as shown in Figure 2.3 of the test reformer bundle of Steinmüller-company in Germany [2]. However the average heat flux is generally lower in this case because of the higher flue gas temperatures available with the conventional steam reformer. With the HTGR as the heat source, an average value of heat flux between 40 and 60 kW/m^2 can be realized in the steam reformer [3] as well as in other relevant heat transfer components.

Operation of the German AVR experimental HTGR over a span of twenty years resulted in consistently achieving a helium temperature of 950°C at the core outlet, thereby generating electricity with a steam turbine under conventional live-steam conditions. Good experiences were also obtained from the operation of HTGR plants in different countries of the world, however up till now the heat of HTGR has only been used for electricity production with the steam turbine cycle.

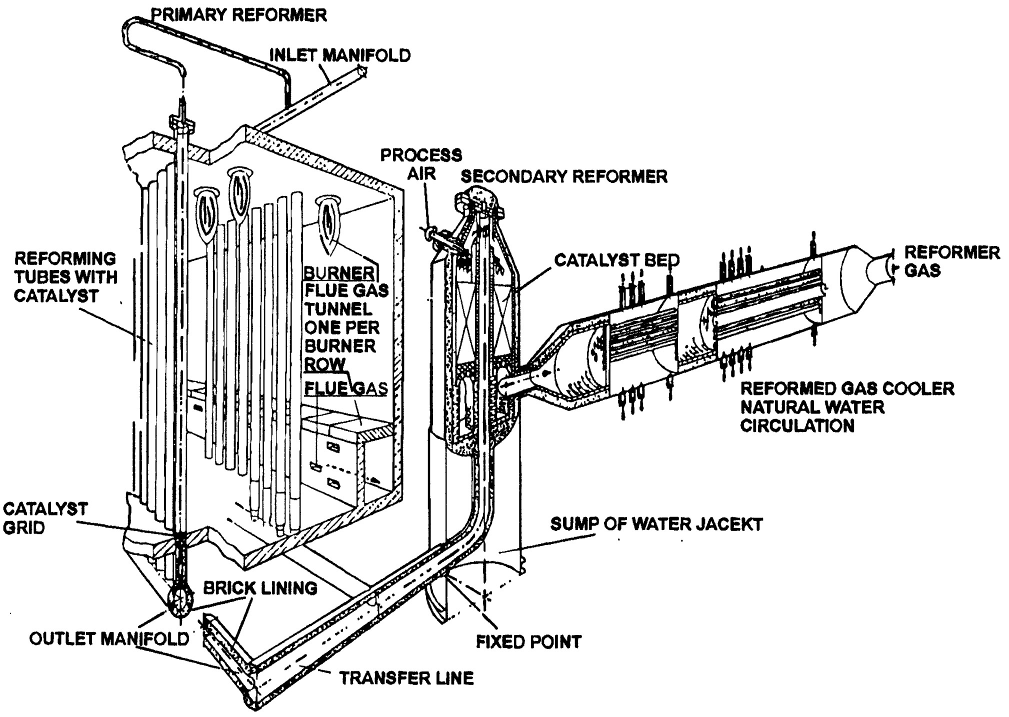


FIG. 2.1: UHDE ammonia technology, reformer section and gas cooling.

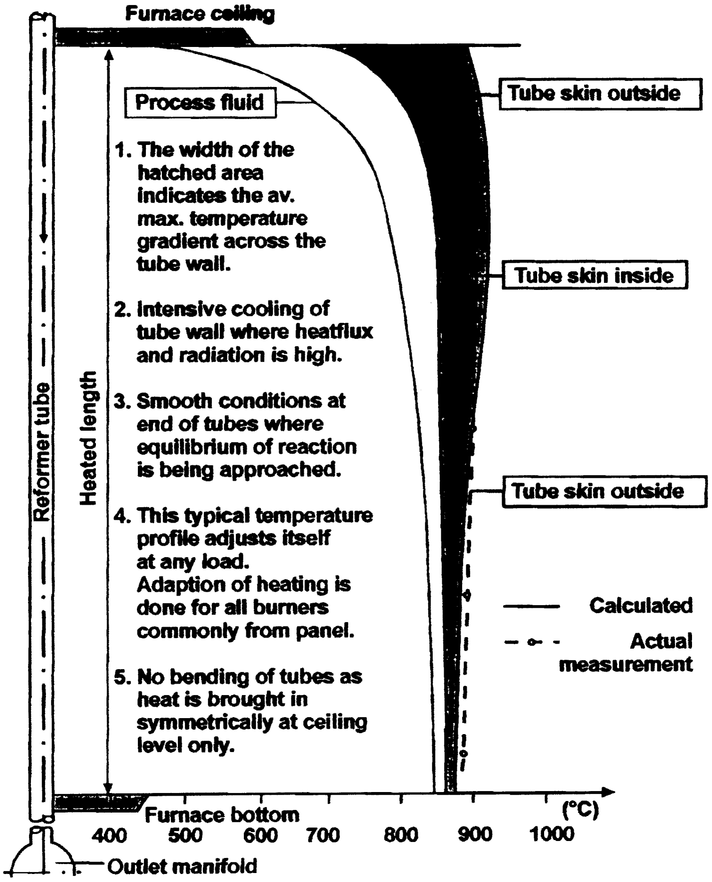


FIG. 2.2: Temperature profiles for top fired reformer, UHDE ammonia technology.

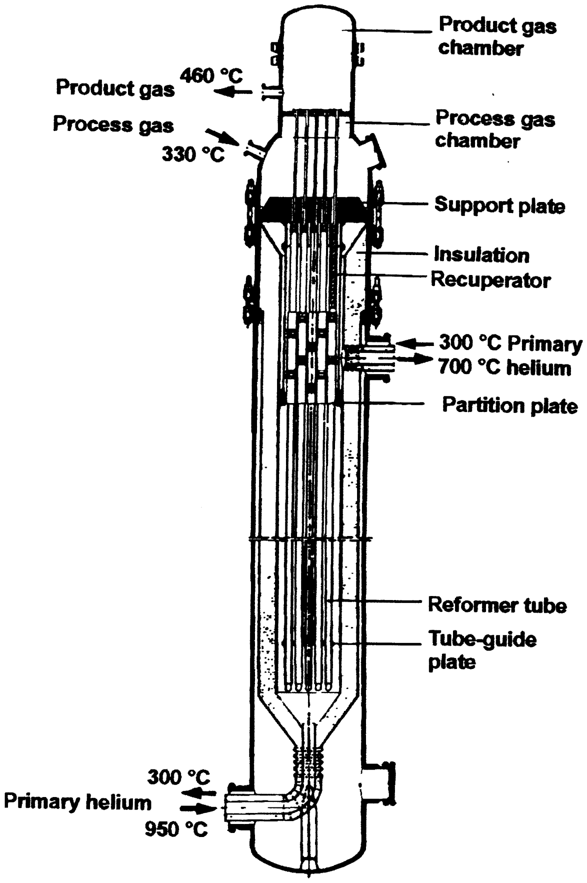
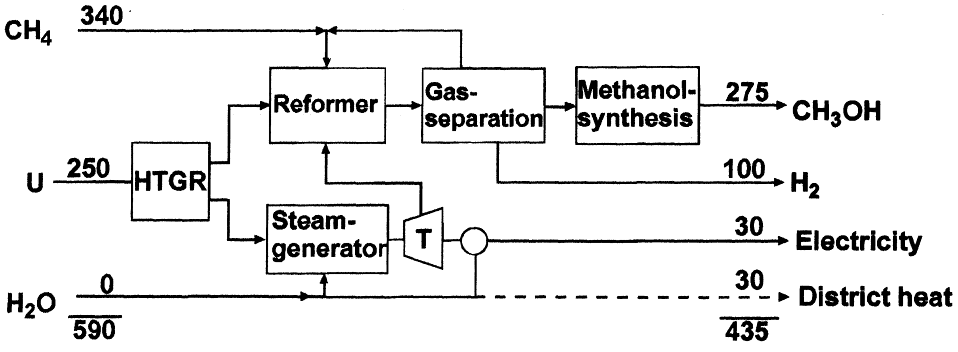


FIG. 2.3. Test steam reformer (5 MW).

2.1.1 Advantages and disadvantages of HTGR heat sources

The potential exists for the HTGR heat source to support a multitude of applications including different chemical processes in the high temperature range, electricity generation in the high and middle temperature range and district heat in the low temperature range. This multiple utilisation of the HTGR can result in the attainment of very high system efficiency. The process for the production of methanol and hydrogen with steam reformer is shown in Figure 2.4, whereby an efficiency of ~ 74% has been achieved. At the same time with this process, the fossil energy carrier has been converted to a benign secondary energy carrier with a conversion rate of about 110 % [4].

The surplus hydrogen from this process can be used with the addition of CO_2 to increase the productivity of methanol. The required CO_2 can be provided from the burning of the fossil energy carrier or CO_2-rich natural gas, e.g. from the Natuna-gas field of Indonesia. This gas can be used directly in the steam reforming of methane to optimize the composition of synthesis gas produced from this process, and for its further synthesis to methanol, as described in detail in the following chapters. A further advantage for the use of this CO_2 is to also improve the global environmental issue.



$$\eta = \frac{435}{590} = 74\% \qquad c = \frac{275 + 100}{340} = 110\%$$

all data are given in MW

FIG. 2.4. Process of the production of methanol and hydrogen [4].

Under the consideration of the aspects of high heat utilization, optimal design-concept of the steam reformer and economic competitiveness of it's products, analyses were performed of different design parameters resulting in changes to the system pressure of the process gas and the equilibrium reforming temperature. The following conditions of the primary helium side have been adopted for further analyses:

Reformer power	70 MW
Mass flow	50,3 kg/s
Inlet temperature	950°C
Outlet temperature	680°C
System pressure	40 bar.

Moreover the feedgas with a temperature of 560°C enters the catalyst region of the reformer with the following composition:

CH_4: 19,77 %

C_2H_6: 0,17 %

CO: 0,02 %

H_2O: 80,05 %

These conditions remain unchanged for further studies.

Compositions of the product gas for an equilibrium reforming temperature of 800°C with different process gas exit pressures are provided in Table 2-1. Compositions and mass flow of the product gas for a process gas exit pressure of 41.9 bar and with different equilibrium reforming temperatures are provided in Table 2-2 [5]. By these analyses, the equilibrium reforming temperature is ~ 10°K lower than the exit temperature of the product gas out of the catalysts region.

Table 2.1. Composition and volumetric flow of the product gas with different process gas pressures [5]

Composition		**Exit pressure of product gas**	
		41,9 bar	20 bar
CH_4	Vol %	5,38	2,83
H_2	Vol. %	37,35	43,06
CO	Vol. %	4,53	6,10
CO_2	Vol. %	5,98	6,23
H_2O	Vol. %	46,76	41,77
Reaction enthalpy	kJ/kg	1554	1933
Mass flow	kg/s	37,2	31,1
Volume flow (process gas)	m^3(STP)/s	47,3	39,5
Volume flow (product gas)	m^3(STP)/s	59,8	52,4
Conversion (CH_4+C_2H_6)	%	66,1	81,3

By reducing the system pressure of the process gas, the methane conversion rate can be increased, whereby the cost of the refinement of the synthesis gas can be reduced to achieve the required product, hydrogen or methanol. However, in this case, the volume flow of the H_2 and CO is not considerably increased, because of higher reaction enthalpy.

Table 2.2. Composition and Mass flow of the product gas with different equilibrium reforming temperatures [5]

Reforming Temp, (°C)		800	825	850	860
Composition of product gas					
CH_4	Vol. %	5,38	4,36	3,42	3,08
H_2	Vol. %	37,35	39,45	41,31	41,97
CO	Vol. %	4,53	5,36	6,17	6,48
CO_2	Vol.. %	5,98	5,89	5,74	5,68
H_2O	Vol. %	46,76	44,95	43,36	42,79
Conversion (CH_4+C_2H_6)	%	66,1	72,0	77,6	79,8
Reaction enthalpy	kJ/kg	1554	1710	1859	1915
Mass flow	kg/s	37,2	34,4	32,1	31,3

The total volume flow has to be decreased with the same heat source capacity. Moreover, the investment costs of the steam reformer has to be increased with the required thicker walls of the reformer tubes in order to compensate for the stresses caused due to the higher pressure difference between the primary helium and secondary process gas. Therefore, the pressure decrease of the process gas has no economical advantage.

With a constant helium inlet temperature of 950°C, the equilibrium reforming temperature can be increased only through the increase of heat transfer area of the reformer tubes. Also in this case, with the increase of the equilibrium reforming temperature the product gas mass flow has to be decreased with the same capacity of the heat source, because of the higher reaction enthalpy. Therefore, the economical advantages cannot be realized by these analyses. Only with the use of better materials or through enhancement of the heat transfer characteristics or through the increase of helium inlet temperature, can the economic advantage of an increase of methane conversion rate be achieved.

To achieve the goal for higher efficiencies by the chemical conversion processes, higher process temperatures are required. Moreover, the heat utilization components, which are the barrier between the primary and secondary circuits such as steam reformer, must fulfill the safety requirements during normal operation and also in accident conditions. Any leakage of the primary helium outside the reactor building must be avoided. Therefore, the walls of the heat transfer components should be built from very good high temperature resistant materials. These should have good properties in regard of creep behaviour, fatigue properties, structural stability and corrosion resistant. Further, through design criteria, the primary and secondary stresses by these components should be kept low.

Commercially available alloys, INCONEL 617, HASTELLOY X and INCOLOY 800H, have been tested in different experimental facilities under operational conditions with good results. However, for long term use of the steam reformer for ~ 100,000 hours, the best results have been achieved by INCONEL 617, which has a higher investment material cost. To realize a higher helium temperature than 950°C, new materials such as ceramics will have to be developed for future heat utilisation components.

Different studies have shown that the economic competitiveness of the nuclear process heat plant has improved through different potentials already applied in the refinement of coal, and it is comparable to non-nuclear alternatives [5]. However the economic competitiveness of the coal refinement in total in comparison to the present market conditions has not existed since the end of the oil price crisis. Similar investigations have also been performed for the conversion of natural gas to methanol e.g. with the gas of the Natuna gas field in Indonesia [6]. The highest potential for economic competitiveness of this gas field is realised with the main product being methanol as a substitute for gasoline and diesel. However, the capital costs of the HTGR heat source plays a dominant role in this economic evaluation. The cost of this process needs to be improved for future deployment in order to justify the introduction of the HTGR in the heat market parallel to electricity generation. However, it is the only nuclear heat source which can supply heat in the higher temperature range of 900° to 1100°C.

Also, this nuclear heat source has significant safety characteristics which would allow its construction directly near the heat market. Under all possible cases of accident, there is no appreciable radiological consequences which can occur outside the plant i.e. no immediate or delayed fatalities, no evacuation, no relocation and no changes in eating and drinking habits. Therefore, in the future, ‘catastrophe-free’ nuclear energy is expected to be realized with the design of HTGR [7]. This NPP should also provide the benefit of not causing risk to the surrounding investments associated with the heat utilization plant. The HTGR plant size can be realized compatible with the consumer demand, so that customers and consumers with smaller financial resources are in position to use such plants.

The HTGR heat source also has advantages in comparison with fossil energy carriers with respect to the global environmental issue, particularly because of it's potential to reduce CO_2-emission. Moreover, if the HTGR can be utilized for co-generation applications including process heat for industry in parallel to electricity generation, it can further provide for the environmental attribute of a reduction in CO_2-emission. At present, the energy demand in the total world is increasing, although it is stagnant in some of the industrialized countries.

The industrialized countries, with a population of only 25 % of the world, produce almost 75 % of the total CO_2-emission in the world. The average CO_2-emission of these countries is ~ 3,6 tC/y per inhabitant in comparison with only 0,4 tC/y per inhabitant for the remaining 75% of the world population [8]. Therefore, according to the goal of the world energy conference, the CO_2-emission in these countries has to be reduced dramatically by the factor of ~ 4 in the coming 20 to 50 years. This can be achieved only if these countries produce and consume their energy through the use of non-fossil energy carriers. The HTGR can play a dominant role for the world energy supply system in the future because of its positive safety and high temperature operational characteristics. This energy source will also be enhanced economically if future additional taxes are assessed on the emission of CO_2.

Positive experiences gained through past operation of HTGR plants has encouraged utilities, particularly in South Africa, to establish new related nuclear technology. Cost savings are achievable due to the excellent safety of these plants as well as through their series fabrication, and the economic competitiveness of their associated products is expected to be realized with future development [9].

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Chapter 3

HIGH TEMPERATURE ENGINEERING TEST REACTOR (HTTR) [1, 2]

3.1. DESIGN OF THE HTTR

The major design specification of the HTTR is given in Table 3-1. As shown in this table, the reactor outlet coolant temperature at full power is set at 850°C and 950°C. The reactor operational mode at 850°C is called "rated operation" and the mode at 950°C is the "high temperature test operation". This is due to the requirement that the HTTR is not allowed to be operated at 950°C for the full life of the initial core. Furthermore, some of the test themes such as the safety demonstration tests and irradiation tests are allowed only at the rated operation. The high temperature nuclear process heat utilization system will be operated within the high temperature test operational mode. The design life of the permanent structural components in the HTTR plant is based on 20 years, with a load factor of 60% of full power operation.

Table 3-1. Major design specifications of the HTTR

Thermal power	30 MW
Coolant	Helium
Core outlet coolant temperature	850°/950°C
Core inlet coolant temperature	395°C
Fuel	Low-enriched UO_2
Fuel element type	Prismatic block
Direction of coolant flow through the core	Downward
Pressure vessel material	Steel
Number of main cooling loops	1
Heat removal system	Pressurized water cooler
	Intermediate heat exchanger
Primary coolant pressure	4 MPa
Containment vessel	Steel containment
Plant lifetime	20 years

The HTGR exhibits inherent safety characteristics for accident conditions with respect to the accidental release of fission products. Nevertheless, the HTTR is required to have a containment vessel in order to meet Japanese safety design guidelines which are provided for light water reactors.

The HTTR achieved first criticality on 11 November 1998, with initial power-up tests to be carried out to attain full power operation with a core outlet temperature of 850°C, and further testing to an outlet temperature of 950°C.

3.1.1 System layout

The Oarai Research Establishment is located approximately 100 kilometers north of the Tokyo metropolitan area near the Pacific Ocean (Figure 3.1), with the HTTR plant in the southwestern part of the Oarai site (Figure 3.2). Two nuclear reactors have already been located near the HTTR. One is the Japan Materials Testing Reactor (JMTR), a 50 MW(th) light water cooled and moderated reactor, located approximately 400 m north of the HTTR; and the other is an experimental liquid metal fast breeder reactor "Joyo" of 130 MW(th), approximately 650m east.

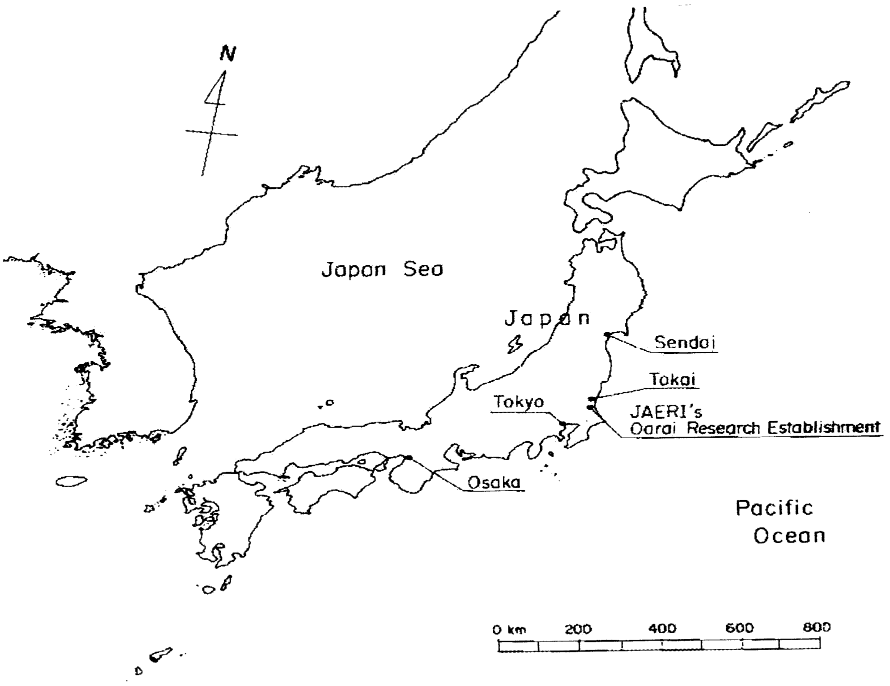


FIG. 3.1. Location of the Oarai Research Establishment of JAERI.

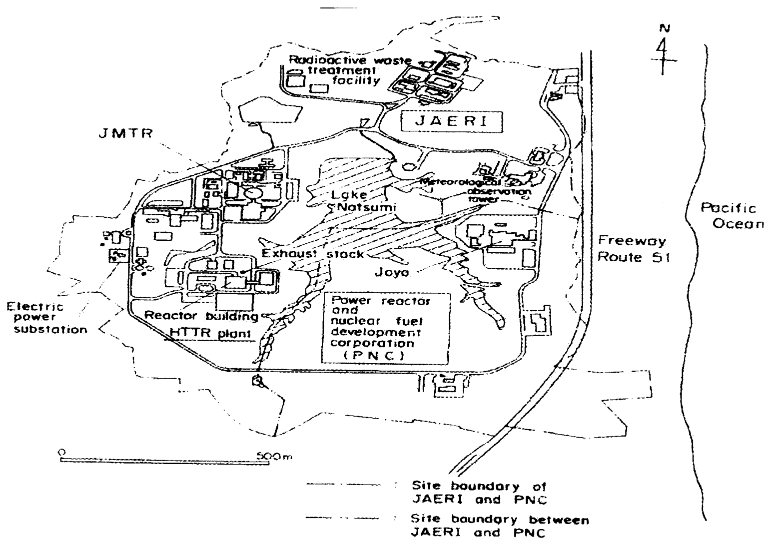


FIG. 3.2. Location of the HTTR within the Oarai site.

As illustrated in Fig. 3.3, the HTTR plant is composed of a reactor building, a spent fuel storage building, a machinery building, cooling towers, an exhaust stack, warehouses, a high temperature process heat utilization system and others. The reactor building of 48 m x 50 m in size is centered in the area of the plant. The exhaust stack (80 m high) is north of the reactor building and provides for the discharge of air ventilated from the reactor building to the atmosphere. A heat utilization system will be constructed on a plain south of the reactor building.

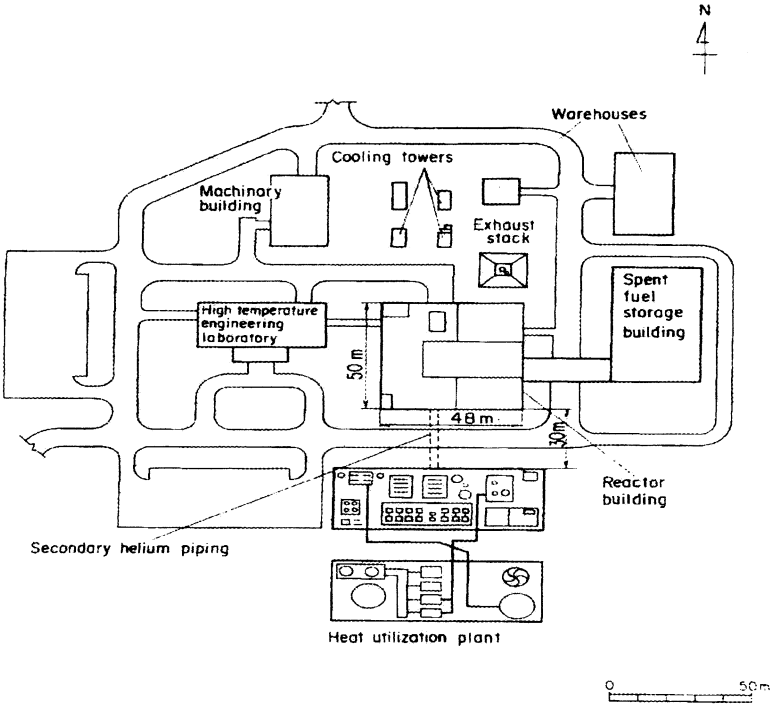


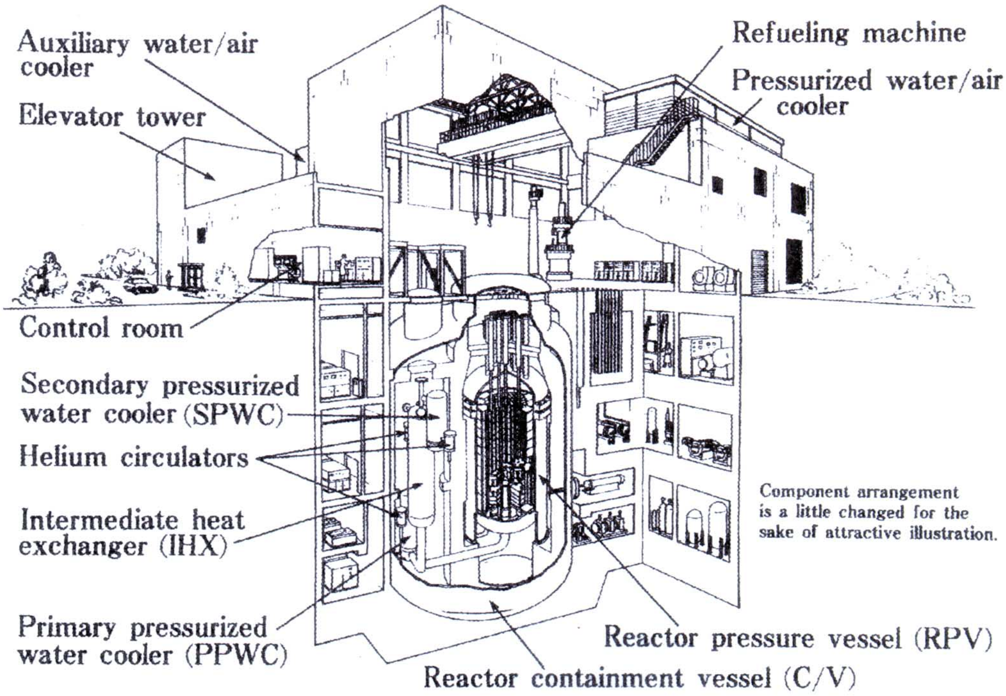
FIG. 3.3. HTTR plant layout.

3.1.2 Reactor building

The reactor building consists of two floors above grade level with and three underground, as shown in Figure 3.4. It is supported on a sand layer formed during the Quaternary era, and was structurally evaluated by boring surveys and shallow reflection seismic surveys that verified the supporting ground is horizontally formed with continuity of strength and stiffness. It was recognized from seismic surveys that the supporting ground at the site has the seismic safety equivalent to base rock.

3.1.3 Reactor containment vessel

A steel reactor containment vessel (C/V) is installed in the center of the reactor building. A refueling hatch is attached to the C/V above the reactor pressure vessel (RPV) at the level of the operating floor. During refueling, the hatch is removed to another location on the operating floor. The major specification of the C/V is given in Table 3-2.



Component arrangement is a little changed for the sake of attractive illustration.

FIG. 3.4. Cutaway view of the HTTR reactor building.

Table 3-2. Main design specifications of the reactor containment vessel

Material	Carbon steel
Maximum allowable pressure	0.49MPa
Maximum allowable temperature	150° C.
Leak rate	0.1%/day
Size	
Inner diameter	18.5 m
Thickness	30 mm
Free Volume	2,800 m^3

The primary functions of the C/V include:

- To contain fission products (FPs) as one of multiple barriers against accidental FP release into the atmosphere, and
- To limit the amount of air which may ingress into the core and react with the core graphite in the event of a primary pipe rupture accident.

The HTTR has four distinct barriers; (a) fuel coatings, (b) RPV, (c) C/V and (d) the reactor building. The double containment concept (RPV + C/V) was applied to the HTTR, because the safety features of an HTGR were not developed in Japan at the time of licensing the HTTR.

Some compartments surrounding the C/V in the reactor building serve as a confinement e.g. the service area. The service area is maintained at a slightly negative pressure with respect to atmosphere by an air conditioning system during both normal operation and accident conditions. The barriers of the C/V and the service area in the reactor building drastically reduce the off-site radiation dose in an accident such as a primary pipe rupture. Major components, including the primary cooling system as well as the RPV, are contained within the C/V.

3.1.4 Reactor pressure vessel

The reactor pressure vessel (RPV) consists of a vertical cylinder, hemispherical top and bottom head closures and 31 standpipes which are categorized as "control rod (CR) stand-pipes", "irradiation stand-pipes", and standpipes for instrumentation, etc. The top head closure is bolted to a flange on the vessel cylinder as shown in Figure 3.5.

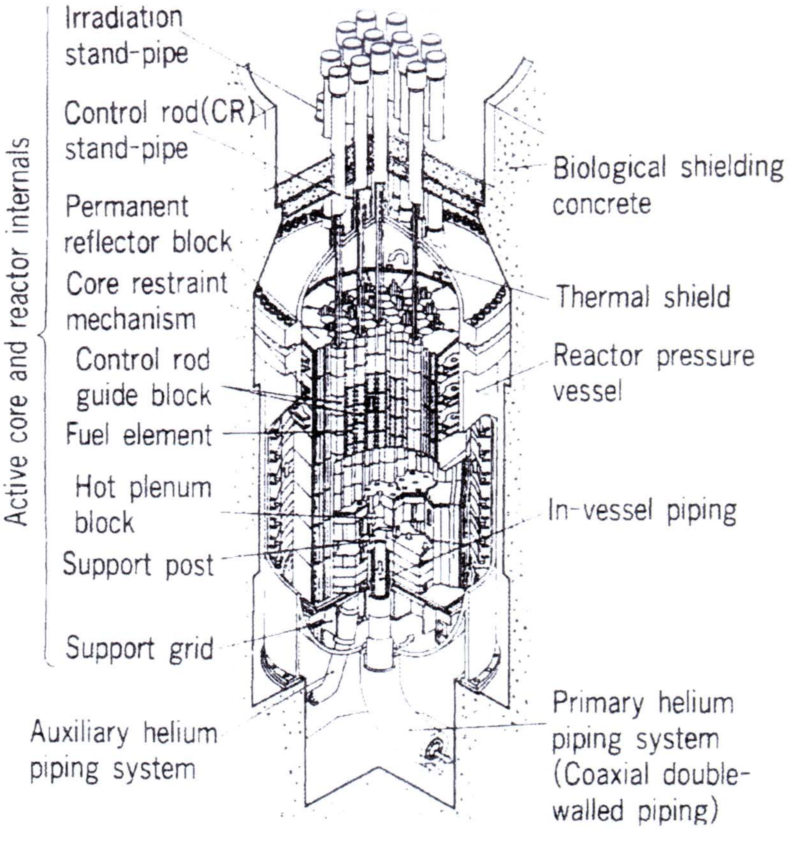


FIG. 3.5. Reactor pressure vessel and internals.

The thermal shield is attached to the inner surface of the top head closure to prevent the closure from overheating during a depressurization accident consisting of a primary helium pipe rupture. The RPV design specifications are provided in Table 3-3.

Table 3-3. Major design specifications of the RPV

Material	2.25Cr-1Mo steel
Design pressure	4.81 MPa
Design temperature	440° C
Size	
Inner diameter	5.5 m
Thickness	120 mm
Height	13.2 m

3.1.5 Core

The active core is arranged as a right circular cylinder 2.9 m in height and 2.3 m in equivalent diameter, consists of hexagonal fuel element blocks and graphite guide hexagonal blocks. The active core is formed with 30 fuel columns and 7 CR guide columns, where the term "column" is one row of the blocks piled axially. 12 replaceable reflector columns, 9 CR guide columns and 3 irradiation test columns surround the core, as shown in Figure 3.6. The permanent reflector blocks are tightened by the core restraint mechanism.

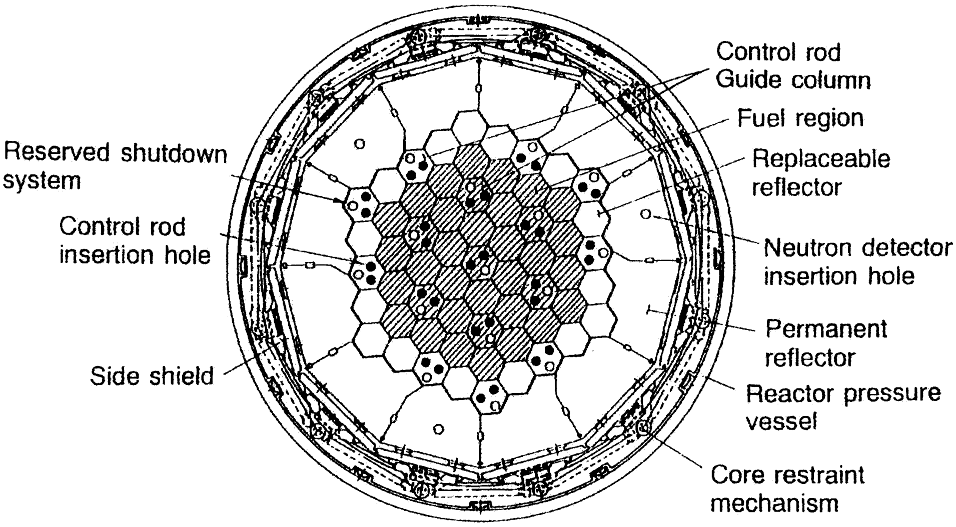


FIG. 3.6. Horizontal arrangement of the core.

3.1.6 Fuel

The fuel elements of the HTTR are of a “pin-in-block” type with each fuel element consisting of fuel rods and a fuel block, 360 mm in width across the flats and 580 mm in length, as shown in Figure 3.7.

The fuel block has three dowels on the top and three mating sockets at the bottom for alignment. Tri-isotropic-coated fuel particles with kernels of UO_2, approximately 6 wt% of average enrichment and 600 μm in diameter, are dispersed in a graphite matrix and sintered to form a fuel compact. Fuel compacts are contained in a fuel rod of 34 mm outer diameter and 577 mm in length. Fuel rods are inserted vertically into holes in the fuel block. The reactor coolant flows downward through annulus gaps between the hole and rod. The maximum design temperature is 1495° C and maximum burnup is 33,000MWd/t.

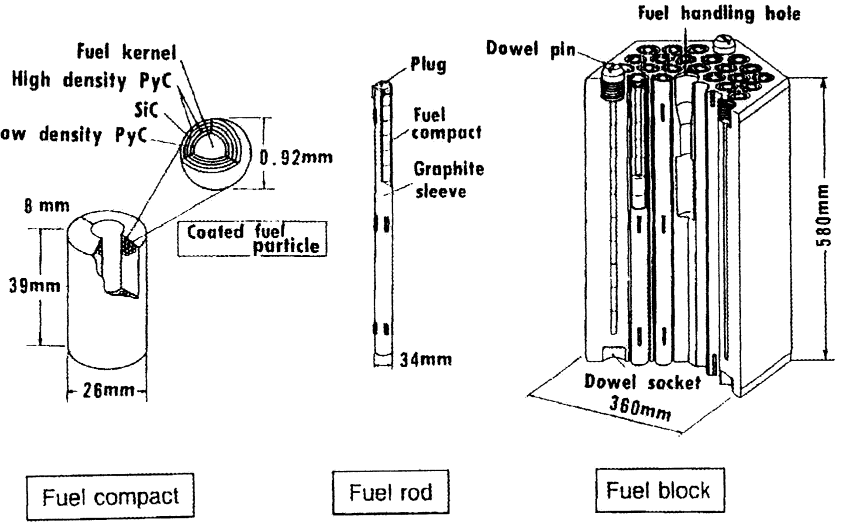
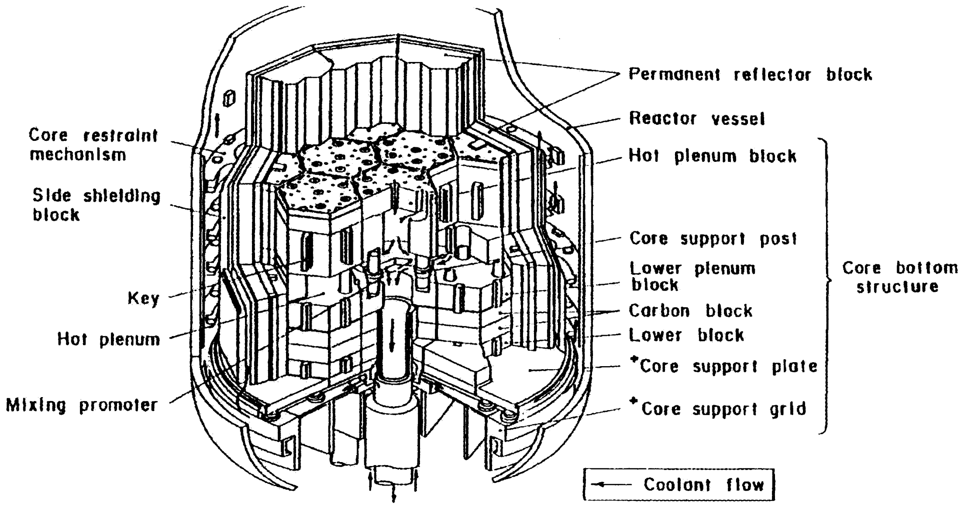


FIG. 3.7. HTTR fuel element.

3.1.7 Reactor internals

The reactor internals consist of graphite, the metallic core support structures and other components as shown in Figure 3.8. The graphite core support structures consist of permanent reflector blocks, hot plenum blocks, support posts, core bottom structures, etc. The metallic core support structures consist of support plates, a support grid, and the core restraint mechanism. They support the active core and replaceable reflector blocks.



* metallic core support structures

FIG. 3.8. Structure of the HTTR core internals.

The permanent reflector surrounding the replaceable reflector is made of large polygonal graphite blocks. These blocks are fixed by key elements and the core restraint mechanism.

The reactor coolant enters the RPV at the bottom from the primary cooling piping system and then flows up through an annulus between the RPV and reactor internals. It then flows down through the core and enters the hot plenum.

The hot plenum and permanent reflector blocks are fabricated from PGX grade graphite, which is a medium-to-fine grained, molded structural graphite. Within the hot plenum block assembly, the hot primary coolant from the core is collected and flows into the pipe inserted in the core bottom block. This is connected to the primary helium piping system in the main cooling system.

3.1.8 Control system

Reactivity is controlled through CRs, which are inserted into the CR guide columns in the active core and the replaceable reflector columns. A reserved shutdown system (RSS) is provided as a back-up shutdown system to the CRs and operates by means of inserting boron carbide/graphite pellets into the third channel in each CR guide column. The neutron absorber is made of B_4C/C, and is sheathed with a cylindrical clad of Alloy 800H.

All pairs of CRs are individually supported by control rod drive mechanisms (CRDMs) which are housed inside the stand-pipes. The CRDM inserts and withdraws a pair of the CRs during normal operation. Upon receipt of a scram signal, the CRs drop into the active core and reflectors by gravity.

During a scram, the sheath temperature reaches approximately 900°C. To prevent the CR sheathes from thermal damage, nine pairs of CRs are initially inserted into the replaceable reflector column holes. The remaining seven pairs of CRs are inserted into the active core column holes after the core is cooled down when the reactor outlet coolant temperature is decreased below 750°C.

3.1.9 Cooling system

The reactor cooling system flow diagram is shown in Figure 3.9. This system is composed of a main cooling system (MCS), an auxiliary cooling system (ACS) and two-reactor vessel cooling systems (VCSs).

The MCS removes the heat energy from the reactor core during the normal operation, while the ACS and VCSs function as residual heat removal systems upon initiation of a reactor scram. With the receipt of a reactor scram, the gas circulators of the MCS stop to prevent the core and heat exchanger tubes from over-cooling. In an anticipated operational occurrence and accident condition when forced cooling of the core is available, the ACS automatically starts up in correspondence with a reactor scram signal. The VCS functions as a residual heat removal system when the forced circulation in a primary cooling system is no longer available due to a rupture of its piping system. This system is also in service during normal operation to cool the reactor shielding concrete wall.

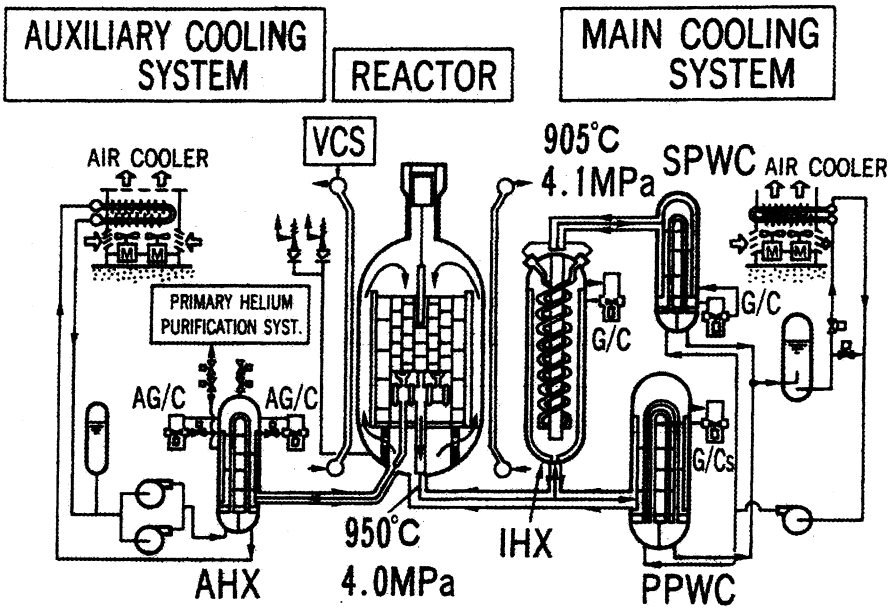


FIG. 3.9. HTTR reactor cooling system.

A brief description of the components and operational requirements for the individual cooling mechanisms which comprise the overall HTTR cooling system are as follows:

- Main cooling system

The MCS consists of an intermediate heat exchanger (IHX), a primary pressurized water cooler (PPWC), a secondary pressurized water cooler (SPWC) and pressurized water air cooler. The MCS has two operational modes; "single loaded operation", and "parallel loaded operation". The PPWC is operated to remove the reactor heat of 30 MW during the single loaded operation, while during the parallel load operation, the IHX removes 10 MW of energy and the PPWC removes 20 MW. The SPWC subsequently removes the heat from the IHX.

The heat removed by the PPWC and the SPWC is transported through water pressurized to 3.5MPa. This water is then cooled by an air cooler. In the HTTR, the reactor heat of 30 MW is eventually transferred to the atmosphere.

During normal operation, the pressure of the secondary helium is controlled to 0.1 MPa higher than that of the primary helium at the IHX heat transfer tubes. This allows a reduction on the pressure load of the tubes and to protect against an accidental leak of radioactive materials into the secondary helium. The water pressure is always controlled at 3.5 MPa so that a large amount of water will not ingress into the core upon a PPWC tube rupture accident.

- Auxiliary cooling system

The ACS consists of an auxiliary heat exchanger (AHX), two auxiliary helium circulators and an air cooler. During normal operation, a small flow of helium (200 kg/h) passes through the AHX to a primary helium purification system so as to remove impurities contained in the reactor coolant, as shown in Figure 3.9. Upon a reactor scram with the reactor coolant pressure boundary intact, the auxiliary helium cooling system automatically starts and transfers the residual heat from the core to the air cooler. The AHX has a heat transfer capacity of ~ 3.5 MW.

- Vessel cooling system

Two vessel cooling systems (VCSs) are provided to prevent the reactor core and the RPV from thermal damage caused by residual heat after a reactor scram when the ACS cannot cool the core. They consist of water-cooled panels surrounding the RPV and two cooling water systems. Each of the systems alone is capable of controlling temperatures of the core and RPV within safe limits.

The heat removal rate from the RPV to the VCS is designed 0.6 MW or less during normal operation so as to effectively allow for the transfer of reactor heat to the MCS as much as possible, and also 0.3 MW or more during an accident to remove the residual heat from the core. The VCS is designated as safety equipment so that there are two independent complete systems which are backed up with emergency power supply. It is in service even during normal operation in order to cool the biological shield concrete wall.

3.1.10 Intermediate heat exchanger

The IHX is a vertical helically coiled counter flow type heat exchanger as shown in Fig. 3.10. To minimize constraints of axial and radial thermal expansions on the helically coiled heat transfer tubes, a floating hot header with a combination of a central hot gas duct has been adopted which passes through the central space inside the helix bundle. An assembled type of tube support allows free thermal expansion of a helix in the radial direction.

The primary helium enters the IHX through the inner pipe of the primary concentric hot gas duct attached to the bottom of the heat exchanger. It flows up outside the tubes thereby transferring the nuclear heat of 10 MW to the secondary helium and then flows back to the annular space between the inner and outer shells. The secondary helium flows down inside the heat transfer tubes and then flows up through the center hot gas passage.

A double-walled shell with thermal insulation attached on the inside surface of the inner shell provides reliable separation of the heat-resisting and pressure-retaining functions. Cold helium flowing through the annulus provides uniform temperature distribution throughout the outer shell which serves the function of a pressure retaining member. Table 3-4 shows the major design specification of the IHX.

Table 3-4. Major design specifications of the IHX

Material	
Shell	2.25Cr-1Mo steel
Tube	Hastelloy XR
Thermal insulation	Kaowool 1400SHA
Design pressures/temperatures	
Design shell pressure	4.81 MPa
Design tube pressure	0.29 MPa
Design shell temperature	430° C
Design tube temperature	955° C
Size	
Shell inner diameter	1.35 m
Shell height	10 m
Tube outer diameter	3.2 mm
Tube thickness	3.5 mm
Tube length	21.5-22.9 m
Number of tubes	96

3.2 MODES OF OPERATION

Along with the modes of "single loaded operation" and "parallel loaded operation", the HTTR has both a rated operation condition and a high temperature test operating condition. With respect to heat utilization, the parallel loaded high temperature test operation is most important. The temperature and pressure distribution in this operational condition are described in this subsection.

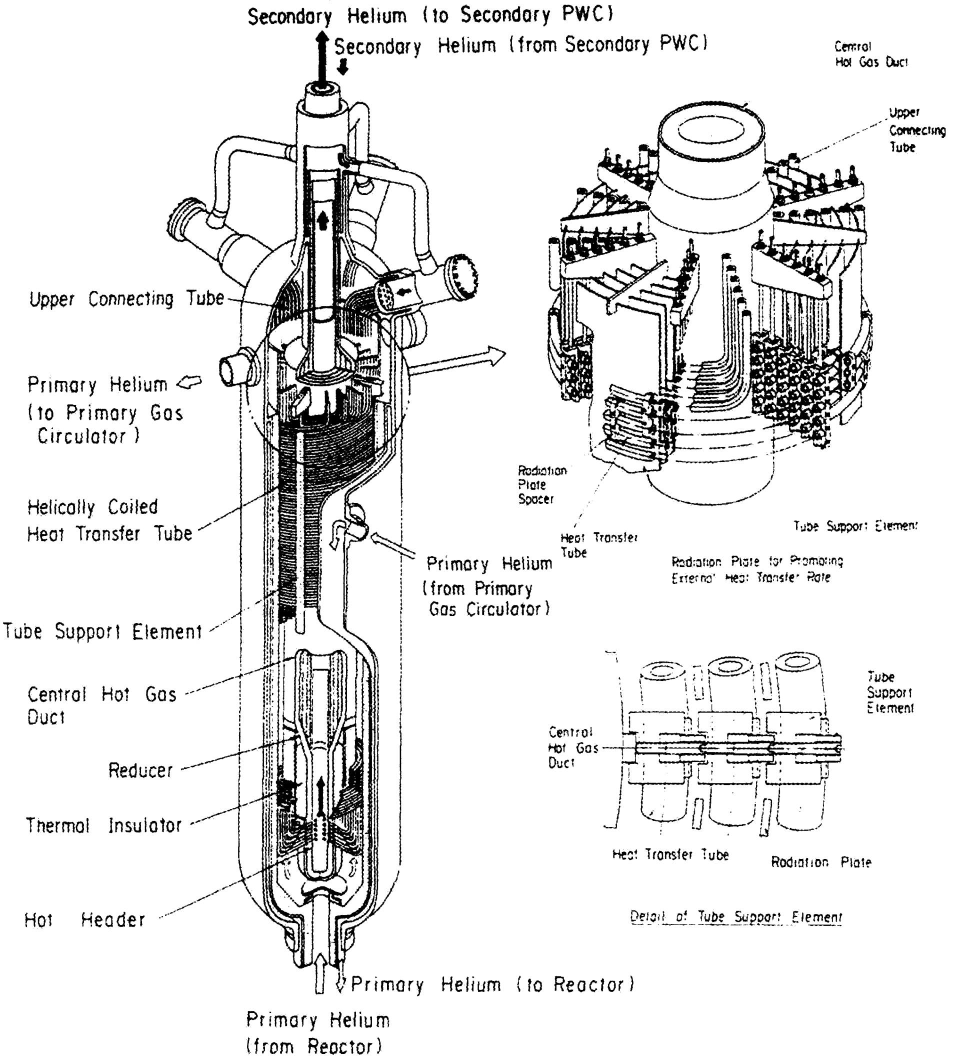


FIG. 3.10. He/He intermediate heat exchanger.

3.2.1 Major technical parameters

A simplified process flow diagram of the MCS in the HTTR is shown in Figure 3.11. This figure shows the coolant flow path and the values of the flow rate, pressure and temperature at key stations in the parallel loaded high temperature test operation mode.

- Primary helium cooling system

As shown in Fig. 3.11, the reactor coolant of 36.7 t/h (10.2 kg/s) in total mass flow is heated to 950° C through the active core in the high temperature test operation mode. A reactor coolant of 36.5 t/h, i.e., 99.5 percent of the total flows into the MCS. The remaining 0.5 percent of the reactor coolant, i.e., 200 kg/h, flows into the ACS.

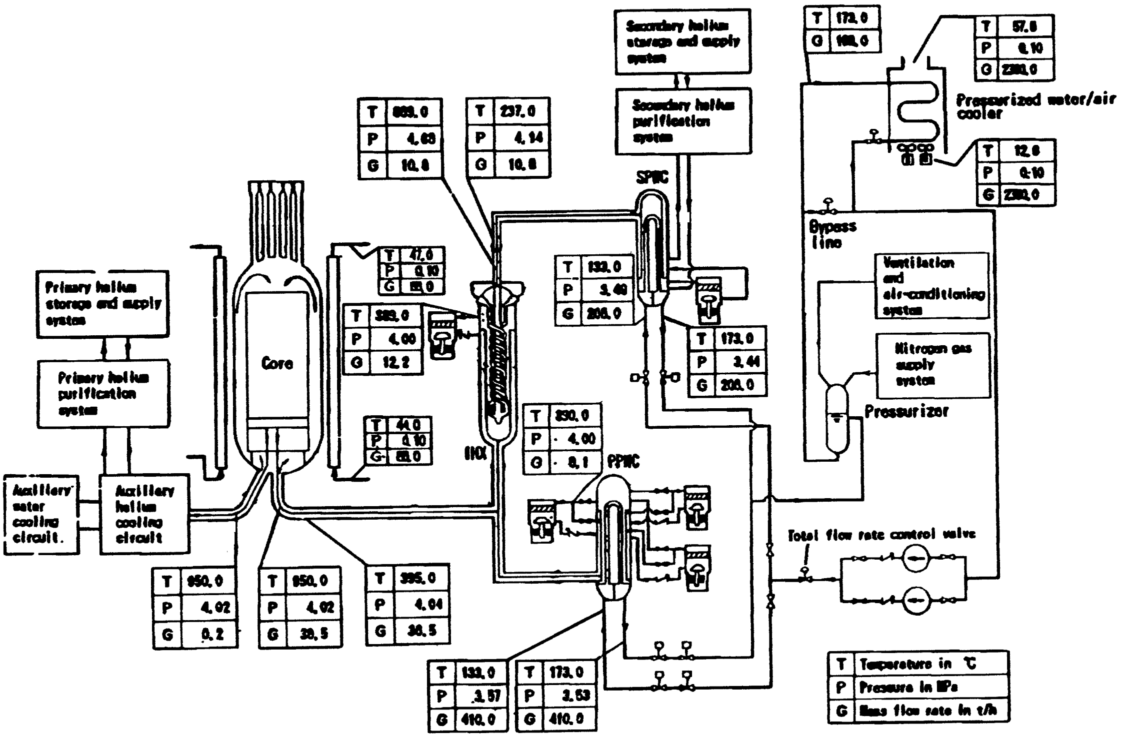


FIG 3.11. Main cooling system flow diagram for the parallel loaded high temperature test operation mode.

Two-thirds of the primary helium enters the PPWC and the remaining one-third goes to the IHX. Each of these helium streams is cooled down to approximately 390°C. In the concentric hot gas duct the cooled helium is slightly heated by hot helium flowing internal to the duct which results in 395°C at the inlet to the RPV. The primary helium pressure is maintained at 4.04 MPa at the inlet of the RPV. Adequate mass flow is maintained by controlling circulator speed.

- Secondary helium cooling system

The secondary helium temperature is strongly dependent upon the operating conditions of the pressurized water cooling system. Under nominal operating conditions, the secondary helium is heated up to ~870°C. The heated helium is then cooled to ~237°C by the SPWC. Also under normal operation, the secondary helium pressure is always controlled to 0.074 MPa higher than the primary helium pressure. The pressure control is achieved by the secondary helium storage and supply system via the secondary helium purification system.

If a heat utilization system is connected to the HTTR, the flow rate of the secondary helium is reduced in order to increase the outlet temperature at the IHX to 905°C.

- Pressurized water cooling system

At rated power operation, the pressurized water temperature increases 40°C through the PPWC and SPWC. After mixing, the water enters the air cooler which is mounted on the roof of the reactor building. The water flow rate through the air cooler is varied with a flow control valve at the inlet of the air cooler in order to maintain a heat-transfer rate of 30 MW as the atmospheric temperature varies. The atmospheric temperature will range from 33°C in a summer season to –13°C in winter. Since the total water flow is required to be 618t/h, a bypass line is provided for the remainder at the air cooler. The water pressure is maintained at 3.5 MPa through control of the amount of nitrogen gas to the pressurizer.

3.3. SAFETY REQUIREMENTS

Operational limits and conditions are specified to ensure the safe operation of the plant at normal operation as well as during anticipated operational occurrences and accidents. A maximum reactor coolant outlet temperature of 950°C has been established so that the HTTR reactor and plant system components such as fuels, graphite blocks and metallic components will not be subjected to significant damage in the event of an accident. As the HTTR is the first HTGR in Japan, strict operational limits and conditions are specified for the plant. Tables 3-5 and 3-6 provide the most essential design criteria for normal and accident operating conditions.

Table 3-5. Design criteria for normal operation

Maximum fuel temperature	1,495° C
Maximum RPV temperature	395° C
Maximum reactor coolant pressure	4.05 MPa
Maximum IHX heat transfer tube temp.	955° C

The maximum fuel temperature of 1495°C for normal operating conditions has been established in order to meet the allowable design fuel temperature of 1600°C at the most severe anticipated operational occurrence of quasi-steady overpower operation.

Table 3-6. Design criteria for accident conditions

Maximum fuel temperature	1, 600° C
Maximum RPV temperature	550° C
Maximum reactor coolant pressure	5.75 MPa
Maximum IHX heat transfer tube temp.	1,000° C

Dimensions of the reactor coolant pressure boundary structures are specified primarily based on design temperature, design pressure and other associated mechanical loads. These design parameter values include allowances of control system error, system configuration effects and measurement inaccuracy. The stipulated service pressure limits are established to meet the design requirements in anticipated operational occurrence and accident as follows.

- During anticipated operational occurrences, the pressure shall be less than 1.1 times the design pressure or the maximum pressure in service.
- During accidents, the pressure shall be less than 1.2 times the design pressure except for the IHX heat transfer tubes and central hot gas dust.

The IHX structures forming the boundary separating the primary and secondary helium shall withstand the creep buckling load sustained in a pipe rupture accident of the secondary helium piping system. This accident is the most severe for these structures.

The maximum allowable rate of reactor coolant temperature change under normal operation depends upon the reactor coolant temperature and the metal temperature of the structural portion in contact with the coolant. The acceptable stress is limited at the higher temperature due to the resultant creep damage on the structures. Based on a parametric analysis of the IHX hot header and reducer structural integrity, the maximum allowable reactor coolant temperature change rate is limited to 15°C/h at a temperature of 650°C or above as the safety consideration.

3.4. HEAT UTILIZATION SYSTEM

The HTTR has three major research and development goals as follows.

- Establishment and upgrading of the technology associated with the high temperature gas cooled reactor
- Establishment of HTGR heat utilization technologies, and
- Innovative and basic technologies.

The HTTR is designed to provide thermal energy of 10 MW at the secondary helium system of the IHX for R&D on heat utilization systems. There are several heat application systems proposed by interested Member States for this R&D programme within the framework of the IAEA CRP on “Design and Evaluation of Heat Utilization Systems for the HTTR”.

The application of steam reforming of methane for the production of hydrogen and methanol has been selected as the first priority system. The thermal energy of the HTTR is 30MWt so that it is potential possibility to scale up to 30MWt for the heat application systems. Detail descriptions of these potential heat utilization systems are discussed in the following chapters.

REFERENCES TO CHAPTER 3

[1] JAPAN ATOMIC ENERGY RESEARCH INSTITUTE, “Present Status of HTGR Research and Development”, (1996).

[2] SAITO, S., “Present status of the HTTR project at JAERI”, High Temperature Applications of Nuclear Energy, IAEA-TECDOC-761, IAEA, Vienna, 11, (1994).

Chapter 4

HEAT UTILIZATION SYSTEMS FOR COUPLING TO THE HTTR

This chapter provides an overview of the individual systems that were evaluated within the scope of the CRP for possible connection to the HTTR. Details of these high temperature process heat applications are provided in Chapters 5 and 6.

4.1 STEAM REFORMING OF METHANE FOR PRODUCTION OF HYDROGEN AND METHANOL

Steam reforming of methane is a mature and economical technology for the production of hydrogen. It is projected to be the major process for the production of hydrogen throughout the next several decades. The basic chemical reactions utilized for this process are as follow:

$$CH_4 + H_2O = 3H_2 + CO - 49\text{kcal/mol}$$
$$CH_4 + 2H_2O = 4H_2 + CO_2 - 39\text{kcal/mol}$$

These reactions are endothermic and consequently there is a need for the consumption of substantial quantities of fossil fuel in order to maintain this chemical process. In this regard, the HTGR can supply the reaction heat instead of fossil fuels. Furthermore, within steam reforming system, CO_2 can be removed before combustion from methane, thereby allowing the HTGR powered steam reforming hydrogen production system to provide an important alternative for the resolution of the global warming issue. The simplified flow chart of the HTTR steam reforming system is shown in Figure 4.1.

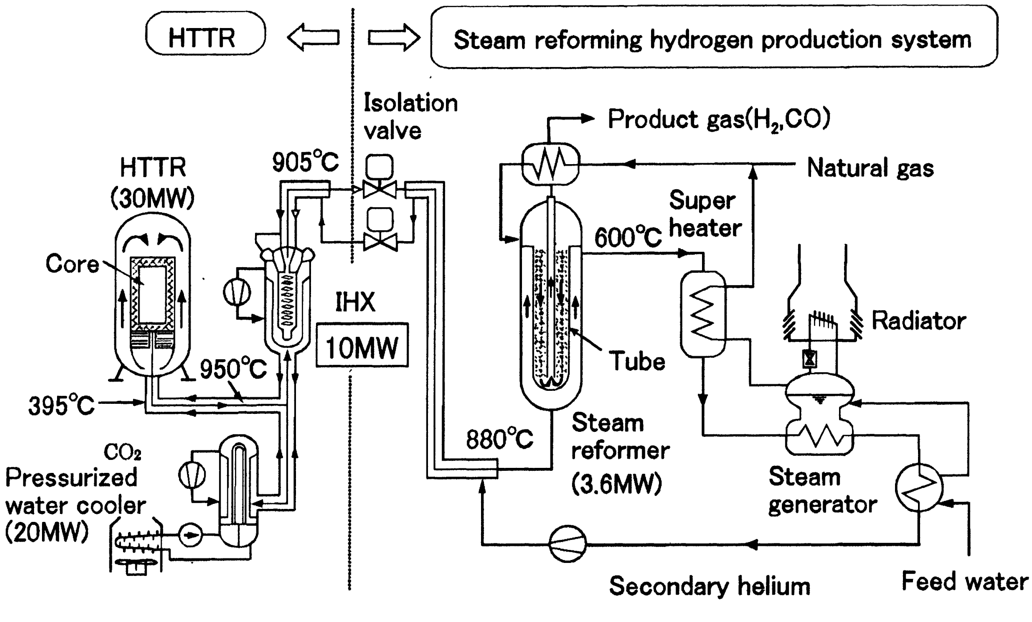


FIG. 4.1. HTTR steam reforming hydrogen production system diagram.

JAERI has been studying application of the HTTR for this process in order to demonstrate industrial capabilities for the use of nuclear heat. The current schedule for testing of the steam reforming hydrogen production process coupled to the HTTR is 2007. Key development achievements include the design of a new concept of steam reformer using a bayonet type catalyst tube, a natural convection type of steam generator integrated with a natural ventilated air cooler and basic safety design criteria for accidents involving fire and explosion originating at the steam reforming plant. Section 5.2 provides details of the following design achievements:

1) Helium heated steam reformer to enable high hydrogen production performance
2) Steam generator for stable controllability and continuous operation at minor accident conditions
3) Safety requirements against tritium transportation, thermal turbulence and fire/ explosion
4) German experience of the PNP project
5) Chinese program for developing a steam reforming system
6) Russian activities

Research and development activities have been on-going at JAERI on the HTTR steam reforming system including the construction of an out-of-pile test facility. Laboratory scale activities including hydrogen permeation tests and corrosion tests have also been performed The preliminary schedule of HTTR steam reforming development in Japan is shown in Figure 4.2.

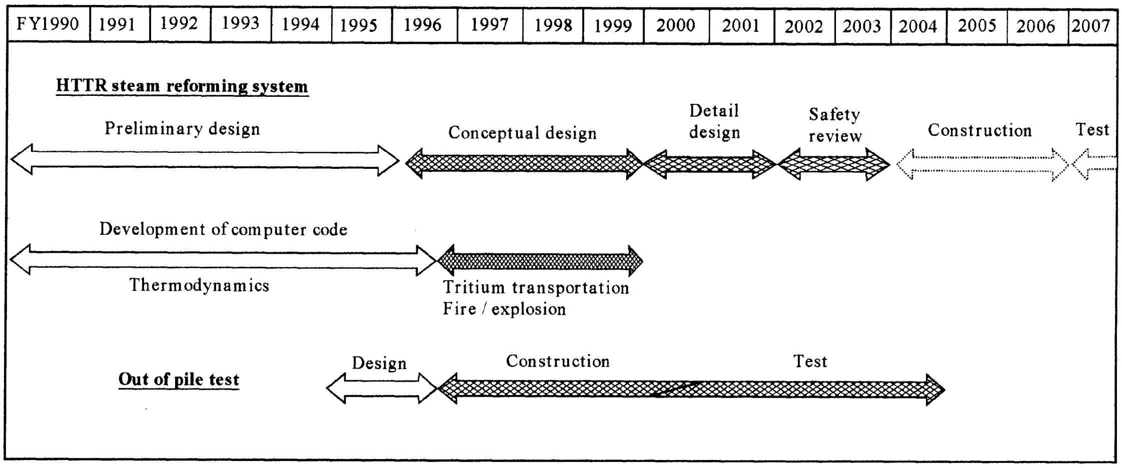


FIG. 4.2. Preliminary schedule of HTTR steam reforming system development.

4.2 CO_2 REFORMING OF METHANE FOR THE PRODUCTION OF HYDROGEN AND METHANOL

4.2.1 Introduction

Carbon dioxide reforming of methane produces syngas with a low hydrogen to carbon monoxide ratio, which can be used directly as fuel for electricity generation and is preferable for many industrial synthesis processes. This reaction also has very important environmental implications, since both methane and carbon dioxide contribute to the greenhouse effect. Converting these gases to a valuable feedstock may significantly reduce the atmospheric emission of CO_2 and CH_4.

Recently, research has focused on the development of catalysts and on the feasible application of this reaction for renewable energy and in industry. Metals pertaining to group VIII, supported on ceramic oxides, are found to be effective for this process. The reforming process requires high temperatures (800–900°C) and high energy input (247 kJ/mole or about 60 kcal/mole). Both can be supplied from the high temperature engineering test reactor (HTTR).

4.2.2 Chemistry

The basic reaction for CO_2 reforming of methane with no addition of steam is:

$$CH_4 + CO_2 \Leftrightarrow 2CO + 2H_2$$

$$\Delta H^0_{298K} = 247\ kJ/mole = 59\ kcal/mole$$

$$\Delta G^0_{298K} = 61770 - 67.3T\ kJ/mole$$

The reverse water gas shift (RWGS) reaction occurs as a side reaction:

$$CO_2 + H_2 \Leftrightarrow H_2O + CO$$

$$\Delta H^0_{298K} = 41\ \ kJ/mole$$

$$\Delta G^0_{298K} = -8545 + 7.84T\ kJ/mole$$

Under the stoichiometric CO_2-reforming, carbon deposition occurs via several undesirable side reactions, as follows:

Decomposition or methane cracking:

$$CH_4 \to C + 2H_2$$

$$\Delta H^0_{298K} = 75\ kJ/mole$$

$$\Delta G^0_{298K} = 21960 - 26.45T\ kJ/mole$$

Boudouard reaction:

$$2CO \Leftrightarrow CO_2 + C$$

$$\Delta H^0_{298K} = -172\ kJ/mole$$

$$\Delta G^0_{298K} = -39810 + 40.87T\ kJ/mole$$

In order to prevent carbon formation, the operating temperatures must be higher than the thermodynamic limit and surplus CO_2 should be used.

CO can be converted to hydrogen by the catalytic shift reaction, known as water gas shift (WGS), as follows:

$$CO + H_2O \Leftrightarrow CO_2 + H_2$$

$$\Delta H^0_{298K} = -41.17\, kJ/mole = -9.84\, kcal/mole$$

The reaction is exothermic, hence, favored by low-operating temperatures. For example, at 220°C and equilibrium, CO is less than 0.2 (% vol.). However, the classical iron-chromium oxides' catalyst (Fe_2O_3/Cr_2O_3) is active only above 320–360°C.

Recently, a new catalyst, based on copper-zinc, was developed. This catalyst enables a reaction temperature at the range of 200–240°C.

This important reaction shows basically that each mole of CO can produce an equivalent mole of hydrogen of inorganic origin (non-fossil hydrogen). It is also a necessary stage in the separation and purification of hydrogen.

4.2.3 Feedstock

Natural sources of the mixture CH_4/CO_2 are available worldwide in large amounts. One example is the giant Natuna offshore gas field in Indonesia. This field is estimated to contain 240 TSCF (trillion standard cubic feet) of gas, with a content of about 70% CO_2. If processed, it could provide 38 million tons of LNG per year over 30 years.

Other important resources are biogas and landfill gas. Biogas is produced via anaerobic digestion of agriculture, industrial and urban wastes, and contains 60–70% CH_4 and 30–40% CO_2. Steam can be added to the feed to adjust the H_2 to CO ratio, according to the final application.

4.2.4 Applications

The applications can be divided into three basic categories:

(1) Direct combustion of the product CO/H_2 mixture (syngas);
(2) Production of fuels, hydrogen, methanol and synthetic gasoline;
(3) Chemicals.

In the case that the syngas is directly combusted in a combined cycle (CC) for electricity generation at high conversion efficiency (over 50% in modern large machines) and the reforming process is carried out with heat coming from a nuclear source, about 30% of methane energy input is added by the nuclear heat, contributing significantly to the reduction of GHG emissions.

Another possibility is to convert the CO/H_2 mixture via the water gas shift reaction to a mixture of CO_2 and H_2. This reaction creates one additional mole of H_2 from inorganic source. The gas is then treated to separate the CO_2 in a concentrated form (compared with the alternative of recovering CO_2 from power station flue gas where the CO_2 is at low partial pressure and is diluted with nitrogen). The recovered CO_2 can be permanently disposed in CO_2 ‘sinks’, which include injection into subterranean cavities or reservoirs, such as aquifers, depleted oil and gas fields, deep un-mineable coal slams, etc.

The product hydrogen can be used directly in a power generating system, such as gas turbines (GT) or fuel cells (FC). This scheme allows a highly efficient utilization of fossil fuels, with dramatic reduction in atmospheric CO_2 emissions, where nuclear heat from the HTGR contributes to about 30% of the feed calorific value.

An additional category of application is the production of fuels and chemicals. The production of methanol is a typical example. Methanol is a clean synthetic fuel and chemical stock. The methanol can be produced according to the reaction:

$$
\begin{aligned}
&2H_2 + CO \rightarrow CH_3OH \\
&\Delta H^0_{600K} = -24\, kcal/mole \\
&\Delta G^0_{600K} = +10.8\, kcal/mole
\end{aligned}
$$

Today, low-pressure (<10 MPa) and low-temperature (220–270°C) commercial methanol synthesis are available, based on a $Cu/ZnO/Al_2O_3$ catalyst. The modern synthesis of methanol is very selective and over 99.5% conversion is obtained.

Since the ratio of H_2/CO is 1:1 for the product of the reaction of CO_2 reforming of CH_4, it has to be adjusted to the ratio of 2:1 for methanol production. In the case where, for instance, feed stock from Natuna gas is used, it is necessary to balance the stoichiometry with water, for example:

$$CH_4 + {}^2\!/_3\, CO_2 + {}^2\!/_3\, H_2O \rightarrow {}^4\!/_3\, CH_3OH$$

Methanol itself can be a precursor to the production of fuel additives like MTBE or synthetic gasoline, as well as other important chemicals, such as formaldehyde, acetic acid, etc.

Besides methanol production, the Fischer-Tropsch synthesis of fuels has been demonstrated on a commercial scale, according to the following reactions:

$$
\begin{aligned}
&nCO_2 + (3n-1)H_2 \rightarrow C_nH_{2n+2} + 2nH_2O \\
&nCO_2 + 3nH_2 \rightarrow C_nH_{2n} + 2nH_2O
\end{aligned}
$$

4.3 COAL CONVERSION

The significant use of natural gas and oil for electric production, transportation, domestic and other industrial purposes has resulted in an increasing deficit of these natural resources. As a result, the world is looking toward coal as a replacement resource to support its ever increasing energy needs.

The world stock of coal is great (> 10^{13} tons of conditional fuel) and significantly exceeds the known resources of oil and gas. However, the use of this solid fuel in its original form will be difficult due to a number of important factors (economical, environmental, regional, etc.).

This problem can be solved to a large extent by gasification or destructive hydrogenation of coal, i.e. its conversion into high quality fuels that are more convenient to use.

Due to the progress made in practical implementation of HTGRs, a number of countries have in recent years been pursuing research into using high-potential thermal power produced by these reactors for gasification of brown coals, hard coals and shale. Owing to employment of inert helium coolant and graphite as structural material in the core, an outlet temperature of 950°C is now attainable in the HTGR. This temperature level suffices for efficient implementation of gasification processes.

The high temperature potential (600° to 950°C) of the HTGR can be used for the steam gasification process or reforming of methane. The low temperature portion of thermal power (300° to 750°C) is used for production of electricity and heating of steam for the gasification process. The efficiency of the gasification process improves with increasing core outlet temperature. As an example, a change of outlet temperature from 950° to 1100°C will provide a corresponding increase of efficiency from 60 to 75%.

Two coal gasification processes are presently being considered for application utilizing the HTGR. These processes are:

- hydrogasification
- steam gasification

In the process of hydrogasification, the reactor thermal power is used for production and heating of the hydrogen that is being supplied for solid fuel gasification. The thermal power from the reactor is conveyed through a tubular steam reformer located in the reactor primary circuit. The process of methane steam reforming produces hydrogen that is further used for coal hydrogasification.

In the process of steam gasification, the heat of the helium coolant is transferred immediately to coal in a gas generator. To ensure the system’s reliability and safety, an intermediate helium circuit is required, which results in a decrease in the coolant temperature potential of approximately 50°C. However, this process can be implemented without the intermediate circuit provided the safety requirements are met.

As compared to conventional techniques, steam and hydrogasification of coal through the use of thermal power produced by high-temperature reactors is a more efficient and environmentally friendly process then utilizing coal. From an equivalent energy standpoint, it would require 1.6 to 1.7 times more coal to produce the same quantity of synthesis gas then that required by using the HTGR. Additional benefits derived from this application of the HTGR include a reduction in fuel transportation costs and a cleaner environment.

4.4 THERMOCHEMICAL WATER SPLITTING FOR HYDROGEN PRODUCTION

Hydrogen can be stored in a variety of ways. Hydrogen can be transported for a long distance with lower transportation loss compared to electricity. Hydrogen is an ideal fuel and also an important industrial feedstock. Furthermore, if necessary, the chemical energy of hydrogen can be transformed into electricity using fuel cells, etc. All of these features make hydrogen an attractive candidate to be the future secondary energy, i.e. an energy carrier.

The attractive characteristics of hydrogen are strongly emphasized when it is produced from water using non-fossil primary energies. Since the combustion product of hydrogen is water, hydrogen production from water using non-fossil fuels completes a very clean energy system, the “hydrogen energy system,” that will play an important role to mediate the environmental issues such as global warming caused by the greenhouse effect of CO_2. In the transition to the hydrogen energy system, hydrogen from water can contribute to the clean and efficient utilization of fossil fuels. For example, it should be very effective to upgrade the highly abundant CO_2-rich natural gas from Natuna, Indonesia.

The thermochemical water-splitting process offers a novel means for large-scale hydrogen production from water. The principle of this process is schematically illustrated in the Gibbs free energy change vs. temperature diagram shown in Figure 4.3. Thermal decomposition of water does not proceed practically below a temperature of several thousand Kelvin. However, it can be carried out with lower temperature heat by combining high temperature endothermic and low temperature exothermic reactions. The reactions should, of course, be selected so that the net chemical change is water splitting. The process works like a chemical engine to produce hydrogen while absorbing high temperature heat and discharging low temperature waste heat.

Numerous chemical reactions and a variety of combinations have been surveyed utilizing the HTGR as the primary heat source for thermochemical water splitting. Among them, the Iodine-Sulfur (IS) process, proposed by General Atomic Co. in early 70’s, is one of the most promising processes due to the comparatively low number of reactions, the potential of high thermal efficiency, etc. The process is composed of the following chemical reactions:

$$I_2 + SO_2 + 2H_2O = 2HI + H_2SO_4 \quad (1)$$
$$2HI = H_2 + I_2 \quad (2)$$
$$H_2SO_4 = H_2O + SO_2 + 0.5O_2 \quad (3)$$

Reaction (1), the so-called “Bunsen reaction”, is an exothermic SO_2 gas absorption reaction to be carried out in the temperature range of 20° to 100°C. Thermal decomposition of hydrogen iodide (2) is slightly endothermic and can be carried out in the gas phase or in the liquid phase. Thermal decomposition of sulfuric acid (3) is strongly endothermic and proceeds in two stages: the decomposition of H_2SO_4 into H_2O and SO_3 at 300° to 500°C, and the decomposition of SO_3 into SO_2 and O_2 at 800° to 900°C.

The IS process has been studied in the USA, Germany, Canada and Japan. These studies have included the chemistry of the reactions and related product separation, the process flow, and the materials of construction.

Recent progress at JAERI has been in the following fields:

- Laboratory-scale demonstration of continuous hydrogen production,
- Modification of the HI processing scheme, and
- Evaluation of materials of construction for boiling sulfuric acid environment.

The results of JAERI’s investigation into the IS process and the associated R&D program are summarized in Section 6.1.

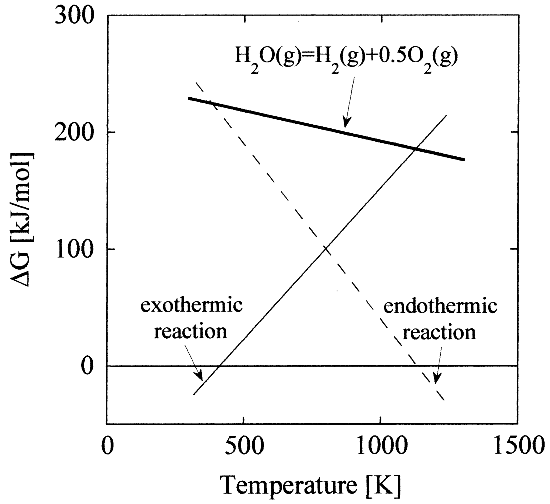
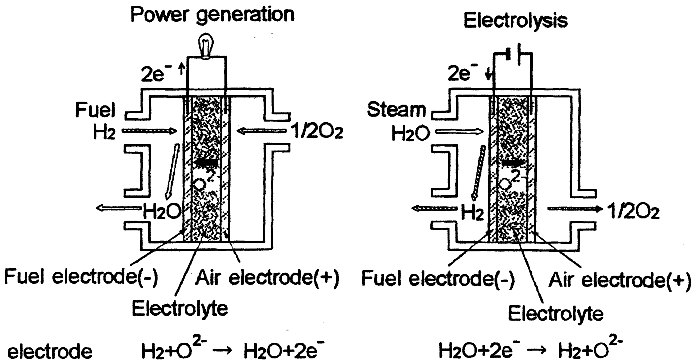


FIG. 4.3. Concept of thermochemical water-splitting for hydrogen production.

4.5 HIGH TEMPERATURE ELECTROLYSIS OF STEAM FOR HYDROGEN PRODUCTION

The high-temperature electrolysis of steam (HTES) using ceramic electrolysis cells is one of the advanced technologies of the hydrogen production process. The HTES is a reverse reaction of the Solid-Oxide Fuel Cell (SOFC) which is presently being vigorously developed around the world. The latest technology of the SOFC (i.e. the electrolysis cell) can be applied to the HTES. From the viewpoint of the energy demand, the HTES could potentially suppress the electric energy required to decompose steam much lower than that of the water electrolysis. Figure 4.4 shows the energy demand for the water and steam electrolysis. The total energy demand (ΔH) is the sum of the Gibbs energy (ΔG) and the heat energy (TΔS). The electric energy demand, ΔG, decreases with increasing temperature as shown in Figure 4.4; the ratio of ΔG to ΔH is about 93% at 100°C and about 70% at 1000°C.

The HTES, however, is at a very early stage of technology development, and thus, it is necessary to take extensive efforts in order to make the HTES feasible. As a first step, JAERI has been carrying out laboratory-scale experiments to examine the effectiveness of the HTES for hydrogen production and to improve the HTES technology. Section 6.2 of this report presents typical experimental results obtained using practical electrolysis cells.



	Power generation	Electrolysis
at fuel electrode	$H_2+O^{2-} \rightarrow H_2O+2e^-$	$H_2O+2e^- \rightarrow H_2+O^{2-}$
at air electrode	$1/2O_2+2e^- \rightarrow O^{2-}$	$O^{2-} \rightarrow 2e^-+1/2O_2$
Total reaction	$H_2+1/2O_2 \rightarrow H_2O$	$H_2O \rightarrow H_2+1/2O_2$

FIG. 4.4. Principle of high temperature electrolysis of steam.

4.6. GAS TURBINE FOR ELECTRIC PRODUCTION

A promising approach for making good use of the high temperature capability of the HTGR is to use the primary helium coolant to drive a gas turbine in a direct closed cycle arrangement. In the 1970s, this was extensively studied in the U.S., Germany, the U.K. and France. At that time, the concept was based on enclosing a large (2000 to 3000 MW(th)) reactor core and the gas turbine power conversion system within a prestressed concrete reactor vessel. After nearly a decade of work, this concept was abandoned primarily because the system achieved only about 39% efficiency and would have required substantial development to resolve design and safety issues [1]. Subsequent technological advancements in the design and operation of magnetic bearings, compact plate-fin heat exchangers and turbomachines, coupled with international capability for their fabrication and testing, and the development of the annular core modular HTGR has resulted in renewed interest in this HTGR concept.

Development of the gas turbine, modular HTGR design was an evolutionary process initially coupling a gas turbine directly to a large HTGR. Subsequent reactor plant modifications to take advantage of the increased safety attributes and the economics of nuclear plant simplification brought about the modular HTGR. This modular reactor plant was originally coupled to an IHX for the production of electricity and other industrial uses via the generation of steam.

However, in order to be economically competitive, the thermal efficiency of nuclear power had to be markedly improved to compete with modern, high efficiency fossil plants. HTGR technology has always held the promise for electricity generation at high thermal efficiency by means of a direct Brayton cycle and fortuitously, technological developments during the past decade provided the key elements to realize this promise. These key elements are as follows:

- The HTGR reactor size had been reduced in developing the passively safe module design. At the same time, the size of industrial gas turbines had increased. The technology was now available for a single turbo-machine to accommodate the heat energy from a single HTGR module.

- Highly effective compact recuperators had been developed. Recuperator size and capital equipment cost are key economic considerations. Highly effective plate-fin recuperators are much smaller than equivalent tube and shell heat exchangers, provide for substantially less complexity and capital cost, and are a key requirement for achieving high plant efficiency.

- The technology for large magnetic bearings had been developed. The use of oil lubricated bearings for the turbo-machine with the reactor coolant directly driving the turbine was problematic with regard to the potential coolant contamination by the oil. The availability of magnetic bearings eliminates this potential problem [2].

A major requirement was for the plant to become substantially simplified in order to provide a significant reduction in the capital expenditure for new capacity additions. This simplification and the benefits derived from the substantial increase in cycle efficiency have formed the basis for development of the gas turbine modular HTGR plant. Figures 4.5 and 4.6 provide a graphic representation of the simplification that can be achieved in going to the basic direct gas turbine cycle and a comparison of nuclear power plant efficiencies, respectively.

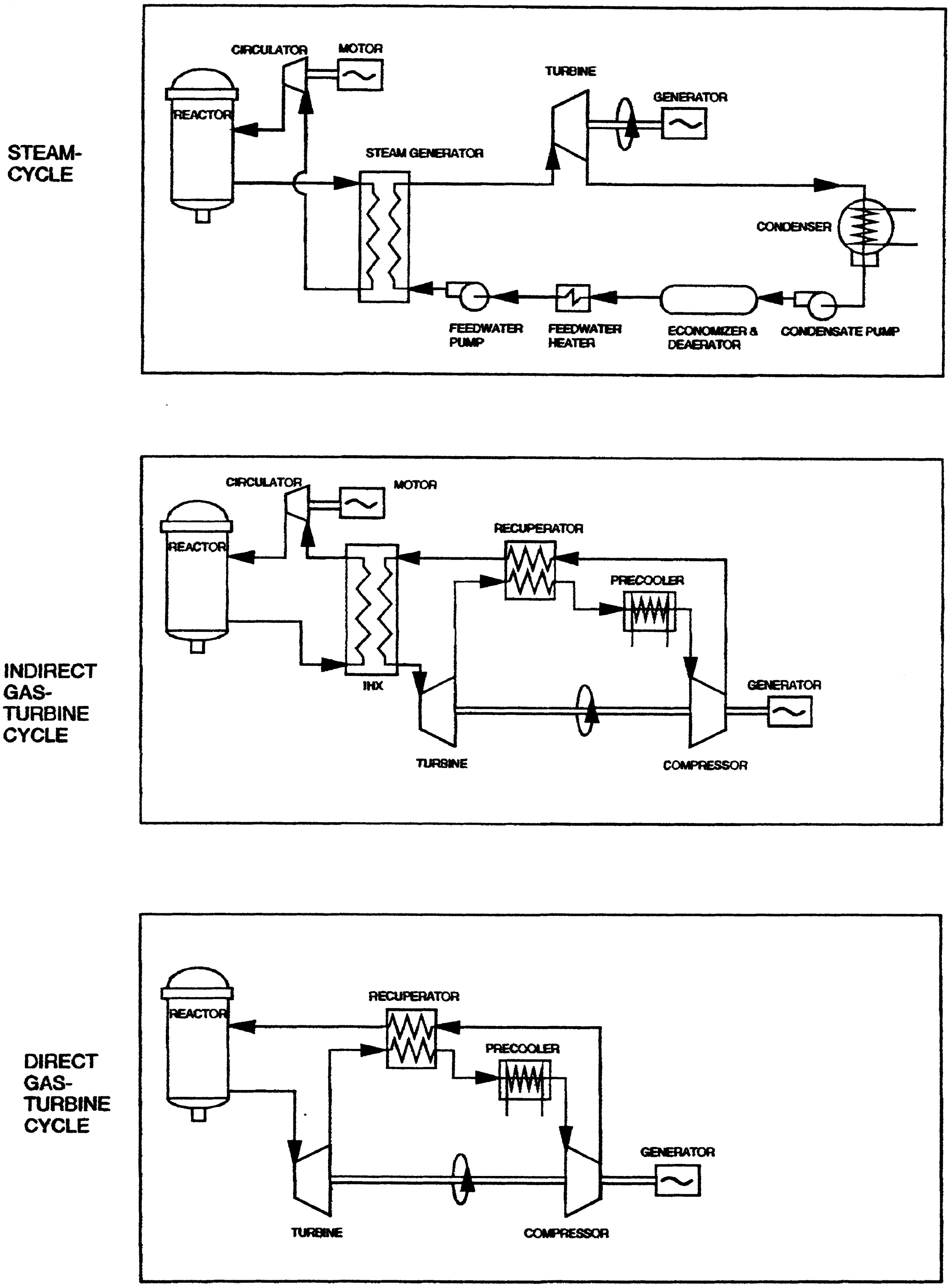


FIG. 4.5. Plant implification, steam cycle to the closed cycle gas turbine plant [3].

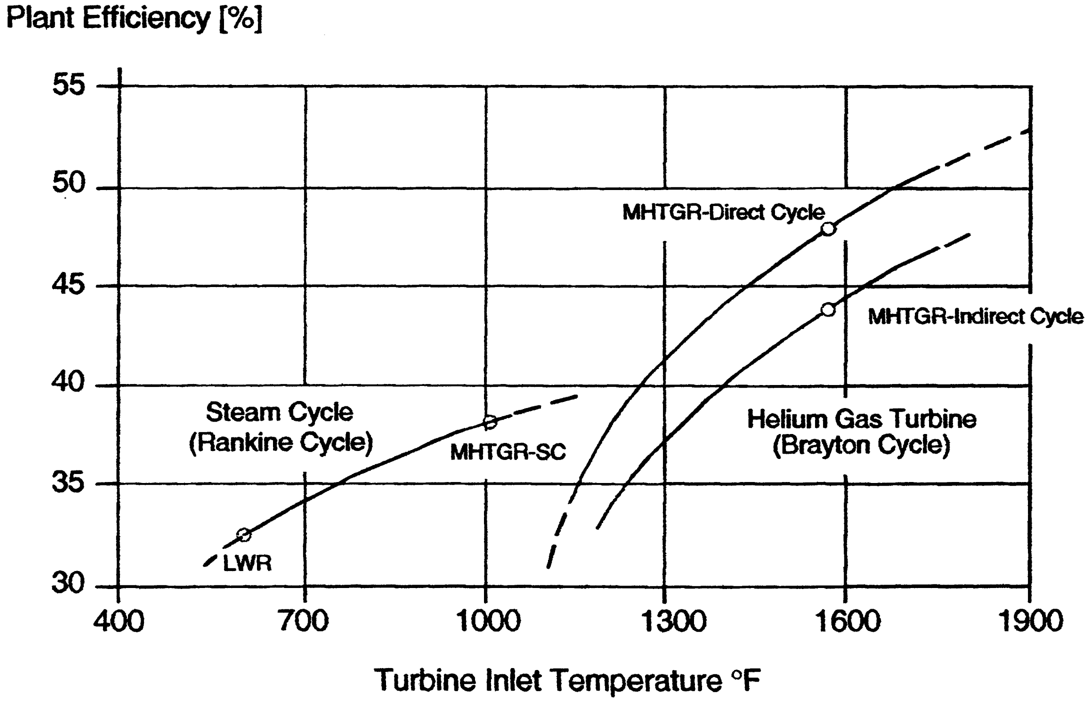


FIG. 4.6. Plant efficiency comparison [3].

In 1993, the electric utility of South Africa, ESKOM, and the U.S. HTGR designer, General Atomics, independently initiated evaluations of the closed cycle gas turbine HTGR concept. Subsequent positive determination of its potential and feasibility resulted in two different, but often similar, designs; the Pebble Bed Modular Reactor (PBMR) and Gas Turbine-Modular Helium Reactor (GT-MHR), respectively. Although the PBMR and GT-MHR programs represent the most significant application to date of HTGR development resources, the international interest in the gas turbine modular HTGR system has also lead to the scientific investigation of other concepts [4]. Section 5.4, herein, provides a general overview of the designs being investigated by the Member States participating in this CRP. These include the GT-MHR, PBMR, the Chinese indirect cycle, and two designs for power levels between 300 and 600MW(th) by Japan.

4.7 STEAM GENERATION FOR OIL RECOVERY

Within the scope of this CRP, Indonesia and Russia presented information associated with national needs for carbonaceous conversion and oil recovery by using HTGRs. Also, General Atomics performed a study which applied a 2 × 600 MW(th) PS/C MHR plant to recovering heavy oil.

As there are currently no new experimental activities underway associated with oil recovery, the feasibility of an associated demonstration test of this industrial application with the HTTR was determined to not be appropriate at the present time. It was agreed that, should this situation change, an out-of-pile demonstration would be initiated before coupling to the HTTR. Subsequently, investigation of enhanced oil recovery systems was deemed as a "Future Candidate" and no further actions were taken within the scope of this CRP.

As the long-term potential for utilizing an HTGR as the energy source for enhanced oil recovery is significant, it was determined that a brief summary of a potential nuclear powered heavy oil recovery plant be presented within this document. This is summarized by applying a 2 × 600 MW(th) PS/C-MHR [5] to recovering heavy oil.

4.7.1 Heavy oil recovery [6]

About 15% of the U.S. domestic oil reserves are in the form of heavy crude oil, defined as having an American Petroleum Institute (API) gravity of < 20°. Recovering this heavy oil can be greatly improved by stimulation methods, such as steam injection. The thermal energy requirements for recovering heavy oil with steam depend on the oil field size and the reservoir characteristics. This study based the field size on the PS/C-MHR providing steam for well injection, dewatering, and other process facilities and co-generating electric power for on-site and off-site uses.

Figure 4.7 shows a typical field arrangement for a heavy oil recovery project using steam from a Modular Helium Reactor. If injection wells are spaced 2.5 acres apart (average), ~ 698 m^2 of heavy oil field may be operated at a time with the 2 × 600 MW(th) plant. Typically, the well injection head injects steam at ~3.4 MPa, which is sufficient to reach depths down to 366m. However, in some locations, the reservoir characteristics and overburden thickness require injection pressures up to 4.5 MPa. Presently, heavy oil (steam drive) operators use steam at ~ 80% quality (dry) to hold dissolved solids in solution. Studies have shown that the oil yield increases significantly with the steam quality. With a PS/C-MHR, which can deliver steam in excess of 538°C, dry saturated steam can be injected into the well if desired.

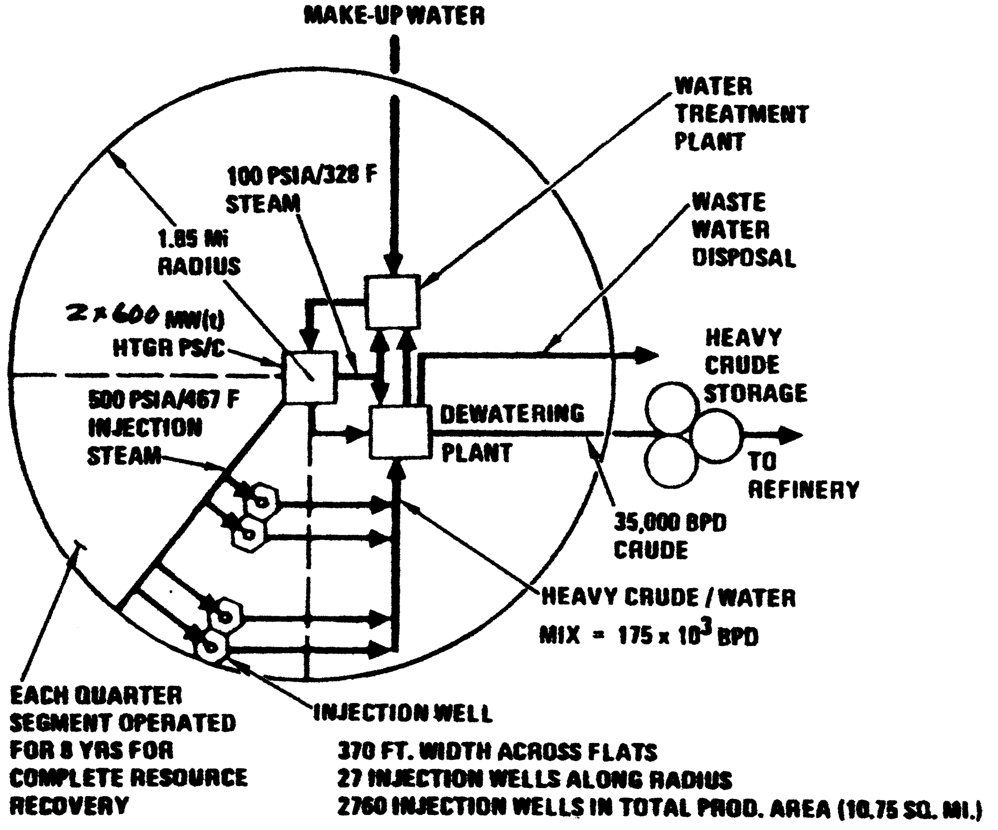


FIG. 4.7. Field arrangement for 5,562 m^3/stream day heavy oil application.

As discussed above, the steam conditions desired at the injection wells are ~3.45 MPa with 85% or higher quality, and very little electrical power is required for the oil field operations. Figure 4.8 shows a typical heat cycle for heavy oil recovery. First, 538°C/16.65 MPa steam from the PS/C-MHR steam generators is expanded through a turbine generator to an intermediate distribution pressure for the oil field injection wells. Part of the exhaust steam from this turbine generator is then expanded through an extraction turbine generator to provide steam for feedwater heating and to produce additional co-generated electric power. For this study, the heat cycle was designed to produce the maximum process steam output consistent with efficient co-generation of electrical power and feedwater heating requirements. Less process steam and more electrical power can be achieved by adding condensing turbine generator capacity; this would be desirable for specific oil field applications with attractive nearby electric power markets.

The heat cycle conditions the main turbine generator exhaust steam by desuperheating before distribution to the injection wells. The amount of desuperheating can be adjusted to suit specific oil fields or different periods during the oil field production life.

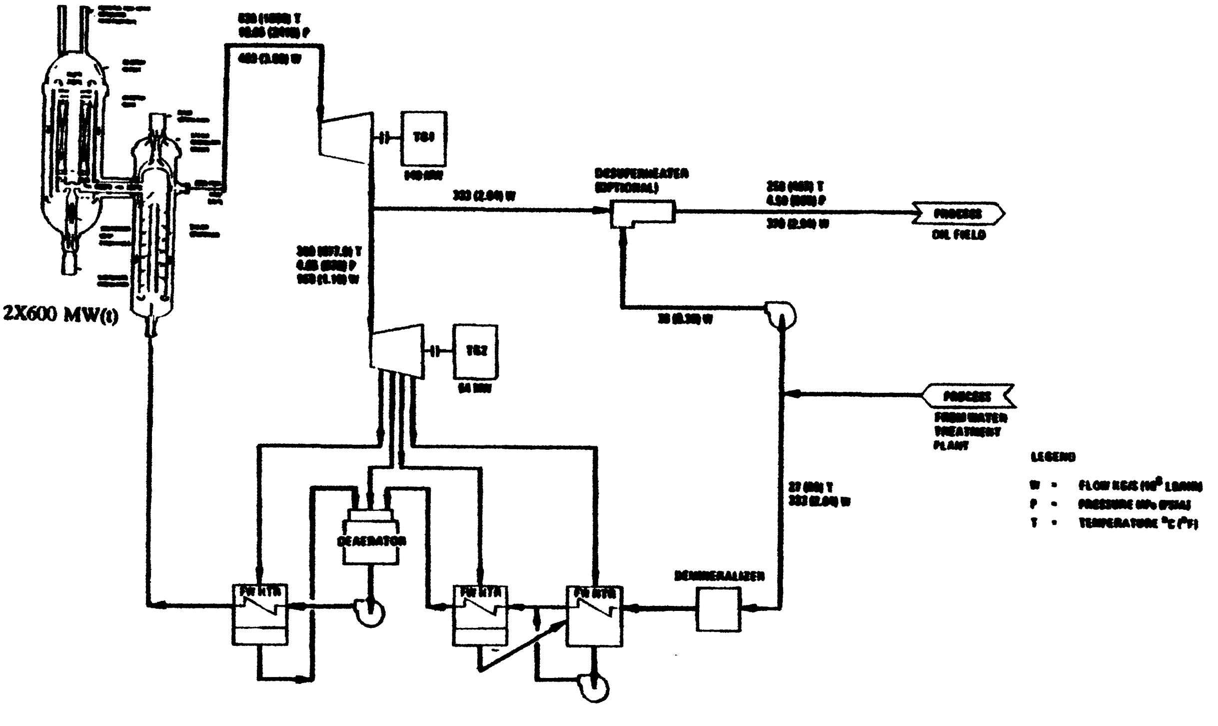


FIG. 4.8. Cycle diagram for 2 × 600 MW(th) PS/C-MHR plant for heavy oil recovery application.

4.7.2 Tar sands oil recovery [6]

Tar sands represent a major energy resource that increases in importance as world supplies of crude oil become limited. The oil potential from tar sands in Canada is estimated to equal the world's known reserves of conventional oil; the potential of U.S. tar sends is smaller, but still substantial [4.8 to 5.6 × 10^9 m^3]. Current Canadian production is limited to deposits suitable for strip mining; however, the major reserves lie at greater depths. To exploit these deep reserves, large-scale pilot projects have investigated in-situ recovery. These projects inject saturated steam into the tar sands deposits.

This section summarizes a study [7] to apply the 2 × 600 MW(th) PS/C-MHR to tar sands oil recovery and upgrading. The raw product recovered from the sands is a heavy, sour bitumen; upgrading, which involves coking and hydrodesufurization, produces a synthetic crude (refillable by current technology) and petroleum coke. Steam and electric power are required for the recovery and upgrading process.

The tar sands fields are generally located in sparsely populated areas of Canada. Therefore, the PS/C-MHR plant can be located at the center of the recovery area, minimizing the required piping and the associated pressure drops and heat losses. When the recovery is complete in one quarter of the operating field, the piping will be shifted to the next quarter until the entire field has been covered. Since it takes ~ 7 years to complete each quarter of the field, the PS/C-MHR will have operated most of its design life (30 years) by the time the recovery is complete.

The nominal steam conditions desired at the injection well are ~ 13.8 MPa and 336°C. Since this steam is obtained by throttling the main steam from 16.65 MPa, adjusting the pressure to account for variations in the distribution pressure drop has some flexibility. A desuperheater using returned water reduces the steam temperature to the saturated condition. The steam required for upgrading, water treatment, and auxiliaries can be further conditioned as required. The balance of the steam, not used by the process, is diverted to a turbine generator, which co-generates electric power and provides a conventional feedwater heating system for the entire condensate flow. The recovery plant processes makeup and clean condensate. To ensure the specified purity for the PS/C-MHR steam generators, the feedwater train includes a full-flow polishing demineralizer.

Figure 4.9 shows the cycle for the 7309 m ^{3}/day plant. In this case, only enough steam for feedwater heating of 147 kg/s is diverted to the turbine generator; the recovery plant uses the balance 439 kg/s. The turbine generator is a noncondensing unit similar to the high pressure and intermediate pressure units of a small conventional turbine generator; its gross output is 101 MW(e), while its net output is 64 MW(e). The difference is used to drive the PS/C-MHR circulators, the feed pumps, the condensate pumps, and other nonprocess auxiliaries.

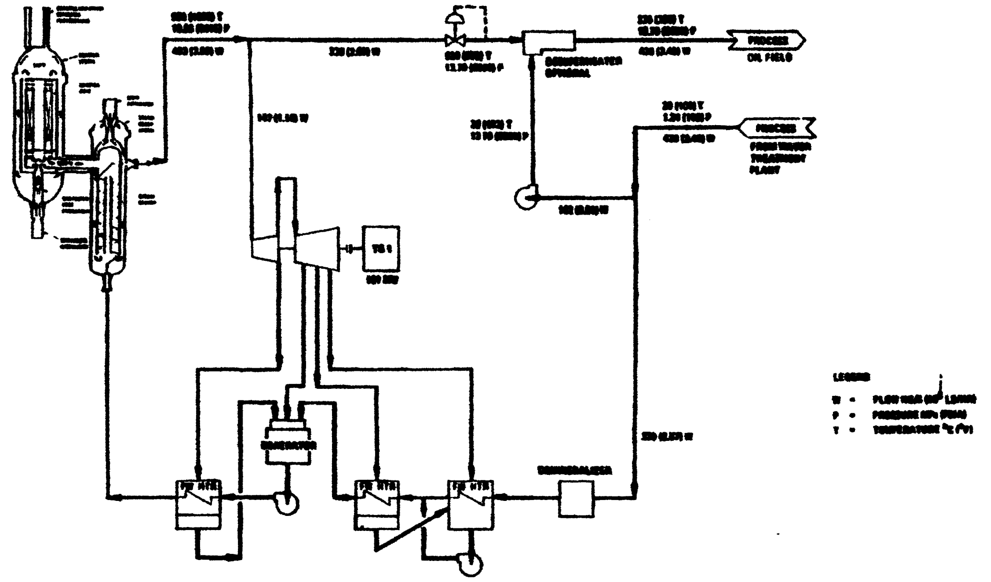


FIG. 4.9. Cycle diagram for 2x600 MW(t) PS/C-MHR for tar sands oil recovery.

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Chapter 5

CANDIDATE HEAT UTILIZATION SYSTEMS TO BE CONNECTED TO THE HTTR

This chapter provides details associated with each heat application system designated by the Chief Scientific Investigators for utilization in conjunction with the HTTR. This includes a description of each system, the status of its technology, purpose and benefits to be derived by its application to the HTGR and proposed testing with the HTTR.

5.1 CONSIDERATIONS DETERMINING PRIORITIES OF THE HTTR SYSTEM

The following seven candidate heat application systems were investigated within this CRP by the participating Member States:

- Steam reforming of methane for production of hydrogen and methanol
- $C0_2$ reforming of methane for production of hydrogen and methanol
- Coal conversion
- Thermochemical water splitting for hydrogen production
- High temperature electrolysis of steam for hydrogen production
- Gas turbine for electricity production
- Steam generation for oil recovery

The objectives to be achieved for coupling a heat application system to the HTTR is to develop related technologies and to demonstrate the technical feasibility of utilizing nuclear heat as the energy source for the system to function. Taking into consideration the importance of these objectives, the following criteria was established in order to select the first priority candidate system for the HTTR:

1) The first priority candidate system should be a technically mature heat application system to allow connection to the HTTR in 2006 according to the draft schedule of HTTR plant operation,
2) The system should have potential market needs,
3) The technologies developed as a result of this programme should provide application benefits to other systems.

With regard to the steam reforming system for the production of hydrogen, considerable investigation has taken place including out-of-pile test in Germany and Japan. Many R&D results currently exist and key technical issues have been proven for this heat application process. Furthermore, the potential needs for hydrogen and methanol as future energy sources have become larger from the viewpoint of global warming. Also, the flexibility exists for demonstration of the $C0_2$ reforming system by using the steam reforming system with only a change in the feed gas composition.

Currently, coal is the most promising resource among all fossil fuels. Needs for the conversion of coal is significant in coal producing countries in order to provide for more efficient energy transportation. The coal conversion system requires steam or hydrogen so that there are only minimal R&D needs specific for this system.

Thermochemical water splitting is one of most attractive HTGR heat application systems. JAERI continues a basic study of the iodine/sulfur process (IS process) in a lab-scale test apparatus. This process requires extensive development and many R&D areas are yet to be proven before connection to the HTTR.

The basic study for high temperature electrolysis of steam was terminated within the CRP-4 activities because of its early stage of technology development, particularly in the area of reliable electrode development. This is further discussed in Section 6.2.

The gas turbine coupled to a modular HTGR for electricity production has a high potential for commercialization due to the simplicity of the system, its safety characteristics and high thermal efficiency with corresponding low capital and operational costs. Both Eskom, the electric utility of South Africa and a consotrium of organizations from Russia, Japan, France and the US are planning commercial HTGR gas turbine plants. Low helium gas pressure and a thermal energy of 10 MW in the secondary helium loop of the HTTR precludes achieving high system efficiency for electricity generation and, therefore, only technical feasibility of the process can demonstrate using the HTTR.

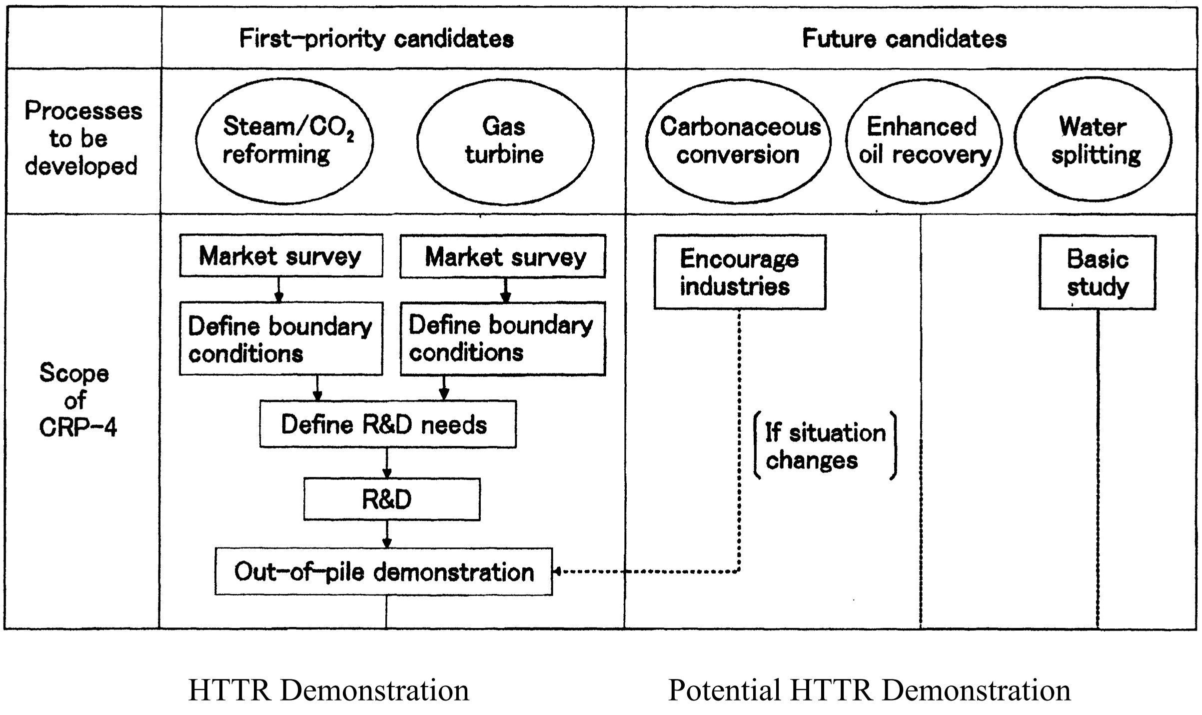


FIG. 5.1. Scope of the CRP in relation to demonstration with the HTTR.

The above considerations were taken into account in deciding on the first candidate heat application system to be connected to the HTTR. These first candidate systems were chosen to be the steam/CO_2 reforming system and gas turbine system as shown in Figure 5.1. The first process heat application test will be the steam/CO_2 reforming system. The demonstration test of the gas turbine system will be performed following the demonstration testing associated with the steam/CO_2 reforming system.

Other candidate systems are capable for demonstration at JAERI in out-of-pile test loops prior to coupling with the HTTR.

5.2 STEAM REFORMING OF METHANE FOR PRODUCTION OF HYDROGEN AND METHANOL

Consumption of large amounts of fossil fuels has resulted in an enhanced global warming problem. In order to diminish the global warming issue and to sustain future societal development, new energy resource/carrier and/or energy technology needs to be developed which meet the following requirements:

(1) Freedom from resource constraint, especially stable and unlimited supply of energy resource,
(2) Environmentally friendly energy resources,
(3) High efficiency energy usage, and
(4) Concentrated energy use for industries.

Such an energy technology includes the reforming of fossil fuels to new energy carriers. Hydrogen and methanol are the key energy carriers to meet these requirements. Several countries such as China, Germany, Japan, Russia and the US have carried out specific design studies associated with these carriers. Furthermore, Germany and Japan had carried out the out-of-pile tests and components tests. [1, 4] The activities of Japan, Germany, China and Russia are described in the following sub-sections.

5.2.1 The Japanese process design (system arrangement and heat balance)

The New Sunshine Program (R&D Program on Energy and Environmental Technologies) was started in Japan in April 1993, with focus on development of hydrogen production and utilization technology supported by Ministry of International Trade and Industry [5]. Within this programme, a HTGR supplies high temperature nuclear heat enabling the attainment of highly efficiency of heat utilization without any emission of CO_2 gas. It was determined that hydrogen and methanol production by means of nuclear heat generated from an HTGR has a high possibility to help in the resolution of the global warming issue.

JAERI has been studying the possibility of developing nuclear-heated hydrogen production technologies and of demonstrating technical feasibility of steam reforming for production of hydrogen and methanol with the HTTR since 1990. A steam/methane reforming system has the advantages as described below:

1) It is highly possible to couple the steam reforming system to the HTTR in the early 2000's because a fossil-fired steam reforming of natural gas (methane) or naphtha is an economical and mature technology for production of hydrogen in the chemical industries.
2) Technical solutions demonstrated by coupling the steam reforming system to the HTTR will contribute to other nuclear-heated hydrogen production systems.
3) Nuclear-heated steam reforming is a practicable means for producing hydrogen because of pre-combustion removal of CO_2 from hydrocarbon resources, such as natural gas and coal, and has significant potential as a method to facilitate the transition from fossil fuels to future hydrogen energy systems [6].
4) Some of key technologies of this system, such as a helium heated steam reformer, have been developed in the framework of ERANS project in 1972-1978 [2].

JAERI developed the framework of the HTTR steam reforming system during preliminary design from 1990 through 1995. The conceptual design of this system has been carried out subsequently. The steam reforming system is to be connected to the HTTR through the IHX as shown in Figure 5.2. This will provide 10MW of thermal energy from the HTTR to the steam reforming system.

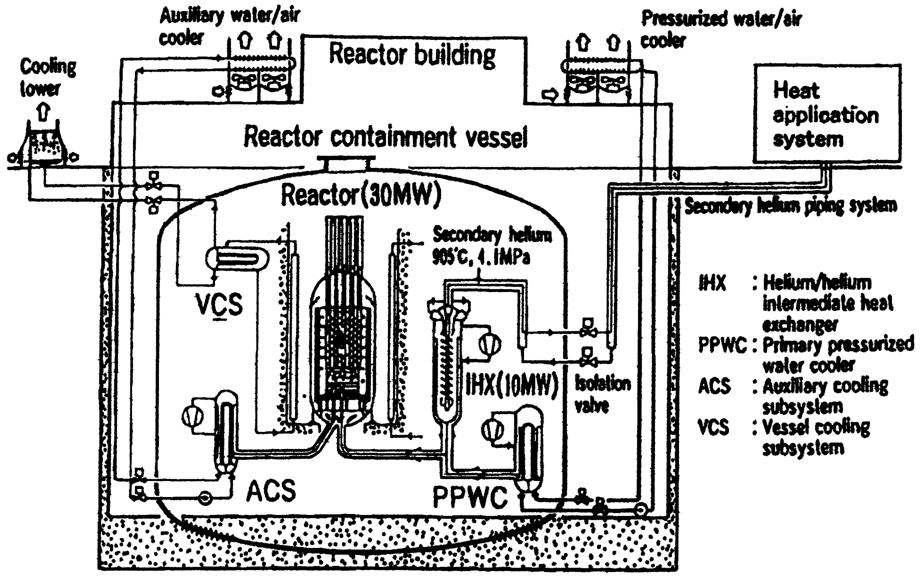


FIG. 5.2. Simplified diagram of the HTTR heat application system.

The process parameters of the HTTR steam reforming system such as temperature, pressure, and flow rate, are shown in Figure 5.3 and Table 5-1. Though the temperature of secondary helium at outlet of the IHX is ~905°C, it becomes ~880°C at the inlet of steam reformer (SR) due to the heat loss from hot gas duct between the IHX and the SR. The following steam reforming reaction and CO sift reaction occur within the SR.

$CH_4 + H_2O = 3H_2 + CO$ -206kJl/mol
$CnHm + nH_2O = (n+m/2)H_2 + nCO$ (endothermic)
$CO + H_2O = H_2 + CO_2$ +41kJ/mol

In conventional steam reforming system, feed steam is supplied excessively for effective conversion from feed methane to hydrogen and for preventing carbon deposition in the catalyst zone. In the HTTR steam reforming system, a steam/carbon ratio of 3.5 has been selected. The required steam is about 5,160kg/h at rated conditions so that the thermal energy necessary to generate steam is 3.1MW. The thermal energy of the product gas at outlet of the SR is only 1.9MW. 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 SR to generate feed steam for the SR. 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.

Table 5-1. Major process parameters

Secondary helium	
Flow rate	**9070kg/h**
Temperature	
Outlet/Inlet of the IHX	**905°C/160°C**
Outlet/Inlet of the SR	**880°C /600°C**
Outlet/Inlet of the SG	**555°C /275°C**
Pressure	**4.1MPa**
Process feed gas	
Flow rate	**6450kg/h**
Composition	
CH_4	**17.7 mol%**
C_2H_6	**1.2 mol%**
C_3H_8	**0.6 mol%**
C_4H_{10}	**0.3 mol%**
H_2O	**80.2 mol%**
Pressure	**4.5MPa**
Product gas	
Composition	
H_2	**38.7 mol%**
CO	**4.5 mol%**
CO_2	**5.7 mol%**
CH_4	**5.1 mol%**
H_2O	**46.0 mol%**
Pressure	**4.1MPa**

Compositions of feed gas and product gas in the HTTR steam reforming system are shown in Table 5-1. The flow rate of natural gas as a feed gas is 1,290kg/h and the flow rate of steam is 5,160kg/h at the inlet of the SR. The temperature of the product gas is ~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 4.5MPa and ~830°C so that the expected conversion ratio from methane to hydrogen is ~68%. As a result, 32% of methane will remain in the product gas. In conceptual design, this residual methane is burned in the flare stack with other gases.

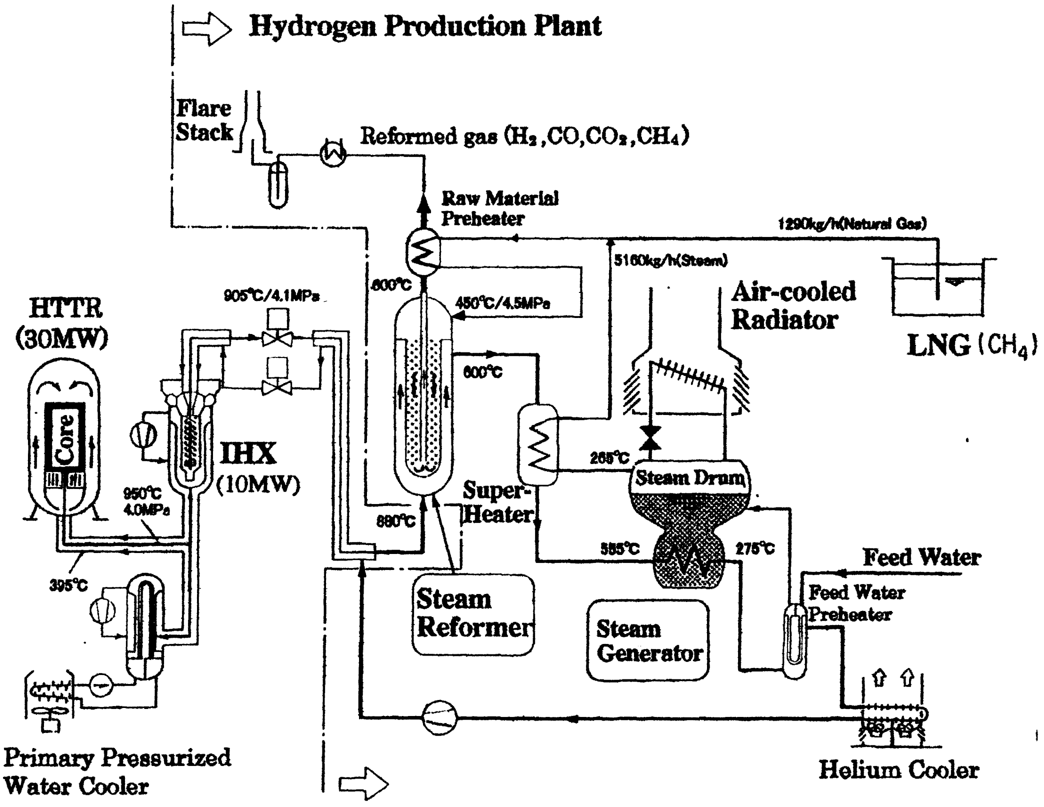


FIG. 5.3. Flow scheme of the HTTR steam reforming system.

The HTTR steam reforming system can provide about 4,200m^3-STP/h of hydrogen production using a Ni-based catalyst with 10MW 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 ~80%.

5.2.2 Engineering design (key components and operation procedure)

The HTTR steam reforming system will be the first nuclear process heat application system in the world. The requirements of this system are for safe operation and high hydrogen production efficiency. To meet these requirements, JAERI has carried out design work and achieved the following improvements.

(1) 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.
(2) 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.
(3) An air-cooled radiator is connected to the steam generator to operate as a final heat sink during normal and anticipated operational occurrence condition.

(1) Development of new concept steam reformer

The high hydrogen production rate derives from the high process feed gas rate and high conversion rate. 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 the temperature and the pressure of process gas.

The secondary helium gas is pressurized up to 4.2MPa in order to prevent the accidental release of fission products from the core to the environment and to assure the structural integrity of the IHX heat exchanger tubes against creep damage. The HTTR can provide high temperature helium gas of 905°C at the outlet of the IHX and of 880°C at the inlet of the SR. Furthermore, in order to generate feed steam by the thermal energy of secondary helium gas, the helium gas temperature at the outlet of the SR is required to be ~ 600°C so that only thermal energy of 3.7MW is supplied to the SR from helium gas. This high-pressure and low-temperature condition is a disadvantage for steam reforming reaction.

A new concept heat exchanger type of steam reformer is required for enhancing hydrogen production rate. In evaluation this, JAERI considered the following:

(1) Increasing heat input into the process gas
(2) Increasing the reaction temperature of the process gas at the outlet of the catalyst zone
(3) Optimizing reforming gas composition to enhance the reforming rate

JAERI adopted a bayonet type of catalyst tube which can use both the outside and inside of the flow gas for heating of the process gas. The thermal energy input into process gas increase from 3.7MW to 5.0MW.

If catalyst tube is infinitely long, the process gas temperature becomes close to the helium gas temperature. But in general, a catalyst tube length limit of ~10m is mandated from the viewpoint of seismic design. It is necessary to enhance the heat transfer rate in order to design for an adequate SR size. There are several means for enhancement of the heat transfer such as baffle, double tube, fins, etc., and JAERI has performed an analytical comparison of the heat transfer rate and selected a double tube with a radial finned catalyst tube which thermal radiation rate is over 1800W/m^2°C.

Excessive steam is supplied to the SR so as to react sufficiently and to prevent carbon deposition. Therefore, in the HTTR steam reforming system, a steam to carbon ratio of 3.5 has been selected. If a high performance catalyst can be used at low cost, the steam-carbon ratio could be decreased to utilize steam for electric generation.

The proposed SR is shown in Figure 5.4. 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 endothermic chemical reactor is an essential technology for the production of hydrogen through the use of nuclear heat.

(2) Steam generator in the secondary helium loop for stable controllability and as a safety barrier

A plant where a process heat application system with an endothermic chemical reactor is connected to an HTGR exhibits thermal dynamics that are quite different than those of an HTGR reactor.

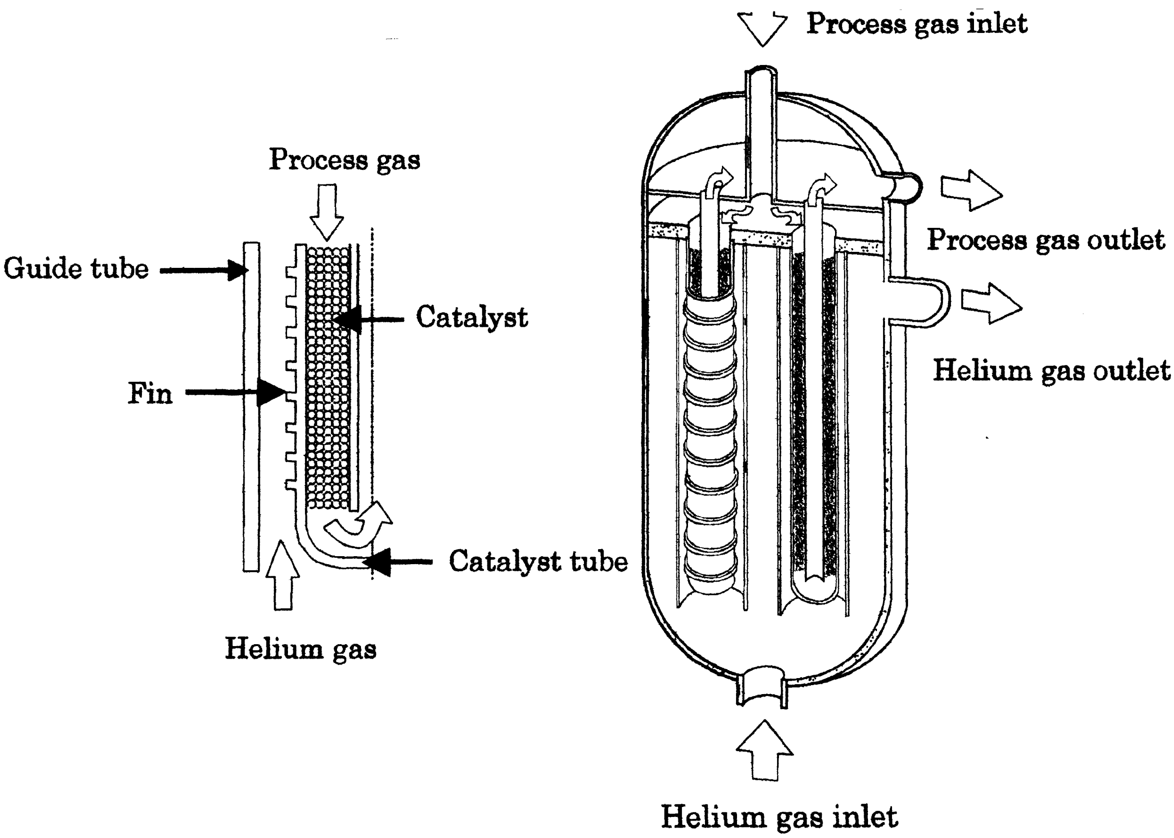


FIG. 5.4. Schematic illustration of the steam reformer.

In an HTGR core, the nuclear generated heat is transformed to sensible heat of the reactor coolant gas and as a result reactor power and helium temperature have a proportional relationship. On the other hand, in an endothermic chemical reactor where an endothermic reaction occurs for the production of hydrogen, the heat input necessary to cause the reaction dramatically increases with increasing reaction temperature due to the Arrenhius type temperature dependence of reaction rate. Development of a new control technology is required in order to balance the difference in the thermal dynamics between the nuclear reactor and the chemical reactor.

JAERI has found that the installation of a steam generator (SG) at the downstream of the SR in the secondary loop provides the stable controllability for any disturbance at the SR due to the large heat sink capacity. For example, when the helium temperature at the outlet of the SR and then at the inlet of the SG goes up due to a malfunction in the process gas feed line, the helium temperature at the outlet of the SG can remain constant at the saturation temperature of steam. A transient analysis result of this is shown in Figure 5.5 for a stepwise decrease in process gas flow rate by 20%. This result indicated that an increased heat input to SG due to increasing the SG inlet helium temperature results in only an increase in steam quality at the saturation temperature due to boiling, but not in an increase in steam temperature.

The operating procedures for startup and shutdown are similar, but reversed. Figure 5.6 shows major process characteristics during start-up. Before start-up, nitrogen is supplied at a pressure of 2.2MPa.

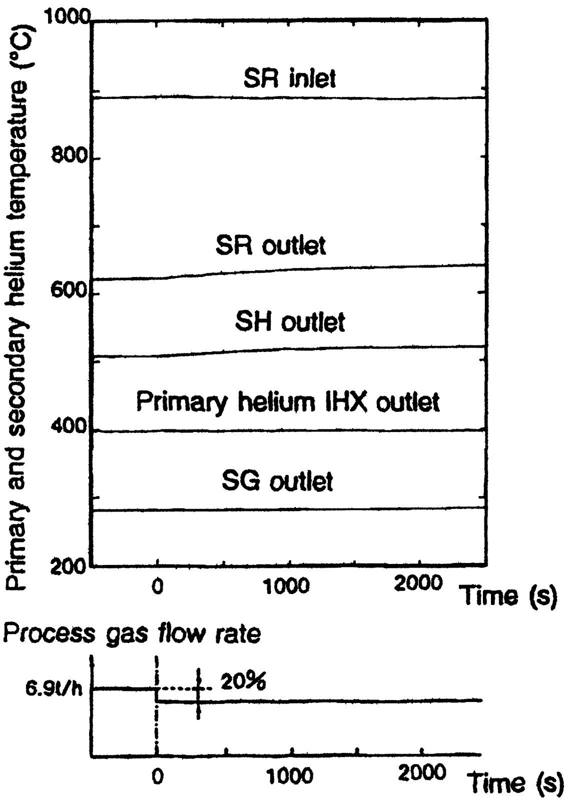


FIG. 5.5. Helium gas temperature at the HTTR steam reforming system by transient thermal hydraulic analysis.

The HTTR is then started. When the secondary helium gas is heated to over 500°C and steam generator is controlled at the rated pressure of 5.0MPa, steam is supplied gradually to the system and nitrogen is released into the environment with this steam by changing the flow line. With the steam flow rate constant at the rated conditions and the helium gas temperature at inlet of the SR increases to 700°C, methane feed gas is supplied gradually to the system. Even during low start-up system operation, a stepwise increase in the feed flow rate by 10% (as it is difficult to control the feed gas at low level), results in stable control of helium gas temperature at the inlet of IHX due to the influence of the SG. After 60h, the helium gas temperature reaches 950°C and the entire system can be operated automatically.

The advantage of the SG is utilized to prevent a reactor scram due to a malfunction or accident within the steam reforming system. Consequently, the secondary helium loop with SG can function as a mitigation system as mentioned above.

(3) Steam generator integrated with the radiator for the final heat sink

A higher probability of malfunction or failure is expected of the hydrogen production system than with the power generation system. This is due to the severe environment of the chemical reactors for the production of hydrogen. A safety measure is required to mitigate the disturbance due to a malfunction or failure of the hydrogen production system to allow for continuous reactor operation without reactor scram.

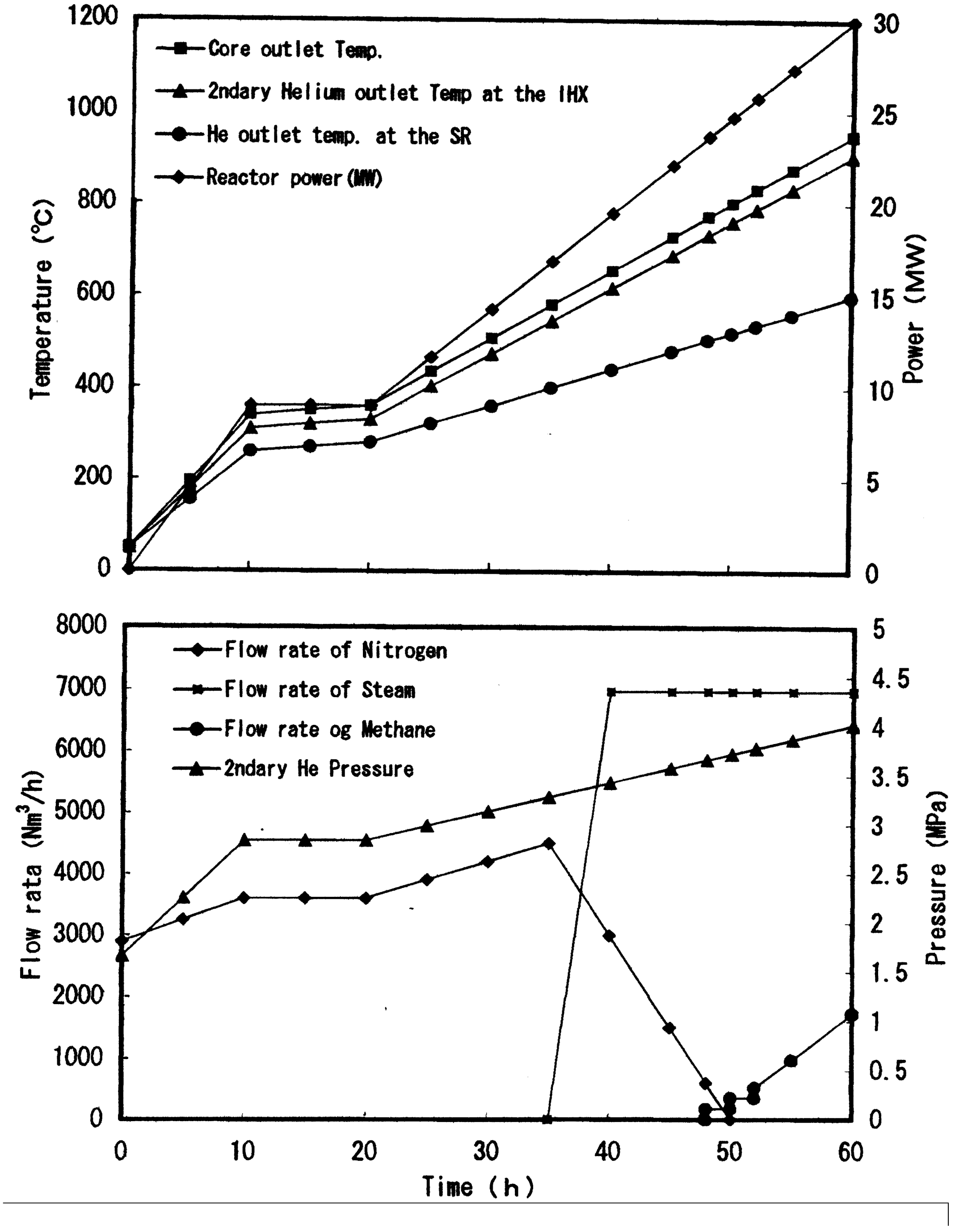


FIG. 5.6. Illustration of the startup profile for the HTTR steam reforming system.

When the feed methane supply system shuts down due to a loss of electric power or a malfunction of control system and results in an increase of helium gas temperature at the outlet of the SR, the SG can cool the hot helium gas to the rated temperature and prevent a reactor scram. But if the feed water stops, the SG cannot continue to operate. JAERI's proposal is that the generated steam be re-used as feed water after condensing in the radiator. The conceptual geometry of this cooling system is shown in Figure 5.7.

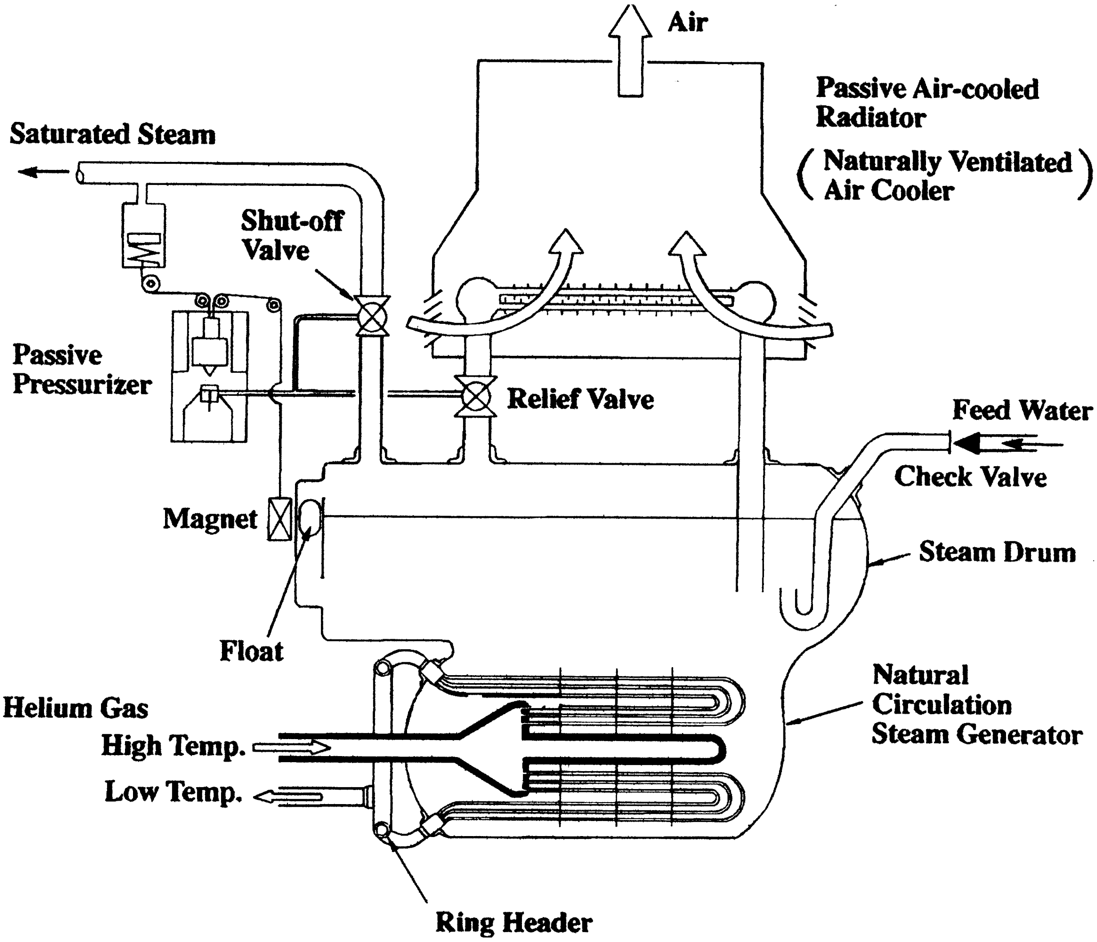


FIG. 5.7. Passive helium cooling system.

In order to prevent a reactor scram due to a loss of feed water to the SG by a loss of power or a malfunction of control system, the hot helium gas is cooled by the SG and the generated steam from the SG flows into a natural ventilation type radiator connected to the SG. This condensed water is then supplied to the SG as feed water. The SG can keep its water contents for normal operation. The heat capacity of the SG and the radiator is ~ 8.8MW. This is the total heat capacity of the SR, the super heater and the SG.

Furthermore, in this cooling system when a pressure drop is detected and water level is low in the SG, the valve in the steam line closes and in the radiator steam supply line opens passively through an automatic air supply system.

5.2.3 Safety requirements

A principal goal of nuclear safety is to maintain the radiation exposure of the public to as low as reasonably achievable in all operational states and accident conditions. Safety items are provided to prevent the initiating events such as malfunction or failure of equipment in order to limit the consequences of anticipated operational occurrences or accident conditions in the HTTR steam reforming system. These safety items are designed to have adequate functions based on the safety requirements.

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 highest class. These items are to have high reliability and redundancy in order to avoid the resulting loss of required safety functions. On the other hand, the items associated with continuous normal operation are categorized into the lowest class and are not required high reliability and redundancy.

It is expected that the steam reforming system connected to the HTTR will not be a nuclear grade system from the viewpoint of economy. Therefore, excessive safety items are not provided in the steam reforming system to prevent the anticipated operational occurrences that may cause damage to safety items or lead to accident condition. Minimum safety items are provided based on the requirements as mentioned below.

Proposed basic design concept 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 with the HTTR reactor system have already been selected. It is therefore important to discuss the additional anticipated design basis events that could originate due to the connection of the steam reforming system with the HTTR.

There are three key areas of concern associated with coupling the steam reforming system to the HTTR. These are:

- The steam reforming system is the final cooling system of the HTTR,
- A large amount of flammable materials are used in the steam reforming system,
- Product hydrogen and methanol will probably be used outside of the nuclear plant in the future.

The three additional anticipated events in the HTTR steam reforming system have been selected as follows.

1) Tritium transportation from the core to the product hydrogen and methanol
2) Thermal turbulence induced by problems in the steam reforming system
3) Fire and explosion due to the feed material and the products

(1) Tritium transportation

Tritium is produced in the core by ternary fission of the fuels and by activation reactions of lithium and boron in the graphite components and control rods during normal operation. In addition to these, helium gas as coolant is a specific tritium source in the HTGR. The coated fuel particles have the potential property to retain the produced tritium within their coatings. Therefore, tritium can be released into the coolant if the coating cracks. On the other hand, tritium produced in the graphite materials can rapidly diffuse through the graphite components at high temperature and be released into the coolant. Tritium in the coolant is removed with other impurities by the helium purification systems provided in each cooling system. However, a very small amount of tritium remains in the coolant and the remaining tritium can easily transport into the product hydrogen or methanol by permeating through the heat exchanger tubes that run at high temperature.

As these products are expected to be alternatives to fossil fuels in order to help the resolution of the environmental problem, it is required that the tritium concentration be reduced in the products below allowable limit in order to treat the hydrogen and methanol as non-radioactive materials. Therefore, the proposed safety requirement for this event is to allow only negligibly low tritium concentration in the products. In this event, the required safety items are not directly related to the reactor safety so they can be classified into lowest safety level.

There are two approaches to reduce tritium concentration in the products; one is removing tritium from coolant as much as possible and the other is protecting against permeation through the heat exchanger tubes. If purification system can be installed directly in the main primary and secondary loops, all tritium will be removed from the helium gas. But, in the HTTR, they are installed in the bypass loop and their flow rates are selected based on concentration of other impurities. Also, it is not desirable to install the purification system in main loops because large equipment would be required to overcome the extensive pressure loss.

Tritium permeation rate depends on the tube surface condition. If the tube surface is clean, tritium permeability becomes high. But if an oxide scale or an effective coating covers the tube surface, permeability becomes low. In the steam reforming condition, oxide scale will develop on the tube surface rapidly. Therefore, it has been determined to be appropriate to combine two approaches in order to reduce tritium concentration. Preliminary calculations have been performed to determine tritium concentration at steady state for 30MW HTTR steam reforming system. The calculation model is shown in Figure 5.8 and the results are shown in Table 5-2.

Table 5-2: Calculation results

Case	Calculation condition				Tritium concentration in product hydrogen	
	Tube surface		Purification rate	Hydrogen		
	IHX	SR,SH,SG	(kg/h)	release	(-)	(Bq/g)
1	Clean	Clean	200	no	1.67×10^{-13}	89.5
2	Clean	Oxidized	200	no	3.83×10^{-14}	20.5
3	Coated	Oxidized	200	no	1.66×10^{-14}	8.86
4	Defect	Oxidized	200	no	2.25×10^{-14}	12.0
5	Coated	Oxidized	400	no	9.96×10^{-15}	5.33
6	Clean	Oxidized	800	no	1.59×10^{-14}	8.50
7	Coated	Oxidized	200	Release	1.27×10^{-14}	6.80

In this calculation, tritium permeability of the IHX is reduced to 1/10 of clean condition and the one of reformer is reduced to 1/100 of clean condition due to the oxide scale of the tube surface developed during operation. From these results, it is found that the oxide scale on the surface of the tubes is effective and the reasonable flow rate of the helium purification systems are enough to reduce the tritium concentration in the products.

(2) Thermal turbulence

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

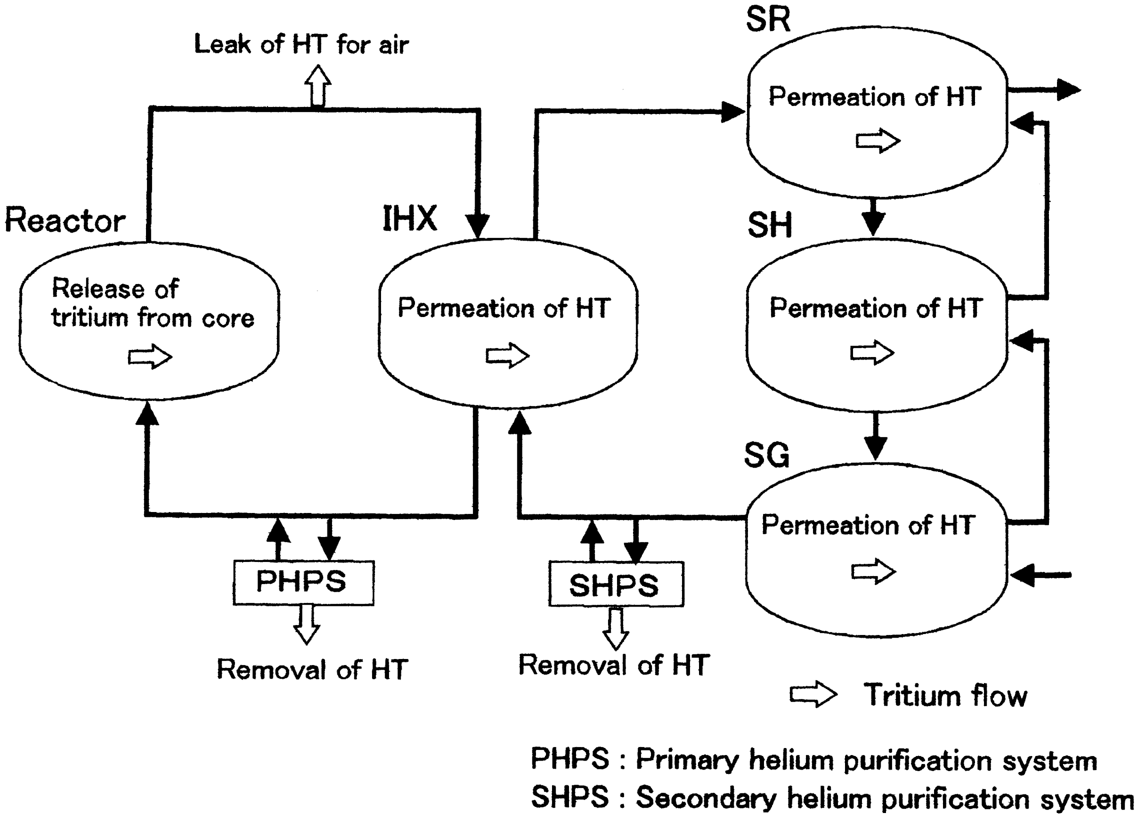


FIG. 5.8. HT balance in the HTTR steam reforming system.

The required function for ternary cooling system is to remove heat from the core during normal operation. Required reliability is not high from the viewpoint of economy. So, problems may occur often during operation lifetime.

The safety design of the nuclear plant is based on the defense-in-depth concept. Therefore, it is required to prevent the propagation of thermal turbulence in the secondary loop as a result of reactor scram. The safety requirement for this event is to limit the secondary helium temperature variation within +/-15°C at the inlet of the IHX to prevent reactor scram.

As shown in the figure of the HTTR steam reforming system layout, the SG is installed downstream of the reformer and has the potential property to stabilize the temperature of content water under constant pressure. This is used to stabilize the temperature returning to the IHX and a passive cooling system using the SG with radiator is proposed to enable continuous normal operation.

The passive cooling system is shown in figure. This cooling system detects a pressure drop due to pipe failure or valve malfunction and low content water level due to interruption of the feed water. When the pressure drop or low water level occurs, the main steam line is closed and the radiator line is opened. Generated steam is supplied to the natural convection type radiator and is cooled down. Condensed water is recycled to the SG as feed water. This system does not require any electric power and feed water.

Static calculations of cooling ability of the SG have been carried out. The results show that the reduction of the feed gas to the reformer, which consists of methane and steam, changes the outlet temperature of the reformer proportionally. But the SG mitigates the temperature variation within 5°C. As a result of continuous cooling the hot helium gas by the SG, it is possible for the HTTR steam reforming system to continue at normal operation.

(3) Fire and explosion

A fire and an explosion resulting from the leakage of flammable materials should be considered because methane and hydrogen are utilized in the steam reforming system. The fire and explosion has the potential of causing significant damage to the safety items so that the safety items related to the fire and explosion should be of the highest safety level.

General safety requirements for the fire and the explosion are already established in the IAEA SAFETY SERIES. In this guideline, it is required that the amount of flammable materials in the plant and in the vicinity of the plant should be reduced as precautionary measures. This design concept cannot apply to the HTTR steam reforming system. The HTTR does not have an inherent capacity to withstand severe radiation heat and blast overpressure resulting from the fire and explosion. It is necessary to avoid the potential possibility of a huge fire and explosion and to provide a distance separation between the accident source and the HTTR. This separation is called a safe distance.

In considering the fire and explosion, three cases need to be addressed, which are a.) inside the reactor building (R/B), b.) in the vicinity of the R/B and c.) far from the R/B, because the required safety requirement and applied safety items are quite different for each of them.

The fire and explosion that originates inside the R/B may cause severe damage to nuclear safety. It is required that the possibility of a flammable gas leak inside the R/B should be low enough to avoid any fire and/or explosion at this location. The potential sequence of flammable gas ingress into the R/B is the simultaneous failure of the secondary helium pipe and the reformer tube. The only cause of the simultaneous failure of these items is an earthquake. These items are designed for a high seismic safety level to avoid simultaneous failure.

The steam reforming system should be designed as a non-nuclear plant so that it is basically not expected to provide additional safety items. When an event occurs in the vicinity of R/B, the thermal load and blast overpressure will be strong enough to cause some damage to the R/B because the inherent capacity of the R/B to withstand a severe blast is not sufficient. Therefore, it is required to prevent significant leakage of flammable gases in the vicinity of the R/B. A double tube has been adopted in the HTTR steam reforming system to prevent leakage of flammable gas. This design concept is also applied for toxic gas in non-nuclear plant. Emergency shut-off valves are also provided to isolate the failure point of the pipe and, thereby, limiting the amount of leakage.

In case of an event far from the R/B, safe distance is available to mitigate the effects of thermal load from fire and blast overpressure from explosion. Comparing to the effect, the explosion causes greater damage than the fire. The explosion is taken into account to estimate safe distance. There are some estimation methods to calculate the effect of blast overpressure. The most prominent of these methods are the TNT equivalent model and a fuel-air charge blast model, such as multi-energy model. These overpressure-distance relationships are very similar for the most conservative calculations. The multi-energy model has been adopted to calculate the distance to meet the allowable limit of overpressure against items important to safety and operational personnel. Furthermore, the effect of movement of vapor cloud by diffusion is considered.

The proposed estimation method to calculate safe distance against explosion is shown in Figure 5.9. The moving distance from the release point by diffusion is calculated by using the Pasquill equation based on the Gaussian plume model. Dispersion parameters in this equation are given in several guidelines. In our calculation, the parameters in the "Yellow Book [68] was adopted.

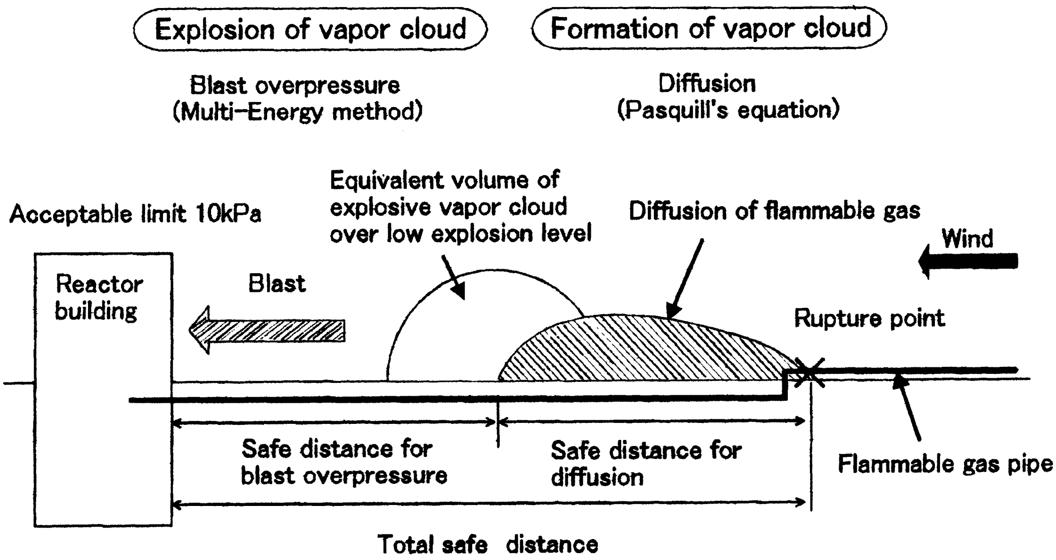


FIG. 5.9. Schematic illustration of the method for estimating the safe distance against explosion.

The vapor cloud contains an equivalent volume amount of explosive vapor over the low explosion level in the spread fuel-air charge. It is assumed that the equivalent vapor cloud of fuel-air charge is a stoichiometric composition and heat of combustion is $3.5 \times 10^6 J/m^3$. The distance needed to meet the allowable overpressure is obtained by reading from the blast charts as shown in Figure 5.10.

The preliminary estimation of safe distance is carried out against vapor cloud explosion in the HTTR steam reforming system. With respect to release conditions, a continuous release and an instantaneous release are both considered within this estimation. A wind velocity of 1m/s and a stability condition of "most stable" are assumed as conservative estimations. If the amount of leakage can be restricted to below 100kg, the required safe distance is only 200m.

5.2.4 Goals of the HTTR demonstration

The HTTR steam reforming system has two major goals. One is to demonstrate a high hydrogen performance from the viewpoint of the competitiveness to a fossil fired system. The other is to demonstrate safe operation from the viewpoint of nuclear safety.

With respect to achieving high hydrogen production performance, it is required to show the following achievements in the HTTR demonstration:

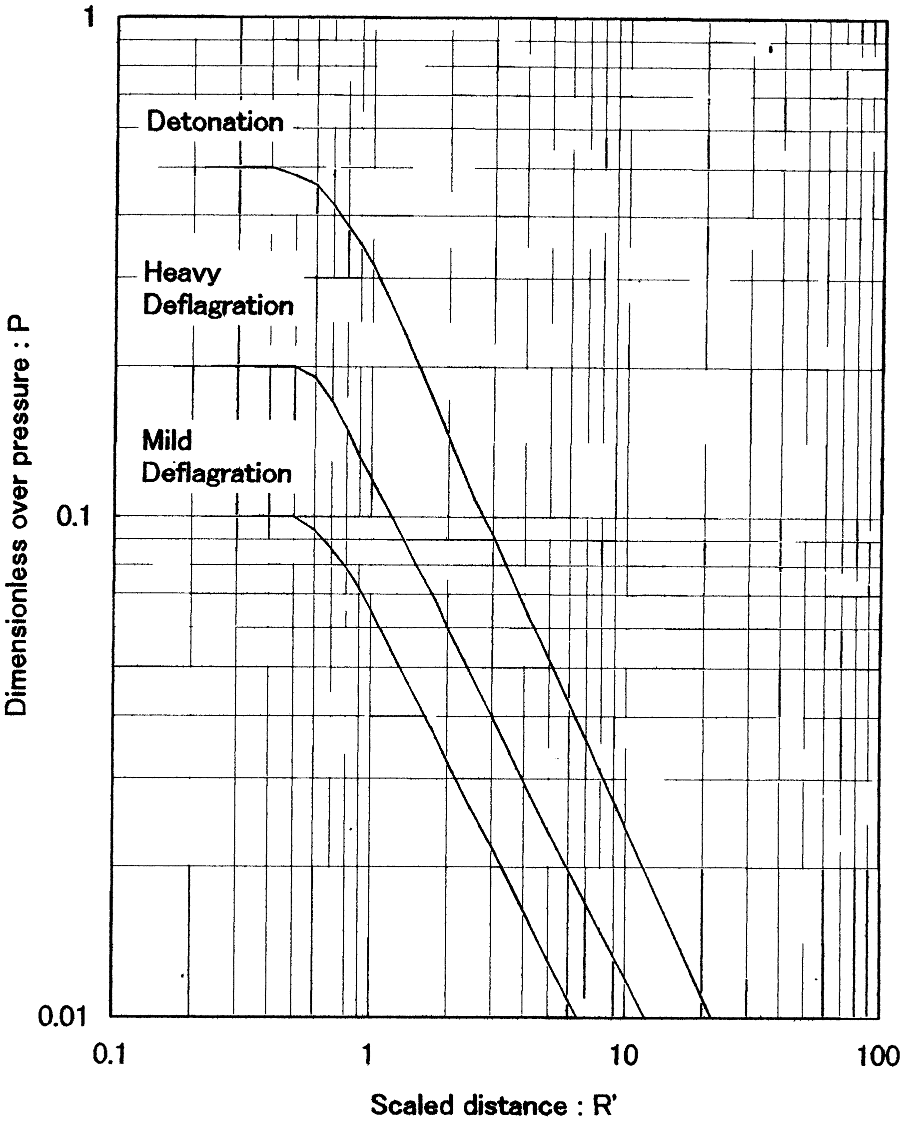


FIG. 5.10. Over-pressure of blast from a hemispherical fuel-air mixture as estimated by the Multi-Energy model.

- A compact and high performance steam reformer
- Low heat loss of the hot gas duct which is installed between the HTTR and the hydrogen production system
- An emergency shutoff valve which can operate at high temperature conditions in case of an event of radioactive release in the reactor containment vessel
- To optimized operating procedures including the feed flow rate
- A hydrogen production rate of > 4,000Nm^3-STP/h
- A thermal energy utilization of ~ 80%.

With respect to safe operation, it is required to show the following achievements in the HTTR demonstration:

- Successful startup and shutdown operation
- Successful mitigation of the thermal turbulence from the steam reforming system to the HTTR by using the SG and the radiator without incurring a reactor scram
- Low tritium contamination in the hydrogen and methanol
- Establishment of the design concept, criteria and regulatory aspects for fire and explosion due to leakage of flammable gas from the steam reforming system.

5.2.5 Alternative designs — German experience

5.2.5.1 Experimental results of the German steam reformer

Research and development work for the steam reforming of methane has been carried out in Germany within the framework of the project Prototype Plant Nuclear Process Heat (PNP), and the project Nuclear Long Distance Energy (Nukleare Fernenergie, NFE). Experimental investigations for this endothermic chemical process have been performed in the experimental plant, EVA-I, and thereafter in the pilot-plant, EVA-II.

The behaviour of a single reformer tube of industrial dimensions with different types of tubes and catalysts has been studied in the EVA-I plant, in which heat is supplied from a 1 MW-electric heater to a helium convective circuit. Also, the main goals of investigation were to determine the heat transfer mechanisms between helium and the process gas as well as the kinetic behaviour of methane and ethane steam reforming. Large process parameters have also been varied for these experimental investigations, including pressure, temperature and flow rate of the process gas and the helium temperature and mass flow at the reformer tube inlet.

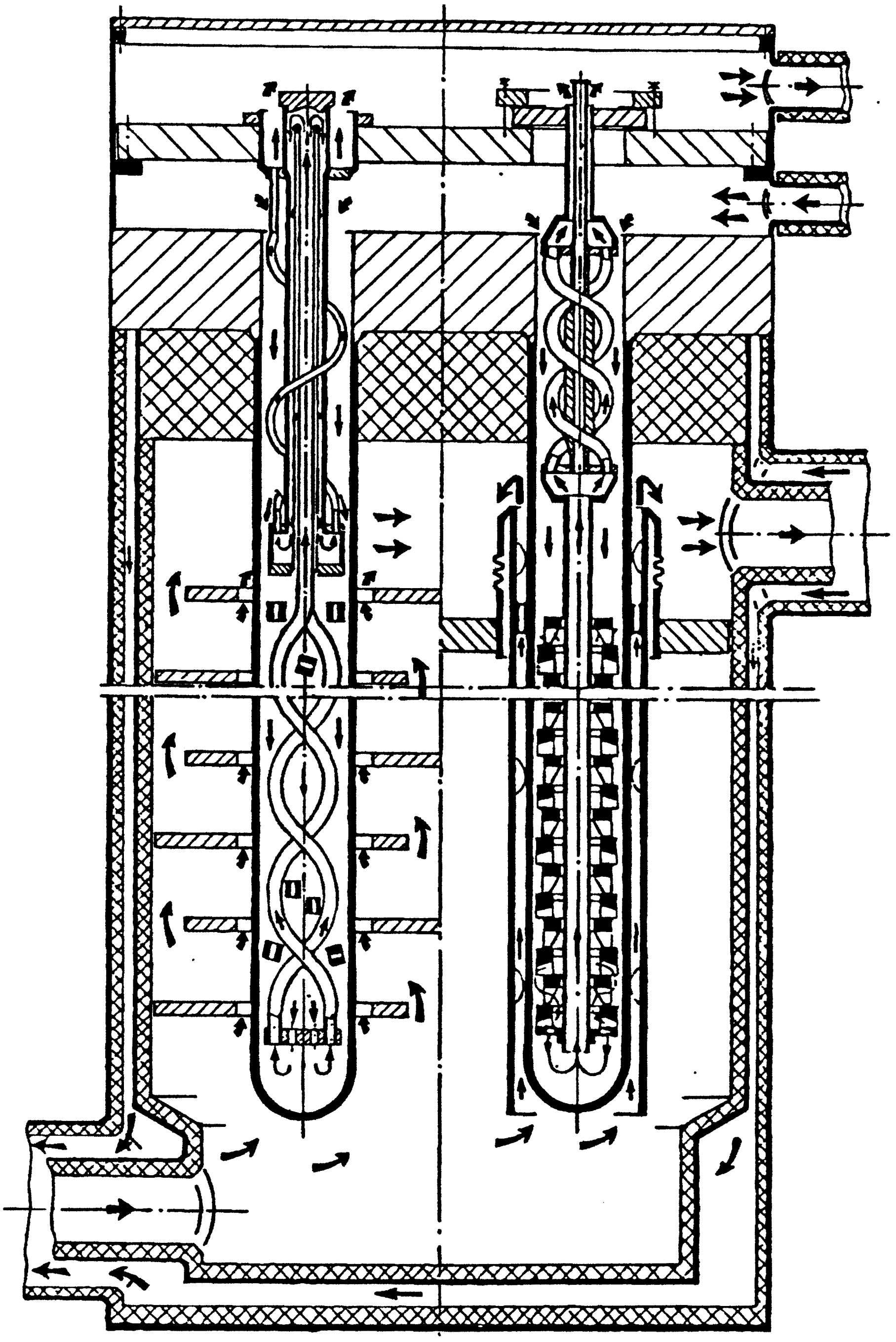
With the experiences gained in the EVA-I plant, a pilot plant (EVA-II) was built at the Nuclear Research Centre in Jülich, Germany, in which two different bundles of full sized reformer tubes have been tested. These bundles had different shapes of helium flow around the reformer tubes and their operational behaviour under the normal and disturbed conditions has been experimentally positively demonstrated. Moreover, the heat and process layout of these bundles has also been experimentally verified.

Variations in the design of the reformer tubes with respect to helium and process gas flow as well as the form of the catalyst have been schematically shown in Figure 5.11, which have been tested as a single tube or as a bundle in the EVA-I and EVA-II plants. In general, hot helium with a temperature of about 950°C flows around the reformer tube from bottom to top in counter flow to the process gas and thereby giving its heat for the endothermic reaction of the steam reforming of methane. Helium flow parallel to the reformer tubes is achieved with concentric guide tubes, which have been tested for a single tube in EVA-I plant and as a bundle in EVA-II plant. Axial and cross flow of helium has been obtained by the use of a large number of baffles in the bundle and this bundle has been tested in the EVA-II plant.

The entrance and exit of the process gas to every reformer tube is at the upper end of the bundle near the tube sheet. This requires an internal return tube, the so-called internal pigtail, for each reformer tube which can be either straight or coiled.

There is a further possibility to arrange a recuperator in every reformer tube, so that the cold process gas can be heated through the reformed product gas before it enters the catalyst region as shown in Figure 5.11. Thereby, it is further possible to keep the temperature of the entrance and exit chambers for the process gas of the reformer bundle at a lower temperature level. However, this recuperator was arranged only in a few experimental investigations of the EVA-I and EVA-II plants.

Moreover, the catalyst region can either be a Raschig-ring bed or a disk catalyst column as shown in Fig.5.11. Further tests have also been performed in the EVA-I plant with different types of catalysts e.g. baskets filled with conventional Raschig-rings. The main goal



Left side: Reformer tube bundle with baffles and Raschig ring bed catalysts

Right side: Reformer tube bundle with concentric tubes and disk catalysts

FIG. 5.11. Schematic of a steam reformer [7].

was to achieve high process performance of the steam reforming of methane as well as to provide an easier replacement method for the catalyst after end of life of the reformer tubes.

These different catalyst concepts have been experimentally verified under identical operational conditions in the EVA-I plant and it was determined that the Raschig-ring bed concept had the best results with regard to the heat transfer coefficient and the reforming temperature, i.e. the process gas temperature at the exit of catalyst region and the measured methane conversion value. However, this measured CH_4-conversion value is lower than the possible equilibrium value at this measured reforming temperature and corresponds to an ~ 10 K lower reforming temperature.

The disk catalyst concept also provided good heat transfer coefficient and thereby also exhibited the same reforming temperature as the Raschig-ring bed, however it's measured CH_4-conversion value corresponds to ~ 40 K lower reforming temperature in comparison with 10 K with the Raschig-ring. This was due to the lower efficiency of this catalyst concept. The basket concept could not achieve the same reforming temperature because of the boundary flow between the reformer tube wall and the basket and this value was ~ 10 K lower than the reforming temperature with the Raschig-ring bed and therefore also had a lower measured methane conversion.

The replacement method for the Raschig-ring bed concept still has to be improved in order to achieve the same low time value of a few minutes as required with the concepts of the disk and basket catalysts. The hydraulic process tested in the EVA-I plant was the first step in this direction, which has to be further developed to fulfil the requirements of the nuclear heated steam reformer bundle.

The helium heated steam reformer corresponds more to the design of a large heat exchanger bundle as shown in Figure 5.11. Therefore, the heat transfer mechanisms have been tested in the EVA-I and EVA-II plants and were also improved to realize higher efficiency of methane conversion.

Two types of helium flow around the catalyst tubes are shown in Figure 5.11 through the arrangement of baffles and concentric tubes. The heat transfer correlation for these arrangements have been experimentally verified. In the first case, helium flows longitudinally along the reformer tube length through the concentric gap between the guide tube and the catalyst tube. The heat transfer correlation of this longitudinal flow is generally well known in literature and is based primarily on thermal convection, and also partly with the thermal radiation between the guide tube and catalyst tube. Experimental results can be expressed with the following correlation in which thermal radiation is also considered:

$$Nu = c \cdot Re^{m} \cdot Pr^{n}$$

Where c = 0,205; m = 0,603 and n = 0,33 are estimated, and the characteristic numbers of Nusselt, Reynolds and Prandtl are defined according to the literature. Moreover, this heat transfer coefficient can be increased by decreasing the width of the concentric gap. However, on the other hand, this change has to be compensated with a higher pressure drop for the helium flow. In the EVA-I plant, this concentric gap for a different single reformer tube arrangement was between 10 to 15 mm and thereafter for the PNP reformer bundle in the EVA-II plant it was optimized to the value of 9 mm.

In the second case of helium flow, a combination of axial and cross flow through the arrangement of baffles is realized. Different types of baffles have been used for the general design of shell and tube heat exchangers, where only an integral average heat transfer coefficient can be given on the basis of different experimental results. However, it is difficult to calculate the local heat transfer coefficient, because it depends on the pattern of flow for a particular place. On the other side, the knowledge of the local heat transfer coefficient is not important, because if any change of local helium temperature occurs, it will automatically be diminished through its further flow along the reformer bundle length, whereby getting thoroughly mixed. The reformer bundle tests in the EVA-II plant utilized disk and doughnut baffles, and the results of the heat transfer coefficient for the helium flow can be described through the following equation:

$$\mathrm{Nu} = c \cdot \mathrm{Re}^{m} \cdot \mathrm{Pr}^{n};$$

Where $c = 1{,}612$; $m = 0{,}502$ and $n = 0{,}33$ are estimated. There are still measured values which deviate from this equation that cannot be totally avoided.

The heat transfer correlation on the process gas side can also be determined in the same simple way as previously shown for the helium side with the following equation:

$$\mathrm{Nu} = c \cdot \mathrm{Re}^{m} \cdot \mathrm{Pr}^{n}$$

Where, for both reformer bundles tested in the EVA-II plant, i.e. with baffles and with concentric tubes, $c = 3{,}12$ and $m = 0{,}413$ are estimated. As the Prandtl number can change in a large range with different feed-gas compositions, the estimated value is 0,4. For the calculation of the Nu-number and Re-number, an equivalent particle diameter has been taken as the characteristic diameter and this can be estimated from the geometry of the Raschig-ring catalyst. Moreover, the velocity of the process gas in an empty reformer tube without a catalyst has to be considered in this correlation. Also, the area occupied through the integral pigtails has to be subtracted by this calculation.

To more precisely determine the heat transfer coefficient on the process gas side, it is necessary to also consider the geometry of the catalyst bed including the form of the internal pigtail as follows:

$$\mathrm{Nu} = 0{,}172 \cdot S \cdot (d_p/D_h)^{0,2} \cdot \mathrm{Re}^{0,75} \cdot \mathrm{Pr}^{0,4}$$

Where:

S = 1 for a straight internal pigtail
S = $1 - (2 \cdot A_{Pr}/A_i)$ for helical pigtails
A_{Pr} = helical pigtail projection area on inner surface area of catalyst tube A_i;
d_p = equivalent particle diameter for the Raschig-ring catalyst
D_n = $4 \cdot A/U$
A = A_{RTI}/A_{PTO}
A_{RTI} = cross-section, reformer tube, inside
A_{PTO} = cross-section, pigtail, outside
U = U_{RTI}/U_{PTO}
U_{RTI} = circumference, reformer tube, inside
U_{PTO} = circumference, pigtail, outside

Re = $d_p \cdot w \cdot \rho / \eta$
W = w_E/ε
W_E = average velocity in empty reformer tube
ε = void fraction of the catalyst bed
ρ = process gas density
η = process gas dynamic viscosity

In the EVA-I-plant, steam reforming of natural gas has been mostly tested, including some heavy hydrocarbon gases that are present in the feedgas mixture, e.g. ethane. Therefore, this also has to be considered by determining the reaction kinetics of methane reforming. However, from the results of the experimental investigations, it can be noted that ethane conversion is much quicker than the conversion of methane with steam and, after passing of the feedgas through the catalyst region of about 1 m length, almost 30 to 40 % of the ethane conversion has been completed. The feedgas has the temperature of about 500°C at the inlet of this region. Moreover, after about 4 m length of this catalyst region, the ethane is totally decomposed.

On the basis of methane reforming reaction

$$CH_4 + H_2O = CO + 3H_2$$

the equation for the reaction rate can be correlated in a simplified way as follows:

$$r_{CH_4} = k_{CH_4} \cdot p_{CH_4} \cdot \text{Bremsterm (Reaction suppression term)}$$

The results of the experimental investigations have shown that the reaction rate constant k_{CH_4} is proportional to feedgas throughput, and also according to the Arrhenius equation, i.e. proportional to reciprocal temperature. The explanation of this behaviour can be described in the following manner. Chemical conversion is effected through the combined processes of reaction and transport. The reaction process is independent from the feedgas velocity. However, similar to the heat exchanging mechanism, the outer transport resistance is velocity dependent. This is based on the diffusion barrier caused through the formation of a film at the outer catalyst surface area. Therefore, any decrease of velocity causes larger film formation and, as a consequence, higher transport suppression. Moreover, the following value of k_{CH_4} has been determined from the experimental results:

$$k_{CH_4} = k_O\,(w - 1{,}5 \text{ m/s}) \cdot \exp(-E/R \cdot T)$$

where:

k_O = $2{,}64 \cdot 10^6$ $m_N^3/(m_{cat}^3 \cdot h \cdot bar \cdot m/s)$
w = Velocity of the feedgas in catalyst bed
E = Activation energy = 73,5 kJ/mol
R = Gas constant
T = Absolute temperature °K
V_{cat} = Volume of the catalyst (m_{cat}^3)
V = Volume of the catalyst region i.e. inner volume of the reformer tube
V_{cat}/V = $1 - \varepsilon = 0{,}534$
ε = void fraction of the catalyst bed

Reaction suppression term (Bremsterm)

$$= \quad 1 - p_{CO} \cdot p^3_{H_2} / p_{CH_4} \cdot p_{H2O} \cdot k_p$$

p_i = partial pressure of the different gas component "i" (bar)
k_p = equilibrium contant

This reaction suppression term at the beginning of the catalyst region is about 1, and at the exit of this region its value is between 0,05 to 0,2, i.e. the equilibrium of the methane conversion is nearly approached at the measured exit temperature.

The reaction rate of methane conversion, r_{CH4}, increases very quickly at the beginning of the catalyst region because of the high methane content in the feedgas as well as through the increase of the feedgas temperature. It reaches its maximum before the middle of the reformer tube length. Thereafter, this reaction rate decreases although the temperature of the process gas increases. This is due to a decrease in the reaction suppression term at the exit of the catalyst region and, as previously mentioned, is more dominant for this reaction rate. From the results of the experimental investigations it has been found that the reaction rate reduced almost linearly along the reformer tube length. By these experiments it was difficult to measure the required parameter data at the inlet catalyst region, so that the reaction rate of methane could not be determined correctly in this region.

Independent to the results of the methane reaction rate, it has been found that the methane conversion at the end of the reformer tube length approached nearly equilibrium conditions according to the measured process gas temperature at the exit. The measured methane conversion value is only about 1 to 2,5 % lesser than the equilibrium value at the measured reforming temperature at the exit of the catalyst region, as shown in Figure 5.12.

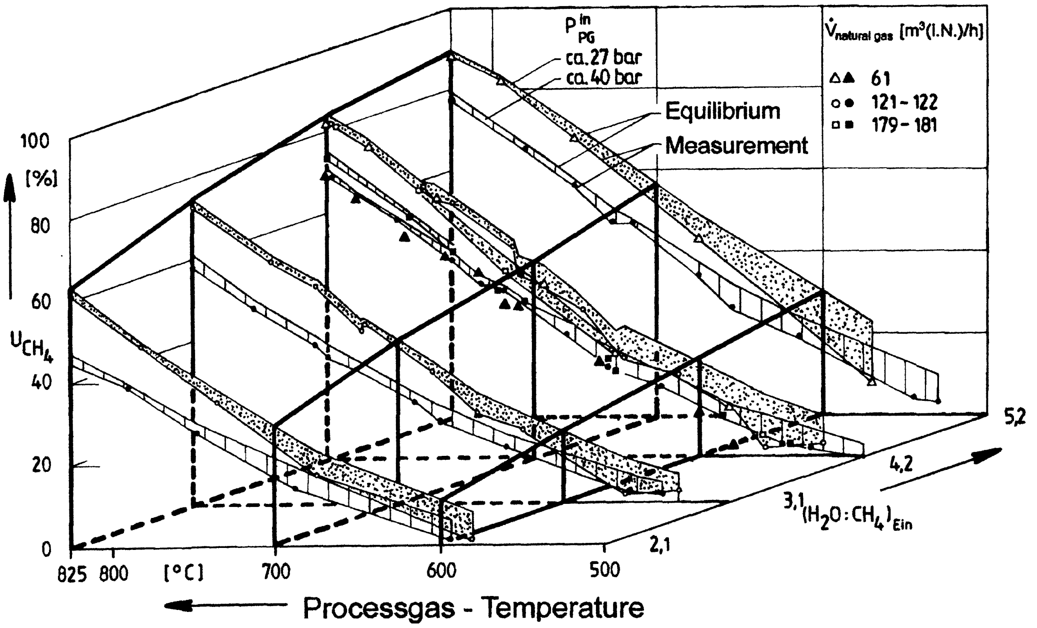


FIG. 5.12. Methane conversion as a function of process gas-temperature steam to methane-ratio and system pressure at the inlet [8].

The temperature difference between this measured temperature and the equilibrium temperature according to the measured methane conversion value has been called the “Approach-Temperature”. Results of the EVA-I-plant had shown that this value lies between 4 to 10 K.

Experimental investigation of the reaction kinetics has also been performed with the addition of carbon dioxide to the feedgas for the steam reforming of natural gas. This was done to change the composition of the produced synthesis gas of the methane conversion in such a manner that its further synthesis to methanol and higher alcohol can be more suitably performed with the required specifications of low H_2/CO-ratio, which is not possible in the normal process without CO_2 addition.

The Arrhenius representation of the reaction rate constant is shown in Figure 5.13. It can be shown that methane conversion with the addition of carbon dioxide in the feedgas stream can be described with the same kinetics relation as the earlier tested pure steam reforming of natural gas without any addition. Also in this figure, the results of two different catalysts with different specific outer surface areas have been shown, although with the bulk volume value of the catalyst remaining the same. Therefore, the reaction rate constant of the BASF-catalyst with a smaller surface area must be higher in value (as shown in curve 2a) if the same activity in the reformer tube volume has to be obtained. Moreover, as shown in this figure, the activity in the inlet region until ~ 700°C is smaller, and at the reformer tube exit it is higher. From this figure it can be shown that at a temperature of 800°C, the reaction rate constant for both catalysts has the same value.

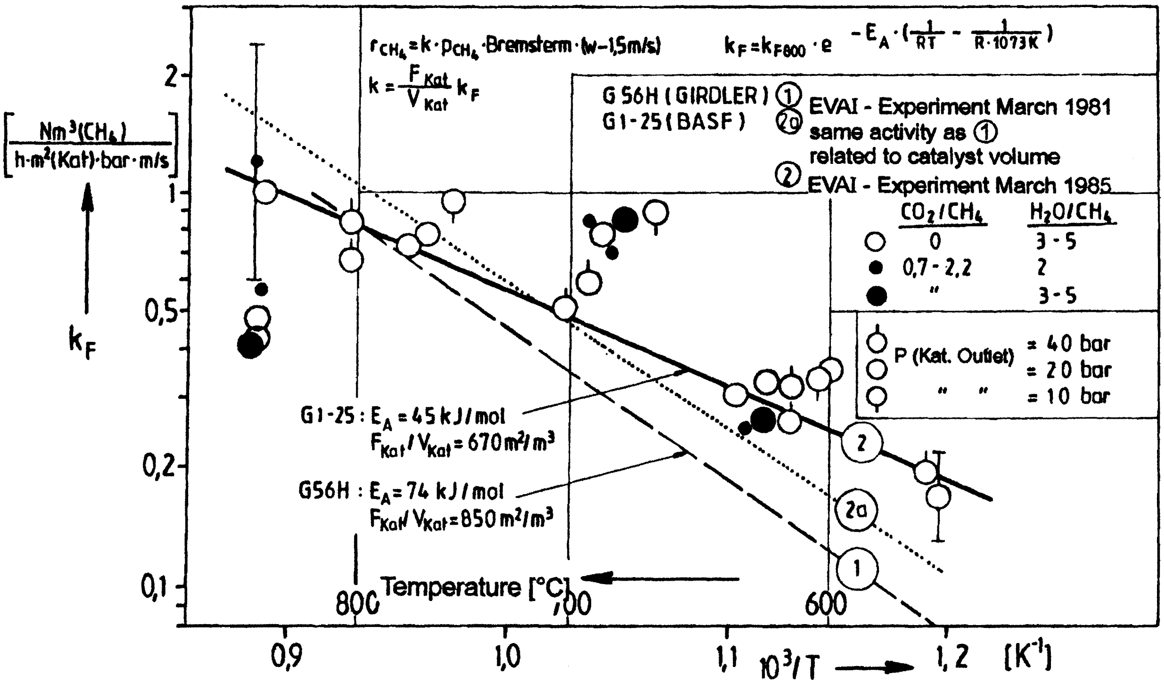


FIG. 5.13. Arrhenius representation of the reaction rate constant of methane conversion related to the catalyst outer surface area [11].

$K_F\ 800 = 0{,}8\ m_N^3 / (m_{cat}^3 \cdot h \cdot bar \cdot m/s)$

The analysis of the experimental results of CO_2 addition in the feedgas stream of the steam reforming of natural gas shows that with increasing values of the CO_2/CH_4-ratio c at the start of the chemical reaction, the formula turns over as shown in Figure 5.14. Thereby, the two reactions, i.e. steam reforming and CO_2-reforming of methane, are competing with each other; i.e. the first reaction goes down and the second reaction goes up equivalently if c increases (4 measurement points shown in Figure 5.14). Therefore, the total reaction, which is the sum of these two reactions, is slightly increasing. This means that the capability of the steam reformer to perform these two reactions in parallel is not reduced, even under the same operational conditions and without any change of the catalyst. Moreover, in general, through this addition of CO_2, the ratio of H_2/CO in the produced synthesis gas is decreasing, which is more suitable for its further synthesis to methanol. Therefore, it has been suggested from the research centre Jülich, that this version of steam reforming of methane should also be tested in the HTTR-plant as methanol may become important in the near future as an alternative energy carrier parallel to hydrogen. Moreover, CO_2-rich natural gas is also abundantly available as natural resources in different countries of the world, and can also be used for future energy demand.

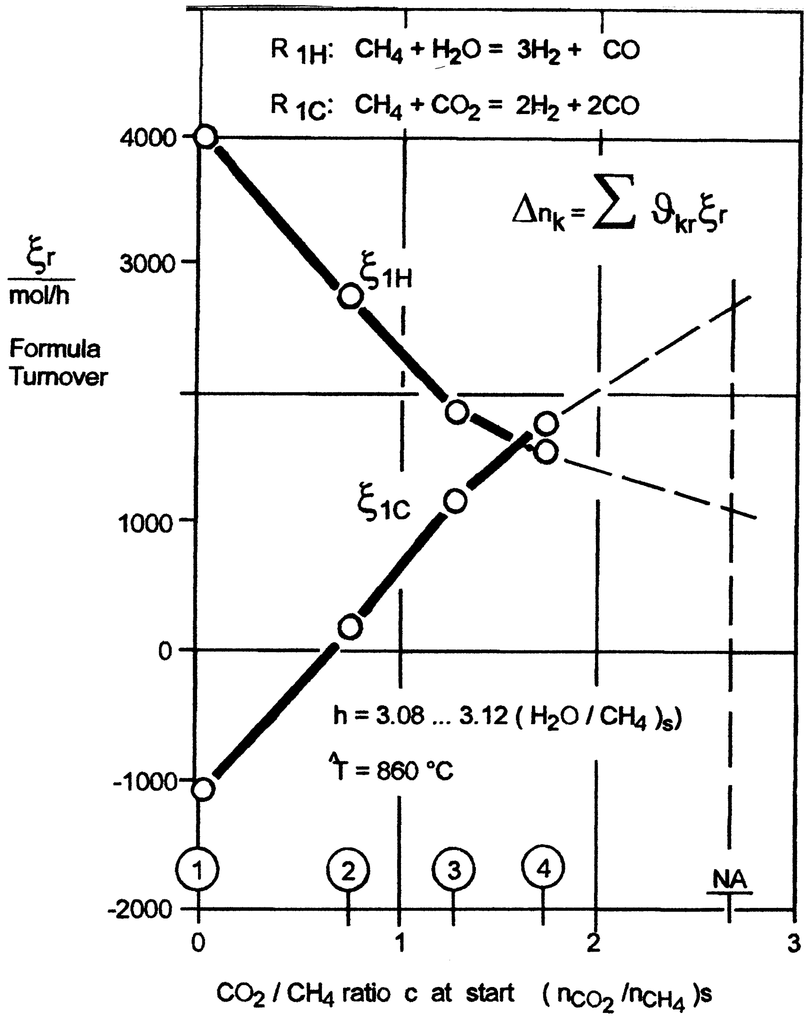


FIG. 5.14. H_2O plus CO_2-reforming of CH_4 formula turnover vs. CO_2/CH_4 ratio [10,15], NA: C=2.65 for the Natuna gas field.

Analysis and experimental results have shown that the reforming temperature at the exit of the catalyst region by steam reforming of methane has a much more dominant effect by the formation and sedimentation of carbon than the presence of carbon dioxide in the feedgas, or of the system pressure of this chemical process. As shown in Figure 5.15, an increase of the reforming temperature, which is 10 to 20 degree higher than the equilibrium temperature, will also increase the risk of carbon formation and it's sedimentation. Corresponding to the equilibrium temperature, minimum value of the H_2O/CH_4-ratios are necessary so that carbon formation can be avoided.

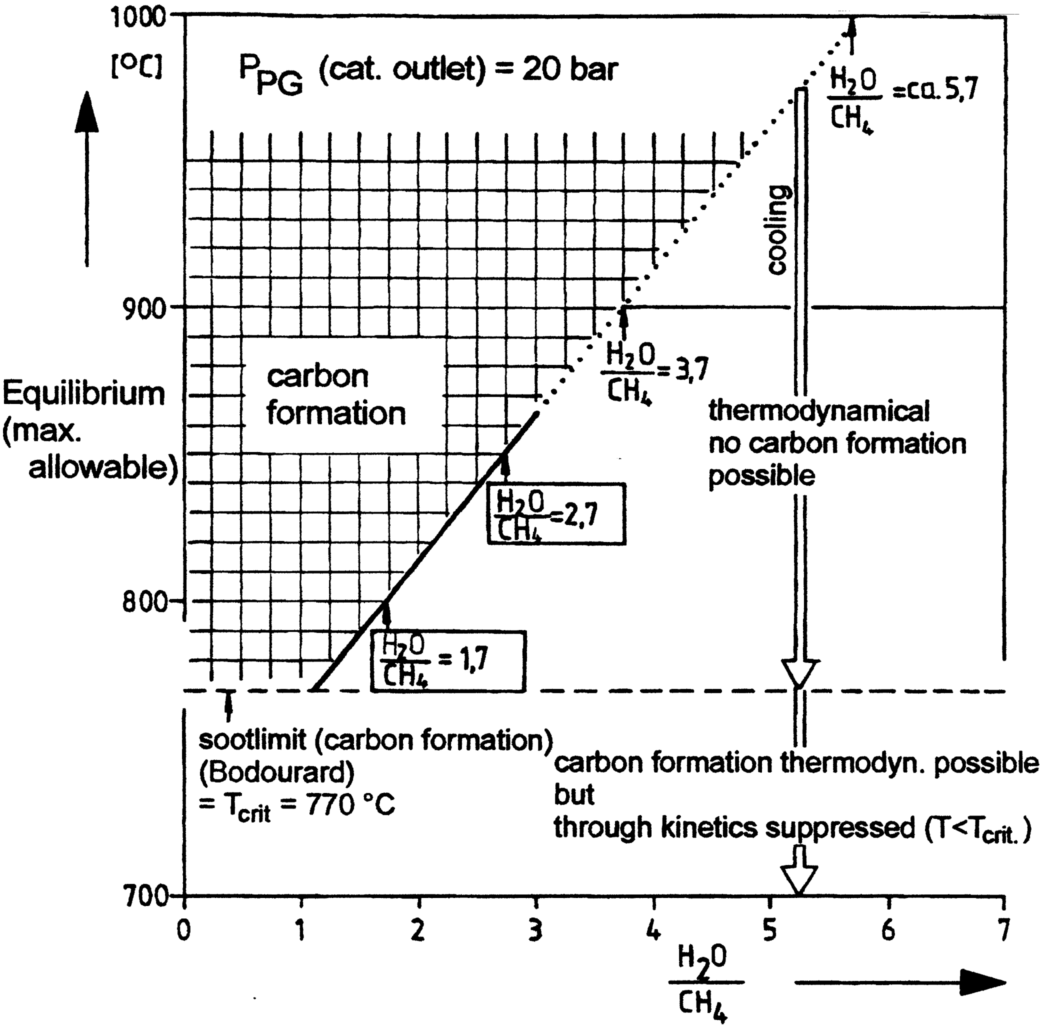


FIG. 5.15. Maximum allowable reforming temperature at the exit as a function of the mole ratio H_2O/CH_4 (equilibrium condition) [11].

5.2.5.2. Safety concepts of HTR-plants for process heat applications

Increased safety requirements are necessary for the HTR-plant for process heat applications in comparison to the electricity generation plant, because of the following reasons:

- The product(s) of the plant are primarily in gaseous or liquid form and these can carry some radioactivity out of the plant boundaries as a result of permeation through the heat exchanger walls and, subsequently, its further transport through the product lines to the consumer market. Through its consumption or its further treatment, some radiation exposure can occur.

- Through endothermic chemical processes, mixtures of burnable gases are produced in the components of the heat utilization system and, therefore, due to any leakage these gases can be released to the surroundings inside or/and outside of the reactor building, which may cause an explosion when mixed with air.

Therefore, safety concepts are required to handle these safety problems, so that contamination due to radiation exposure from such radioactive material, e.g. tritium, by the consumer may be avoided or, at least, its effects can be mitigated. On the other side, consequences of the release of such burnable gases can be controlled through safety measures depending on the arrangement of the chemical conversion components inside or outside of the reactor building. In the German design, the steam reformer for the conversion of methane is directly arranged in the primary helium circuit. Therefore, safety measures were designed accordingly.

The main sources for the release of tritium in the HTR-plant are based on impurities in the graphite components, e.g. lithium, helium coolant and boron contained in the control rods or in carbon blocks.

Two main sinks for the tritium released in the primary helium are the gas purification system and permeation through the heat exchanger walls. The purification system is based on the molecular sieve arranged in a bypass helium flow. Permeation is primarily influenced through the oxide layers on the tube walls, mostly on the secondary side due to the oxidizing conditions prevailing at this location.

Tritium transport behaviour depends on the partial pressure of hydrogen in the primary helium circuit, which can be reduced by enhancing the efficiency of the gas purification system. Many efforts were made in the past to achieve this goal. Parallel to the conventional method based on the copper oxide bed and molecular sieve, a promising alternative system has been tested in the Research Centre Jülich, which is based on the getter technique, e.g. with “Cer Mischmetal”. With this developed technique, it was possible to achieve higher purification throughput rates and, at the same time, only low energy inputs were necessary [16]. A long term test of the “Cer-Mischmetal-Getter” for the purification of hydrogen, tritium and other impurities in a large scale experiment with the AVR-plant at an operational temperature of 300°C was carried out in 1988, and the results were very promising. However, this experiment could not be completed for the total getter lifetime because the AVR-plant was shutdown at the end of 1998 [17].

A further alternative purification system was also developed in the Research Centre Jülich, where selective and continuous separation of tritium with a new type of permeation filter concept was possible. The behaviour of tritium permeation will be described later. Wall materials for the interchanging surfaces, which were based on metals with high tritium permeability properties, e.g. Pd or Pd-Ag-alloys, were used. This can be arranged in the primary helium circuit so that part or all of the helium flow can be purified by this filter. Experimental investigation was also performed for this system with positive results [18].

Many experimental investigations have been performed in the German R&D programme for enhancement of the permeation barrier through oxide layers. At the Research Centre Jülich, many experiments under realistic geometries and boundary conditions were systematically performed to determine the permeation behaviour of tritium, deuterium and hydrogen (protium). Thereby, different materials are tested under different temperatures and prevailing hydrogen partial pressures. Some results of permeation rate are shown in Figure 5.16 [19] and in Figure 5.17 [20] under consideration of the oxide layers. Thereby, it can be seen that the permeation rate is proportional to the square root of the offered pressure of the hydrogen or deuterium ($\sqrt{p}$) in the lower pressure range. In Figure 5.18 [21], the correlation between the hydrogen pressure and permeation for the oxide layers in comparison to the blank metal is shown. Moreover, the positive effect of the temperature treatment as well as of the pre-annealing with hydrogen for the prepared oxide films before operation, can also be recognized.

A high Hemmfaktor (inhibitive factor) can be realized with these oxide films on different materials. This gives the ratio of reduced permeation rate with these protective layers of 40 to 300 for Inconel 617 and 50 to 900 for Hastelloy X.

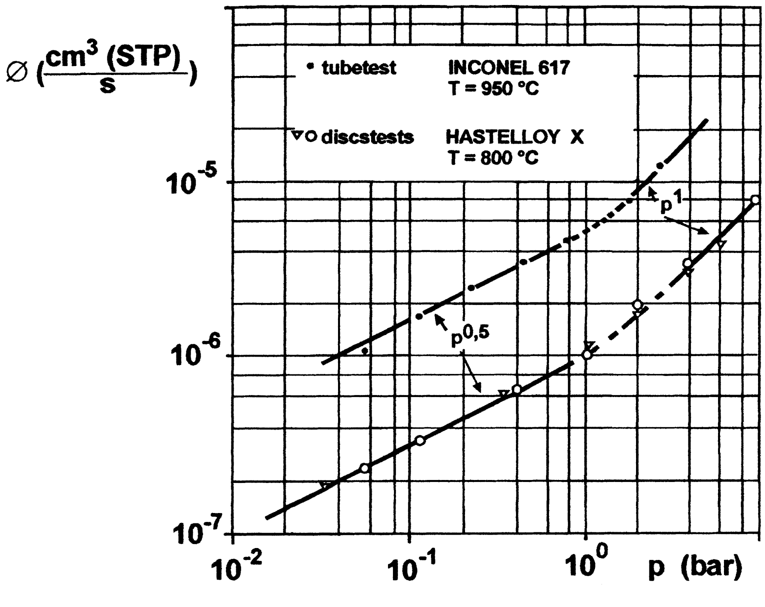


FIG. 5.16. Permeation rate as a function of partial pressures with oxide films [19].

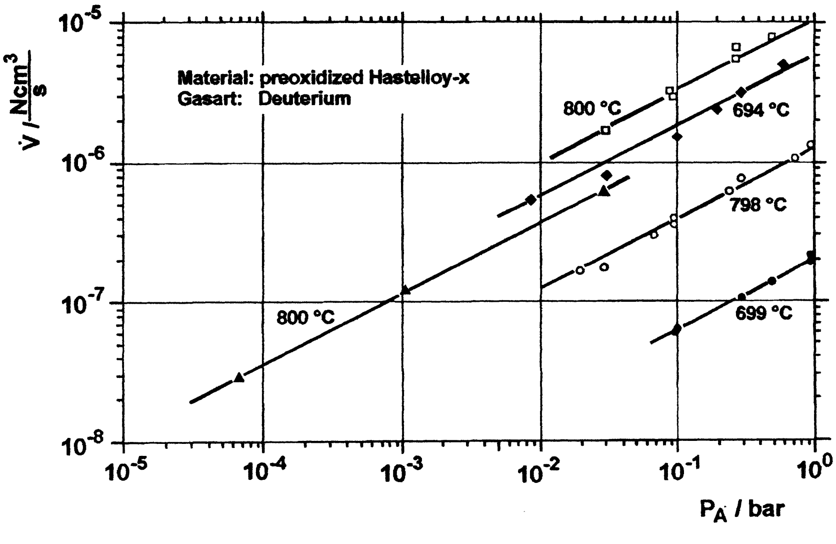


FIG. 5.17. Permeation rate "V" as a function of partial pressure under different conditions of oxide layers [20].

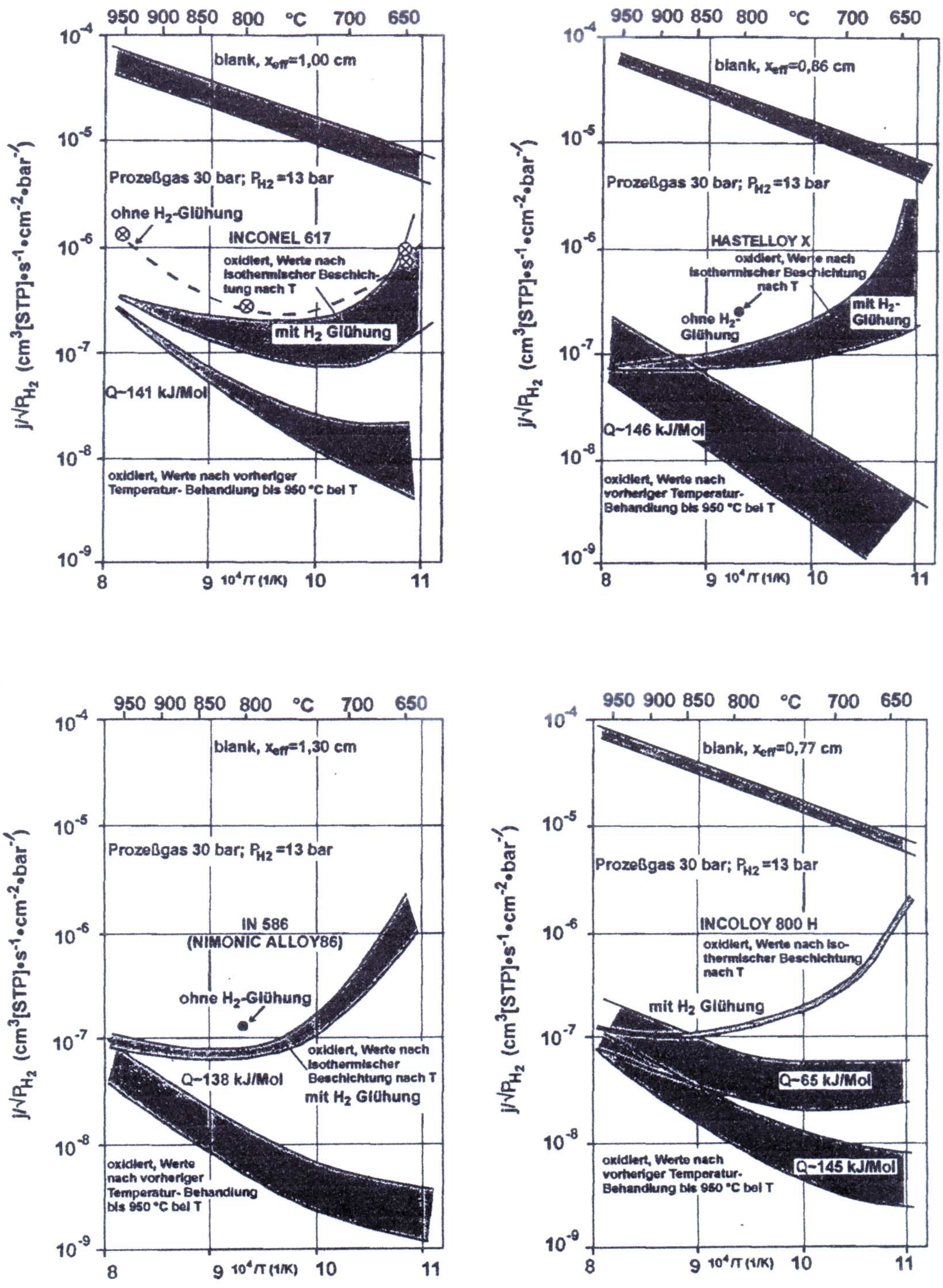


FIG 5.18. Permeation flow as a function of temperature for different materials and preparation of oxide film with prior temperature treatment and annealing with H_2 [21].

One of the goals of the AVR-reconstruction and its following operation was to demonstrate the restraint characteristic of the nuclear plant for tritium by using HTR-heat for chemical processes. Thereby, it was necessary to find whether the use of the product gas from this plant is subject to authorization.

On the basis of measured data of the AVR-plant, the anticipated value of the source of tritium in normal operation was fixed at about 1,2 MBq/s and of hydrogen to be about 0,9 cm^3(STP)/s. Therefore, the reference data for the gas purification constant was 0,03 h^{-1} [22].

On this basis, the tritium balance in the AVR reconstructed plant has been calculated and is shown in Figure 5.19. Steam for the steam reformer was supplied from the process steam generator and was contaminated. The product gas was supposed to be pure methane so that the feedgas methane was not contaminated. However, this was not used as the feedgas, and as shown in Figure 5.19, the product gas methane was contaminated to a value of about 1,5 Bq/g during the normal operation.

This calculated value has to be compared according to the legal Guidelines of Radiation Protection Ordinance (StrlSchV) § 4 (4) 2C, which defines the free limit in Germany for tritium for further manufacturing to a value of 10 pci/g = 0,37 Bq/g. This value was valid at the time of these project studies. However, since then, this value has been increased to 5 Bq/g for the products of coal gasification, because of the low radiological toxitate of this nuclide. Therefore, the product methane of the AVR reconstructed plant can fulfill these requirements during normal operation. However, higher values are anticipated during start-up, because of the damage of the oxide film. Therefore, for short periods, e.g. a few hours, higher contamination values of the product gas are to be considered, which may still be under the new free limit value of 5 Bq/g. Highly efficient gas purification systems can be planned for future projects. Also, in the AVR-plant, the carbon structure utilized higher impurities of lithium, so that future plants can better fulfil these legal Guideline requirements.

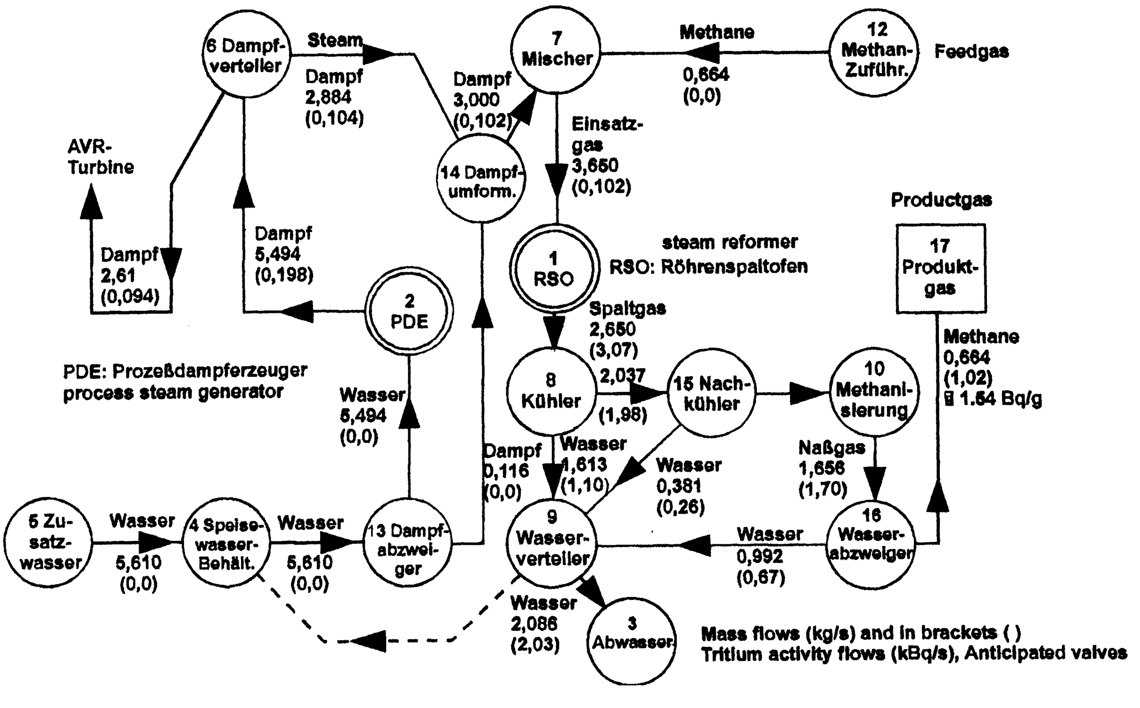


FIG. 5.19. Tritium balance [22].

Control of the release of burnable gas mixtures inside and outside of the reactor building is described in the following safety concepts.

Components which form the barrier between the primary and the secondary circuits must fulfill the safety requirements during normal operation and also under accident conditions, so that the possibility of any ingress form one side to other is kept very low.

Therefore, the qualification of the material should be based on creep behaviour, fatigue properties, structural stability and corrosion resistance. Moreover, through design criteria, the primary and secondary stresses should also be kept at low levels.

General reactor safety measures are necessary for all accident conditions, where reactor shutdown, shutdown of the primary circulator and closing of its isolating valve and the isolation of all secondary circuits are required.

Safety concepts to control the burnable gases inside the reactor building require active and constructive measures through which the formation of the inflammable mixtures within the building can be prevented. Active measures limit the ingress of process gas in the primary system, whereas constructed measures prevent the ingress of the burnable gases inside the reactor building. These counter measures depend on the design arrangement of the components containing this burnable gas mixture in the primary and in the secondary circuit.

In the AVR reconstructed plant, the steam reformer is directly arranged in the primary helium circuit and is located inside the reactor building, whereas the steam reformer vessel is connected with the gas fabrication plant with connecting pipes at the inlet and outlet. Safety counter measures are based on the double pipe system and isolating valves at the inlet and outlet ducts, as shown in Fig 5.20 [22].

In case of any leakage of the ducts, this gas mixture is diverted through a concrete channel which is filled with an inert gas, to a sufficiently long distance (more than 100 m), where a relief valve allows the mixture to be released to the surroundings.

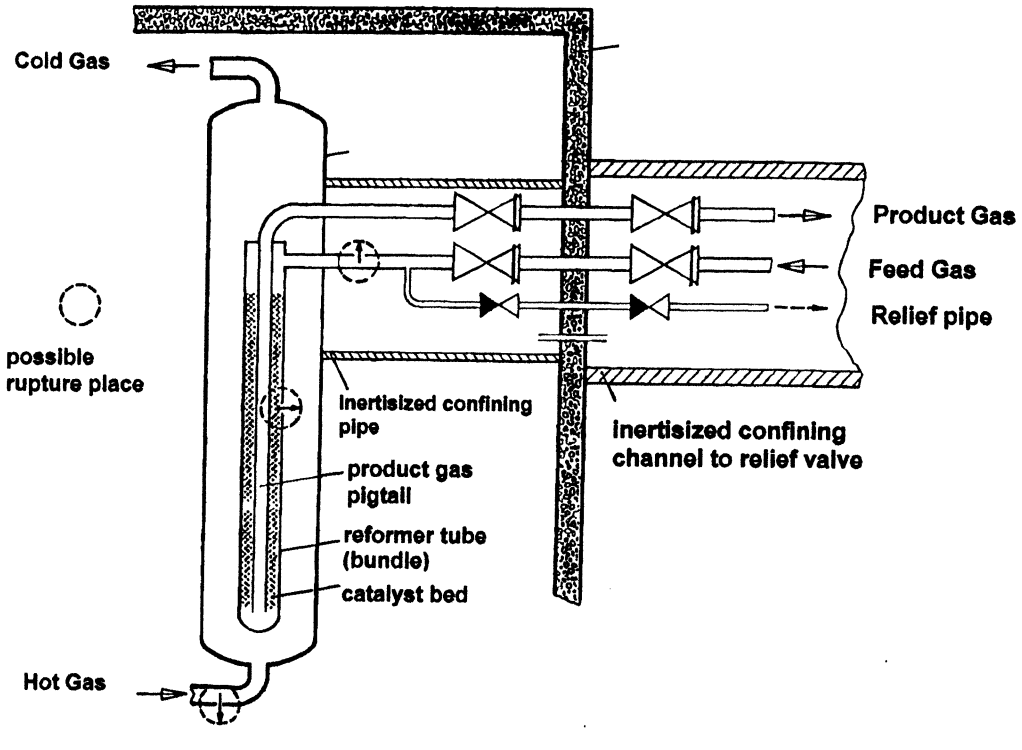


FIG. 5.20. Principals of the safety measures on the process side [22].

On the other hand, the possibility of the release of the burnable gas mixture into the reactor building is given through the leakage of the primary circuit and subsequently the rupture of the reformer tube, although with low provability. To avoid the build up of an explosive gas mixture in the reactor building, quick closing of the isolating valves at the inlet and outlet of the steam reformer limits the leakage of the process gas. However, the reliability of these valves is more important than the quick closing time.

In the case of the use of an intermediate He/he-heat exchanger for a project such as the steam gasification of coal, the combination of the failure of the secondary helium circuit and, subsequently, the rupture of the heat exchanger tube immersed in the fluidized bed of the gas generator will also cause ingress of burnable gases into the reactor building. The probability of such an accident combination, although very low, is possible, so that safety measures are necessary. Also for this design, a double pipe system for the secondary circuit in the reactor building including isolating valves is planned.

Process gas release outside of the reactor building is probable in the case of the arrangement of the gas fabric around its surroundings. Therefore, the possibility also exists for the formation of an explosive gas mixture with air and, subsequently, in the worst case situation, an explosion can occur and its behaviour must be considered in detail.

Without taking into account the arrangement of the gas fabric outside the reactor building, external impacts due to chemical reactions must be considered for all nuclear power plants in Germany in licensing procedures according to the legal Guidelines. Under these aspects, events due to the pressure waves caused by chemical reactions outside the reactor building, because of their low risk value, are not design relevant accidents. However, to minimize the risk, counter measures are necessary.

According to the design concept for the AVR reconstructed plant, the possibility of a process gas release around the reactor building is very low and, therefore, all accidents which are considered for the safety analysis, are only hypothetical.

These accidents are also investigated to determine whether future process heat HTR-plants can accomplish the legal Guidelines for the protection of nuclear plants against pressure waves caused through chemical explosions, and whether risk minimization and, consequently, the protection for the public can be adequately assured.

This guidelines prescribes the time dependent pressure function, whereby a linear increase of the overpressure within 0,1 sec to a value of 0,45 bar on the wall of the reactor building takes place. This overpressure subsequently decreases linearly within the time range of 0,1 to 0,2 sec to the value of 0,3 bar and, thereafter, this overpressure remains unchanged at the value of 0,3 bar in all directions until the end of the first second after the pressure rise. It has to be presumed that pressure waves can come from all directions and, moreover, it cannot be excluded that this pressure wave hits the reactor building many times in succession. Such a pressure wave is generated primarily by fast deflagration of a large gas cloud which can appear after the release of the burnable gas mixture outside the reactor building. Therefore, this guideline requires that the reactor building and the safety related components can withstand such static and dynamic loads without any severe damage.

Furthermore, according to this guideline, a minimum distance between the nuclear power plant and the place at which explosive mixtures are handled is required and should be at least 100 m. This distance depends on the amount of the gas cloud and is given through the following equation:

$$R = 8 \cdot L^{1/3}$$

Where:

R is the safe distance in m
L is the amount of gas cloud in kg .

Therefore, the minimum amount of the explosive gas mixture which has to be considered is about 2000 kg, and is equivalent to the 100 m safety distance. The design of the AVR reconstructed plant includes a relief valve with a reliable ignition source (Fackel) at the distance of about 400 m from the reactor building so that any release of the process gas can be directly burned before the build up of any explosive mixture.

For risk minimization, the following accidents are considered and are given in Figure 5.21. The thickness of the arrows shown in this figure represents the probability of accident occurrence.

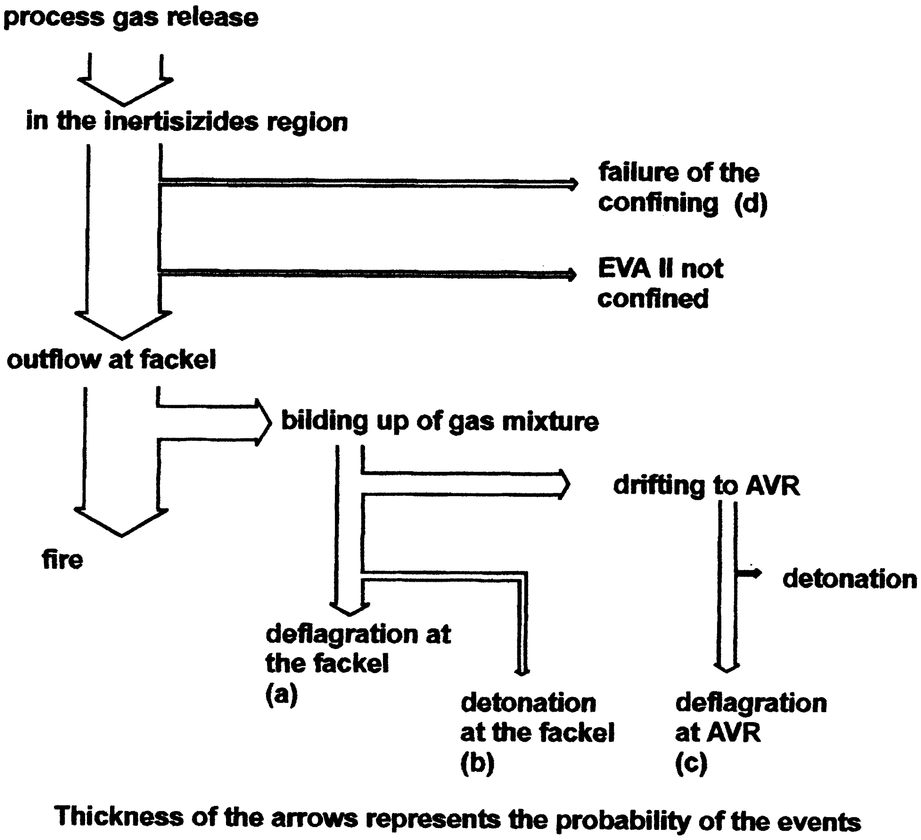


FIG. 5.21. Progress of different events with the release of process gas [22].

a) A process gas release flows in the concrete channel filled with inert gas. Outflow of the gas then occurs through the relief valve thereby building up of an explosive gas mixture with air with a subsequent explosion in the form of deflagration.

The hypothetical development of this incident includes:

b) Detonation of this gas mixture at the relief valve location.
c) Drifting of this gas cloud from the relief valve to the reactor building with subsequent deflagration.

The extremely hypothetical course of this accident includes:

d) After the release of the process gas in the inertisized confining channel, failure of this channel and outflow of the process gas next to the reactor building with it subsequently exploding.

For these accidents, only the amount of the gas released out of the inlet and outlet ducts and from the steam reformer is taken into consideration. This is because the synthesis gas utilising plant has been separated reliably by closing the isolating valves. The total amount of the gas cloud has been estimated for the connecting ducts to the value of 2 x 49 m^3, and in the steam reformer at the value of about 6 m^3.

The volumetric concentration of the dry gas (without taking into account the high steam concentration of about 47%) was estimated under assumption of an average pressure of 15 bar. The concentration includes 71 % H_2; 9 % CO; 9 % CH_4; 11 % CO_2.

The total amount of this process gas is about 60 kmol which is equivalent to about 600 kg. Approximately 57 kmol out of this gas mixture is burnable and the content of hydrogen is about 18 kmol. If this gas can be optimally mixed with air, the stoichiometric mixture of this burnable gas will built to about 12000 m^3 [22].

Three accidents have been further investigated to estimate the destructive load on the reactor building:

Place	Nature of explosion	Wave pressure in free field
Fackel 400 m	Deflagration 160 m/s	0,3 bar
Fackel 400 m	Detonation 2000 m/s	15 bar
Reactor building	Deflagration 100 m/s	0,12 bar

It is very difficult to calculate the static and dynamic load which the reactor building must withstand, because the wave pressure reflection from the obstacle and other details must be considered. It has been estimated that under the third condition, the maximum impulse worked on the reactor building for a relatively long duration, with the progress of the total force at the reactor building is shown in Figure 5.22. This load has been taken into account in the structural and mechanical calculations of the reactor building. The results have shown, that the values of dynamic load at different sites of the reactor building due to this deflagration are lower than those due to the simulated safety earthquake. Therefore, the evaluation of the design based accident for an earthquake cover all events due to gas explosion outside the reactor building. Thus risk minimization can be achieved according to the legal guidelines.

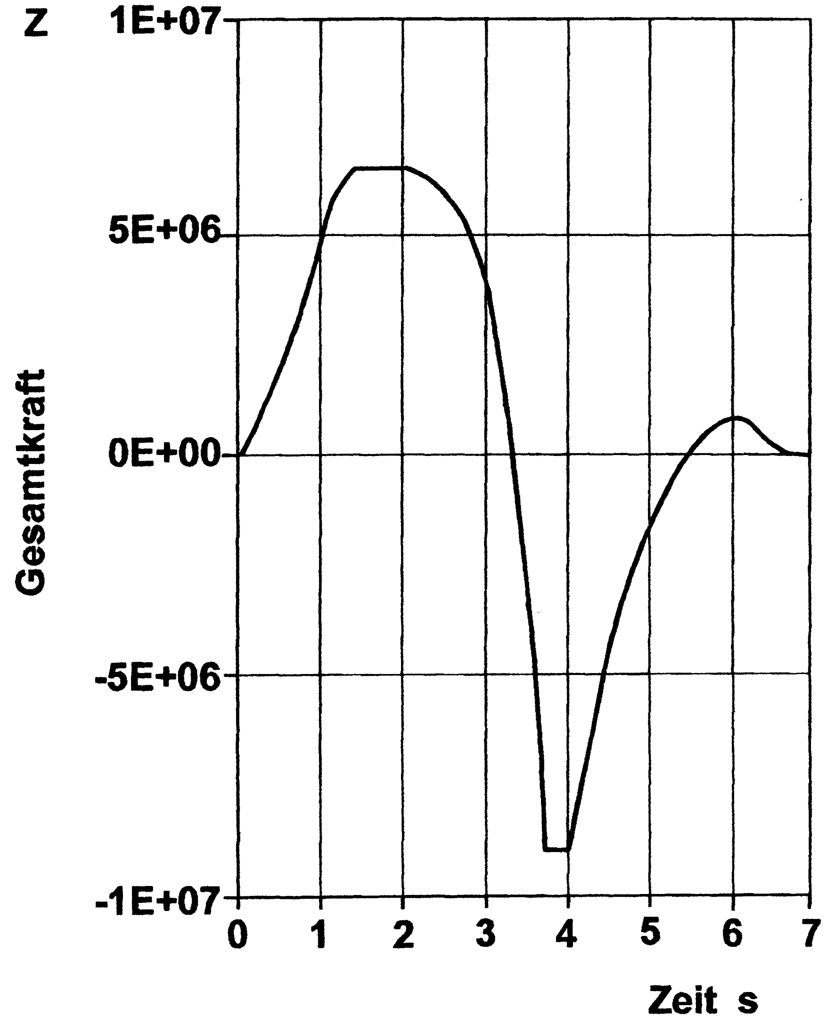


FIG. 5.22. Progress of the total force at the AVR reactor building [22].

5.2.5.3. Review of the general design concept of the steam reformer

Experiences gained through research and development work carried out in Germany for the methane conversion with a steam reformer were generally positive in almost all fields of the design concept. Safety requirements can be fulfilled with the design of the steam reformer and also with its arrangement in the primary helium circuit. However, there are some improvements which can be better realized with an alternative design concept of a steam reformer, e.g. higher performance of the steam reformer, easier removal of the used catalyst, lower stresses of the reformer tubes, etc.

This steam reformer consist of a uniformly filled bed of spherical catalysts in a vertical cylindrical vessel. At the top of the vessel, inlet tubes for the fresh catalysts are arranged, and at the bottom of the vessel a conical discharge tube is provided through which used catalysts can be removed according to the operational conditions as shown in Figure 5.23.

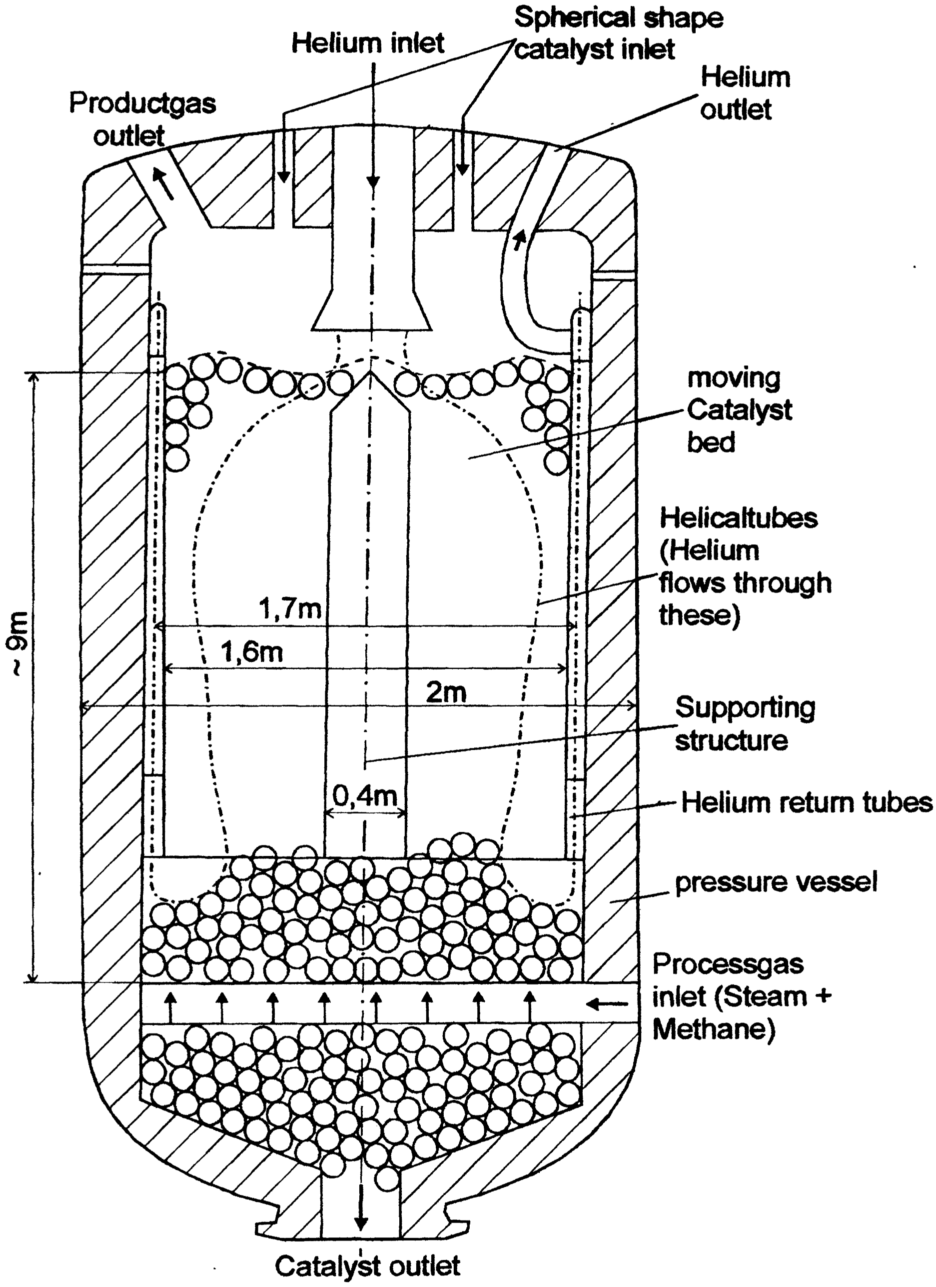


FIG 5.23. Schematic of the design of a steam reformer for testing with the HTTR.

Process gas inlet tubes are arranged in the lower part of the vessel through which methane with steam is uniformly distributed in the catalyst bed and, thereafter, it flows upwards. The process gas is heated to a temperature of more than 800°C by the hot helium in a counter flow to the synthesis gas. This hot product gas flows out of the steam reformer at the top of the vessel and its heat can be used in a separate heat exchanger for preheating the inlet mixture of methane and steam to a temperature of > 500°C before it enters the bottom of the steam reformer vessel.

A cylindrical hot helium header is arranged in the upper part of the steam reformer through which this hot helium is distributed in the many helical tubes. These tubes are arranged in the region of the catalyst bed as a bundle, and the helium flows downward from top to bottom to supply heat uniformly to the process gas for the endothermic steam reforming process of methane. These helical tubes are arranged in such a manner that the movement of the catalyst bed with gravity is possible. Helical tubes are connected to the straight tubes along the side wall of the cylindrical vessel for further flow of helium to the top, where the outlet of cold helium is located. In this way, accessibility to the helium tubes is also provided which forms the barrier between the primary helium and secondary side of the steam reformer. This also allows inspection or, if needed, repair of these tubes to be easily performed. Therefore, the pressure vessel of the steam reformer is not representing the primary barrier. Moreover, this design concept of steam reformer has a similarity with an intermediate He/He-heat exchanger and includes some further advantages as described in the following:

- Higher heat transfer coefficients on both helium and process gas sides are possible without increasing the pressure losses on respective sides.
- The steam reforming temperature at the outlet can be raised with the same hot helium inlet temperature of the steam reformer.
- Pressure loss on the process gas side is relatively low with this design because of the larger cross-sectional area of the catalyst bed. Therefore, the steam to methane ratio can be increased without increasing the number of reformer tubes. With this measurement and (probably) by raising the maximum reforming temperature, the content of methane in the product gas can be considerably reduced.
- Helical tubes for hot helium flow have a smaller diameter as well as reduced thickness. As a consequence, the same heat flux will result in a smaller temperature gradient in the tube wall and, as a result, lower thermal stresses will prevail in the tubes. Also, the transient operational conditions can be handled in a better manner.

This design concept of the steam reformer has not, as yet, been experimentally tested anywhere in the world. Therefore, FZJ has proposed that, if possible, this design be tested in the future with the HTTR plant. For this reason, preliminary investigation has been performed to evaluate a steam reformer design for about 3,7 MW as required in the HTTR-plant. A catalysts bed of about 1,6 m diameter and about 9 m height has been evaluated for this power as shown in Figure 5.23. Thereby, a total of 62 helical tubes of 3 cm outer diameter transfer about 2,5 kg/s hot helium of ~880°C from the top to bottom and provide heat to the process gas in the catalyst bed for methane conversion. These helical tubes are arranged in this catalysts bed at four different diameters. Cold helium of ~ 600°C is transferred to the outlet at the top of the vessel. A support structure for the bundle with an outer diameter of ~ 0,4 m has been arranged in this catalyst bed. The feed gas of ~ 4 kg/s mass flow flows upwards, whereby the mole ratio is ~ $H_2O/CH_4 = 4/1$. Moreover, in this experiment the addition of CO_2 in the feed gas can also be tested in parallel with the same catalyst bed, so that an optimal synthesis gas composition can be realized for its further synthesis to methanol. In this way the production of hydrogen and methanol by steam reforming of methane can be optimally achieved.

5.2.6 Alternative designs — Chinese programme

The 10 MW high temperature reactor test module (HTR-10) is under construction at the Institute of Nuclear Energy Technology, Tsinghua University, Beijing. This test reactor will be utilized to transfer and develop the Modular HTGR technology as well as to establish an experimental base for nuclear process heat application. Figure 5.24 is a schematic representation of the HTR-10 and the process heat application system.

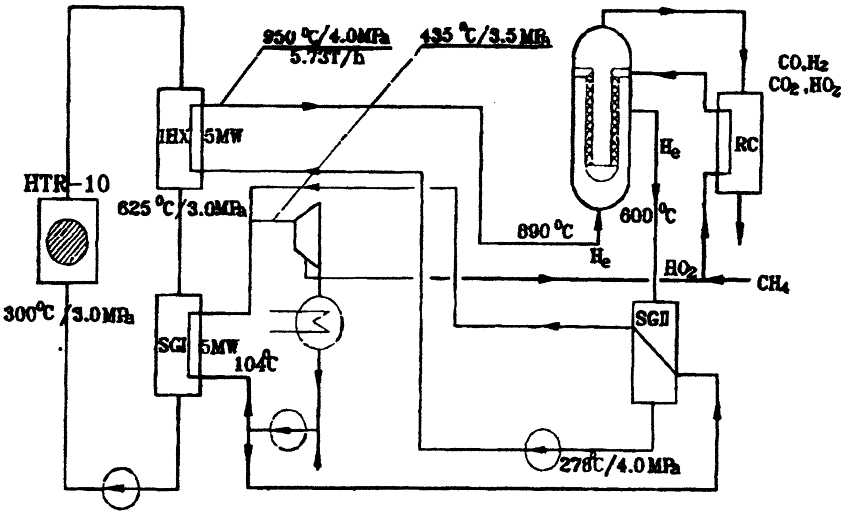


FIG. 5.24. HTR-10 and nuclear process heat application system.

5.2.6.1 HTR-10 reformer optimization

The methane-steam reformer (Figure 5.25) is one of the key facilities for the nuclear process heat application system. Generally speaking, the nuclear heating reformer is a tube-shell type reformer. In comparison with the fossil fuel reformer, its heat transfer efficiency and the reforming transform ratio is low and the cost is high. So in order to make it commercially competitive, it is necessary to optimize the design to increase the productivity and decrease the cost of the reformer.

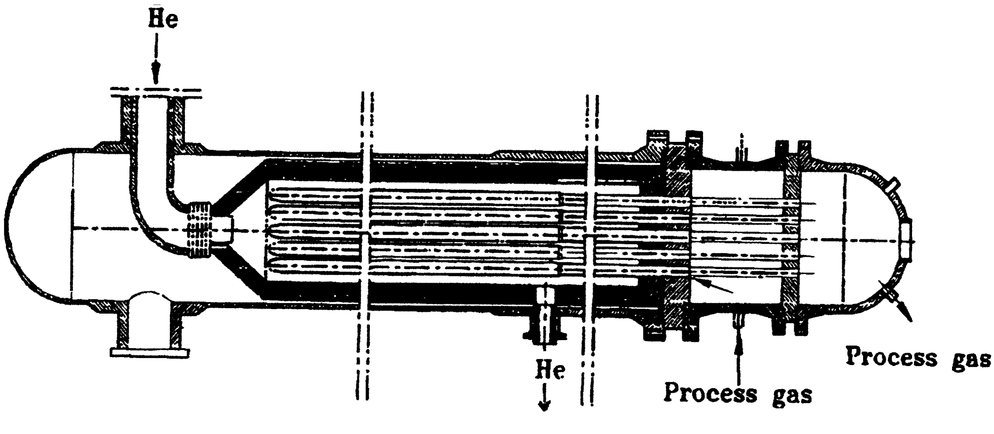


FIG 5.25. Steam reformer structure of the HTR-10.

The following design considerations have been adopted in order to increase the hydrogen productivity:

- to decrease the working pressure of the catalytic bed;
- to decrease the H_2O/CH_4 ratio in the raw gas as low as possible;
- to preheat the process gas by residual heat of the production gas in order to increase the process gas temperature at the inlet of the catalytic bed;
- to intensify the heat transfer at out side of the catalytic bed.
- to increase the temperature difference between the helium (or nitrogen) and the process gas;
- to improve the heat transfer characteristics of the fixed catalytic bed.

5.2.6.2 Reform code

A computer code (REFORM) was developed to optimize the reformer and was used for parameter sensitivity analysis. The basic equations for the REFORM are:

$CH_4 + H_2O \rightarrow CO + 3H_2 - 206$ kJ/mol (1)
$CO + H_2O \rightarrow CO_2 + H_2 + 42$ kJ/mol (2)

The mass conservation equation

$$Fdx = r_1 dw \qquad Fdy = r_2 dw \tag{3}$$

Where: F is feeding rate of the raw gas, kmol/s;
X is methane transformation rate;
y is CO_2 formation rate;
w is weight of the catalyst, kg;
r_1 is reaction rate of the equation (1);
r_2 is reaction rate of the equation (2).

The energy conservation equation:

$$\sum m_i c_{pi} dT = U_1 A_1 (T_{s1} - T)\, dZ + U_2 A_2 (T_{s2} - T)\, dZ - Fdx\Delta H_1 - Fdy\Delta H_2 \tag{4}$$

Where: ΔH_1 and ΔH_2 are the reaction heat of the reactions (1) and (2), respectively;
m_i is the mass of the i component, kmol/s;
C_{Pi} is the specific heat (at constant pressure) of the i component, kJ/kmol.K;
T is the process gas temperature in the catalytic pipes, K;
T_{S1}, T_{S2} is the heating gas temperatures, K;
U is the heat transfer coefficient, kJ/m^2.s.K;
A_1, A_2 is the heat transfer area per length of the tube, m^2/m;
Z is the length of the tube (beginning at the inlet), m.

The momentum conservation equation

$$dp = f'\left(\frac{\rho u^2}{d_s}\right)\left(\frac{1-\varepsilon_B}{\varepsilon_B}\right)dZ \tag{5}$$

Where: ε_B is porosity of the catalytic bed;
ρ is density of the fluid;
u is average empty bed velocity;
d_s is equivalent diameter of the particle.

The following hypotheses are made in the calculation:

- The code REFORM is developed on the basis of quasi-homogeneous phase model, with the solid particles and the fluid considered as a homogeneous phase system.
- Only one dimension (axial) flow is considered. That means the fluid temperature and concentration in a cross section are equal.
- The fluid integral moving model is used in the code.
- The ideal gas equations can be used for the mixed gas.
- The temperature distribution of the heating gas along the tube is linear.

Using the computer code REFORM, the relationship between the methane transformation ratio and the heat transfer, optimized reforming power and effect of reactor outlet temperature on the transformation ratio were analyzed. From analyses, it is suggested that for the HTR-10 the core outlet temperature should be raised up to 950°C and the helium-helium IHX would be utilized. The series combination operating mode can be adopted for the process heat application system. The parameters can be selected as follows: (a.) the reformer power is 2.5MW, (b.) the steam generator power is 5MW and (c.) the reforming temperature is 840°C. The transformation ratio is ~ 0.7.

5.2.6.3 Parameters of HTR-10 Steam Reformer

Design parameters and structure dimensions of the steam reformer for the HTR-10 are listed in Table 5-3 and 5-4, respectively.

Table 5-3: Main Parameters of the Steam Reformer for the HTR-10

Power of the reformer	2.398 MW
Working pressure of the helium	3.0 MPa
Helium flow rate	2.325 kg/s
Outlet/inlet helium temperature	600/890 $^{\circ}$C
Composition of the process gas	
Raw gas	CH_4 / H_2O =1:4
Product gas	CH_4 : 3.11% CO: 6.32% CO_2: 5.74% H_2: 42.89% H_2O: 41.93%
Process gas flow rate	1.57 kg/s
Outlet/inlet process gas temperature at the catalytic bed	840/500 $^{\circ}$C

Table 5-4: Structure Parameters of the Steam Reformer for the HTR-10

Parameters of the tube-bundle	
Number of the reformer tube	37
Diameter of the tube bundle	1400 mm
Diameter of the catalytic tube	ϕ116 X 8 mm
Active section length of the catalytic tube	10140 mm
Diameter of the helium outside tube	ϕ140 X 4 mm
Diameter of the centre guide tube	3 X ϕ18 X 1.5 mm
Parameters of the vessel	
Working pressure	4.4 MPa
Working temperature	350 $^{\circ}$C
Outside diameter	2276 mm
Height	20010 mm
Parameters of the regenerator	
Working pressure	4.4 MPa
Outlet/inlet temperature	
Raw gas	320/500 $^{\circ}$C
Product gas	600/400 $^{\circ}$C
Diameter of the tube	ϕ18 X 1.5
Number of the tubes	3 X 30
Outside diameter of the helical tube	ϕ54 mm
Height	3.72 m

5.2.6.4 New conceptual design of the intermediate heat exchanger for the HTR-10

In order to make a more compact arrangement and increase the power density of the IHX for the HTR-10, use of the multi-concentric helical bundle was taken into consideration. Compared with the modular helical tube-bundle, the structure of the multi-concentric helical bundle has the following distinguishing features:

- The structure is more compact, and the power density and surface compactness is greater than that of modular helical tube-bundle.
- The diameter of the helical tube is large, the hydraulic resistance at the tube side is lower.
- The rigidity of the tube structure is poor.
- The manufacture and installation are complex.

A compact structure can reduce both the diameter of IHX's tube-bundle and the diameter of integrated IHX and SG pressure vessel, therefore it can reduce the cost of the component and the reactor building.

Lower hydraulic resistance not only reduces the power consumption of the compressor, but also makes the pressure difference between the inside and outside of the tubes easy to control. In the case of IHX operation at high temperature, the choice of materials with the necessary properties is limited; and even for the best available materials, the allowable stresses are small. In order to assure safe operation of the IHX, one of the important means is to control the pressure difference between the two sides of the IHX and to reduce the pressure load. The main parameters for the IHX with a multi-concentric helical tube bundle are given in Table 5-5.

Table 5-5: Main parameters of the IHX for HTR-10, (Multi-concentric helical tube bundle)

Thermal power	MW	5
Working medium at primary side		He
Working medium at secondary side		N_2
Inlet/outlet temperature at primary side	°C	900/600
Inlet pressure at primary side	MPa	3.0
Mass flow at primary side	kg/s	3.21
Inlet/outlet temperature at secondary side	°C	483/850
Inlet pressure at secondary side	MPa	3.1
Mass flow at secondary side	kg/s	11.17
Pressure loss at primary side	kPa	15
Pressure loss at secondary side	kPa	70
Diameter of the central pipe	mm	ϕ950×20
Diameter of the heat transfer tube	mm	ϕ22×2
Number of heat transfer tubes		127
Number of radial layers		7
The largest diameter of the helical tube	mm	1344
Heat transfer area	m^2	153
Tube material		Inconel-617
Inside diameter of pressure vessel for integrated IHX and SG	mm	2500

5.2.7. Steam reforming of methane by nuclear heat — Russian activities

Preliminary design work is being performed in Russia on a low powered thermal conversion facility which has the capability to utilize thermal power from the HTGR for methane steam conversion. A pipe methane converter made of reaction assemblies in an ordered catalyst arrangement is being investigated for this application.

The methane converter used to produce hydrogen containing gas utilizes the method of catalytic steam conversion of natural gas. Thermal power from the nuclear reactor is delivered by the circulated helium coolant through a high temperature intermediate heat exchanger (IHX) to the converter. The process steam used for the steam conversion of methane is produced in a steam generator included as part of the helium circuit.

Some of the reactions are expected to take place simultaneously in the process of methane steam conversion, the main reaction is:

$CH_4 + H_2O \Leftrightarrow CO + 3H_2$ - 60 Kcal/mole

The process is significantly endothermic, therefore significant heat has to be supplied. The reaction proceeds at an appreciable rate with a temperature of >1000°C. The process is made active in the presence of a catalyst, with the most active catalysts being nickel, cobalt, and noble metals.

In almost all recent industrial systems for natural gas conversion, a catalyst containing nickel oxide was used as the active component and is reduced to metal in the system. A nickel surface area of approximately $0.5 m^2/g$ makes up the operating catalyst with the nickel crystals being about 1 μ in size. The crystals can grow in the presence of steam at t >1023°C which results in a reduction in their specific area. To prevent this, a small amount of special material such as Al_2O_3, Cr_2O_3, MgO and others are added to the catalysts to make the area constant.

Besides the stabilizing additives, the catalyst contains small amounts of calcium, titanium and other oxides, which serve to improve its thermal stability and mechanical strength.

The design of the methane converter for a pilot-industrial facility is shown in Figure 5.26. It is designed as a cylindrical heat exchanger with six-facet reaction assemblies. The assemblies contain a heat-exchanging surface of the methane converter consisting of bunched Field's tubes of ∅ 38 × 4 arranged as a hexagonal lattice. The Field's tubes layout is made with wire coiling.

A nickel catalyst consisting of two separate sizes is fixed in an annular gap of the Field's tubes and is easily replaced upon completion of its service time.

Each reaction assembly (Figure 5.27) has a built-in heat exchanger in the top part that is constructed as a three-row multi-stepped coil of tubes of ∅ 14 × 2. The heat exchanger is 1500 mm in height. During operation, the gas/steam mixture enters the converter with the converted gas exiting through ducts in the top.

Helium of 900°C in temperature is delivered from the IHX to the converter and after passing through the inter-tube space of reaction assemblies is delivered to the steam generator at the temperature of 700°C (Table 5-6).

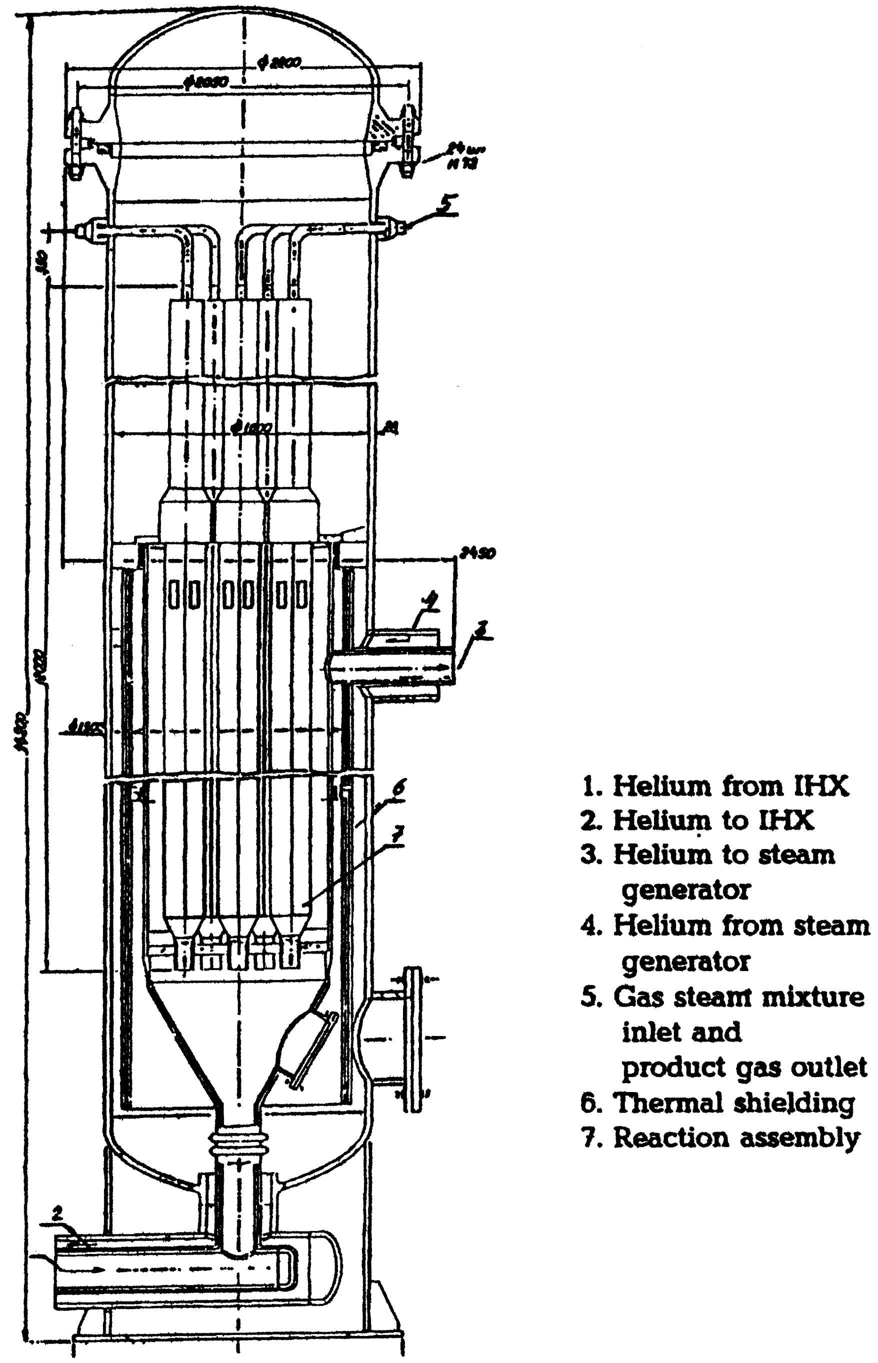
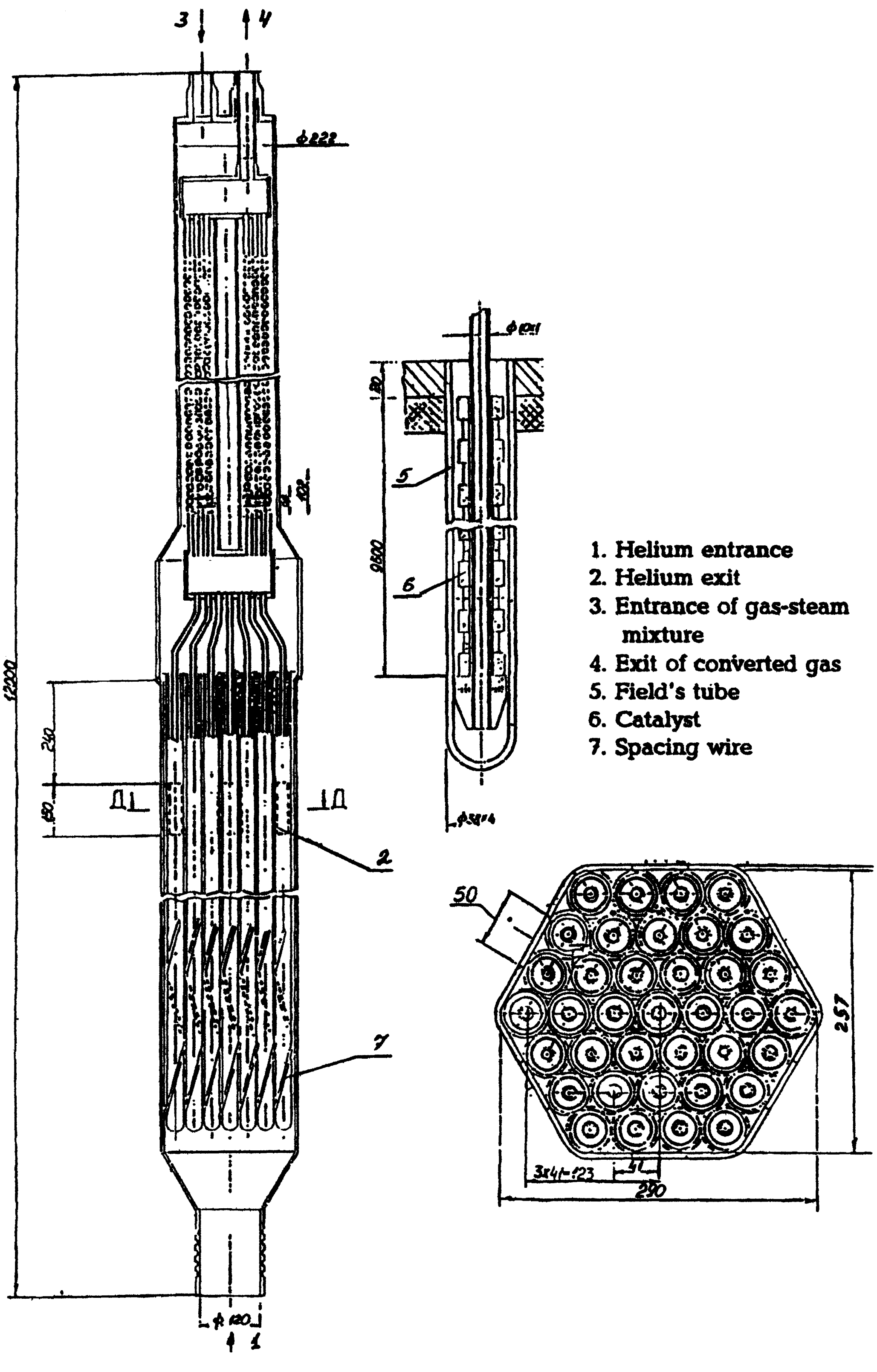


FIG. 5.26. Methane converter.

5.2.7.1 Technical and operational requirements for steam reforming

The methane converter design should provide the following:

- Dependable operation throughout its lifetime, including transient and start-up modes and accidents.
- Capability for being easily decontaminated.
- Operation with uniform distribution of helium and process gas along the pipes.
- Ability for loading/unloading of the catalyst.



1. Helium entrance
2. Helium exit
3. Entrance of gas-steam mixture
4. Exit of converted gas
5. Field's tube
6. Catalyst
7. Spacing wire

FIG. 5.27. Reaction assembly.

- Be leak resistance at the design parameter of helium leakage less than 0.3 volumetric %.
- Exhibit vibration stability.
- Have a catalyst heating/cooling rate of no more than 50 K/hour.
- Ability for stable operation at 50 to105% of rated load.
- Ability for easy drainage of the system.
- Ability for inspection, maintenance and replacement of components and units.

Table 5-6: Technical characteristics of the methane converter.

Parameter	In the facility	In the tubes
Thermal load, MW	5.1	
Temperature, °C inlet/outlet	900/700	350/500
Pressure, MPa inlet/outlet	5.4/5.35	3.75/3.45
Environment, %vol.	Helium	Gas-steam mixture inlet/outlet CH_4 — 21.2/5.77 CO_2 — 0.13/5.83 H_2 — 2.81/41.0 CO — -/5.65 H_2O — 73.44/40.9
Flow rate, kg/sec	5.0	2.5
Assemblies in the unit	7	
Field’s tubes number in an assembly	37	
Tubes diameter, mm	38 × 4	
Tubes material	10 × 23H18	
Tubes number in the built-in heat exchanger	18	
Tubes diameter	14 × 2	
Tubes material	12×18H10T	
Converter height, m	16.2	
Pressure vessel diameter, m	1.656	
Unit mass, t	39.5	

5.3 CO_2 REFORMING OF METHANE FOR PRODUCTION OF HYDROCARBON AND METHANOL

5.3.1 The process

5.3.1.1 Introduction

The reforming of CH_4 with CO_2 to produce synthesis gas with a H_2/CO ratio of approximately one has had, until recently, no significant commercial application. In recent years, considerable attention has been paid to global warming due to the greenhouse effect. This reaction has important implications, since both methane and carbon dioxide contribute to the greenhouse effect.

The major source of synthesis gas, which is mainly used as a feedstock for hydrogen, ammonia and methanol synthesis and Fischer-Tropsch conversions, is from the steam reforming reaction:

$$CH_4 + H_2O \rightarrow CO + 3H_2 \qquad \Delta H_{298} = 206 kJ/mole \qquad (1)$$

A proposed alternative to the steam reforming process is reforming of CH_4 with CO_2 [23]. This reaction was first proposed by Fischer and Tropsch in 1928 [24]:

$$CH_2 + CO_2 \rightarrow CO + H_2 \qquad \Delta H_{298} = 247 kJ/mole \qquad (2)$$

Because of the high endothermicity of the reaction and other advantages, it was proposed for the conversion, transportation and storage of solar energy in chemical form [25].

Similar to the concept developed for transportation of high temperature nuclear heat over long distances in a closed loop by the Kernforschungszentrum Jülich GmbH (KFA), Jülich, Germany [26,27], using the steam reforming process, the CO_2 reforming process was developed by the Weizmann Institute of Science (WIS), Rehovot, Israel, for the transportation and storage of solar energy [28-30].

In addition, CO_2 is a by-product of many industrial processes and available for utilization. For instance, power plants emit a large amount of CO_2 mixed with nitrogen. It contributes to around 25% of the global CO_2 emission. Landfill gas usually consists of 50% CH_4 and 50% CO_2. There are natural gas fields that contain a large CO_2 content, such as the Natuna gas field in Indonesia [31].

In the past, several attempts have been made to reform CO_2-rich hydrocarbon feedstock for the production of synthesis gas on a commercial scale, i.e., the Calcor process [32,33]. But until recently there was no established industrial process for CO_2 reforming of CH_4, primarily due to the problem of coking on the existing commercial Ni-based catalysts. The renewed interest in this process, mainly in connection with transportation of solar energy in a closed reforming methanation loop as an alternative for the steam reforming cycle, initiated several studies on the reactor parameters and catalysts.

McCrary, *et al.* [34] have reported a successful closed loop Solchem system, including the CO_2/CH_4 reforming methanation cycle. Nickel catalyst deterioration has been solved by the use of Rh on stainless steel catalyst. Using CO_2 reforming of methane as the process for storage and transport of solar energy, a solar chemical heat pipe (SCHP) was tested at WIS [35]. High conversion was achieved for both endothermic reforming reaction and the exothermic methanation reaction. The whole process was carried out in a closed loop and the performance was satisfactory. Based on the results from a 10-kW_{th} scale (energy into the process) solar reformer, a 500-kW_{th} unit was built and tested successfully at WIS [30].

In a catalytically enhanced solar absorption receiver (CAESAR) project, carbon dioxide reforming of methane in a solar volumetric receiver/reactor was successfully demonstrated by the Deutsches-Zentrum für Luft-und Raumfahrt e.V. (DLR), Germany [36]. The total solar power absorbed reached 97 kW_{th} with 70% methane conversion. A scale-up of this solar reactor was tested at WIS with a peak power of 286 kW, 3.5 bar (abs) pressure and 84% methane conversion [37].

5.3.1.2 Thermodynamics of CO_2 reforming of methane

The reforming of CH_4 with CO_2 involves the highly endothermic reaction (2). This reaction is always accompanied by the reverse water gas shift reaction (RWGS):

$$CO_2 + H_2 \rightarrow CO + H_2O\,(g) \qquad \Delta H = 41\,kJ/mole \qquad (3)$$

and the carbon gasification reaction:

$$C + H_2O\,(g) \rightarrow CO + H_2O \qquad \Delta H = 131kJ/mole \qquad (4)$$

As with the steam reforming of CH_4, the CO_2/CH_4 reforming is likely to be conducted under conditions leading to carbon formation. The carbon deposition results from methane cracking, from the Boudouard reaction and from the reduction of CO.

The methane cracking reaction is as follows:

$$CH_4 \rightarrow C + 2H_2 \qquad \Delta H = 75\,kJ/mole \qquad (5)$$

The Boudouard reaction is as follows:

$$2CO \Leftrightarrow C + CO_2 \qquad \Delta H = -171kJ/mole \qquad (6)$$

The reduction of CO is:

$$CO + H_2 \Leftrightarrow C + H_2O \qquad (7)$$

Thus, the CH_4/CO_2 reforming reaction (2) can be seen as consisting of reaction (5) and the reverse of reaction (6). Ideally, the carbon formed in reaction (5) should be rapidly consumed by the reverse of reaction (6) and, to a lesser extent, by the steam/carbon gasification reaction (4). Reaction (4) can play a role in CO_2/CH_4 reforming because steam is almost always formed via the RWGS reaction (3).

If reaction (5) is faster than the carbon removal rate, there will be a net build-up of solid carbon, which creates serious problems in practice, because it would lead finally to catalyst deactivation and disintegration and reactor blockage. Apart from reaction (5), reaction (6) and the reverse of reaction (4), both of which are favored at low temperatures, are also potential sources of carbon. Reaction (3), which consumes product hydrogen, also represents a disadvantage unless both reactions, (3) and (4), are taking place efficiently and consecutively so that the overall stoichiometry of the CO_2/CH_4 reforming reaction is maintained. Luckily, chemical equilibrium is not equally established among all the reactions in the presence of a single catalyst.

For example, copper catalysts [38] selectively establish a near equilibrium state for the WGS reaction:

$$CO + H_2O\,(g) \Leftrightarrow CO_2 + H_2 \qquad \Delta H = 41\,kJ/mole \qquad (8)$$

But these catalysts leave other reactions far from equilibrium. In the H_2O/CH_4 system, the nickel catalysts bring the reforming methanation reactions close to equilibrium, but at low temperatures they do not allow the accompanying carbon deposition reaction (5) to occur. Thus, the role of suitable catalysts in the CO_2/CH_4 system would not only be to speed up the overall reaction but also to adjust the appropriate elementary steps in a way that prevents both net carbon deposition and water formation.

Figure 5.28 shows the equilibrium composition calculated for the reforming reaction [25] at the temperature range 400–1000°C, using a CO_2/CH_4 feed ratio of 1/1 at pressures of 1 and 10 atmospheres. This calculation is based on the assumption that the reacting mixture is equilibrated with respect to reaction (5), (6) and (3). It can be seen that some water is always formed, although at 1 atm it effectively disappears above approximately 900°C. At 900°C, the CH_4 conversions are 97 and 90% at 1 and 10 atm, respectively.

Figure 5.29 shows the carbon deposition threshold curves [25], derived by assuming equilibrium of the Boudouard reaction (6) under two different pressures of 1 and 10 atm. Carbon deposition regimes lie on the left of each curve. Clearly, carbon deposition is thermodynamically possible for a CO_2/CH_4 reforming feed ratio of 1/1 at temperatures up to 1000°C at 1 atm and 1100°C at 10 atm. Working at the temperature range of 800–900°C will require a CO_2/CH_4 ratio of 3–2, respectively, to be in the carbon-free zone. However, the carbon-free operation with CO_2/CH_4 feed alone should be possible if the reaction is controlled kinetically by using suitable catalysts, as described below.

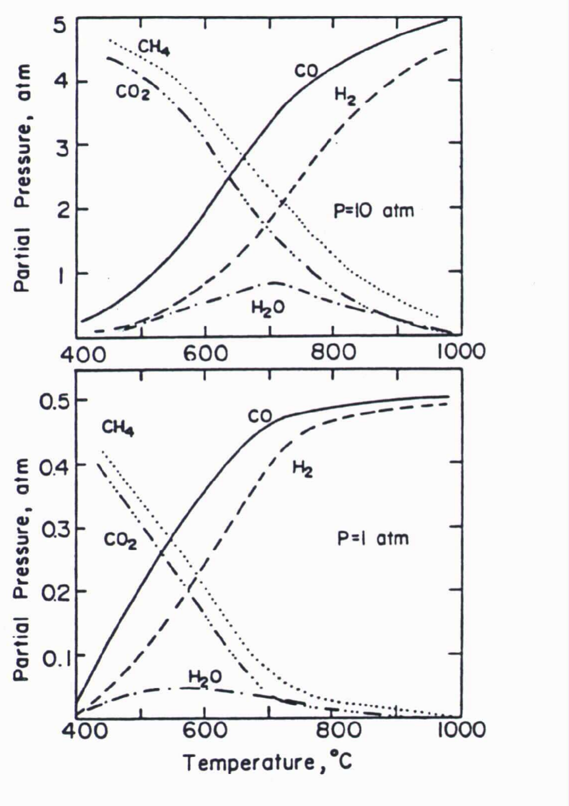


FIG. 5.28. Composition-temperature diagrams for the CO_2/CH_4 (1:1) system (1 atm = 0.101 MPa).

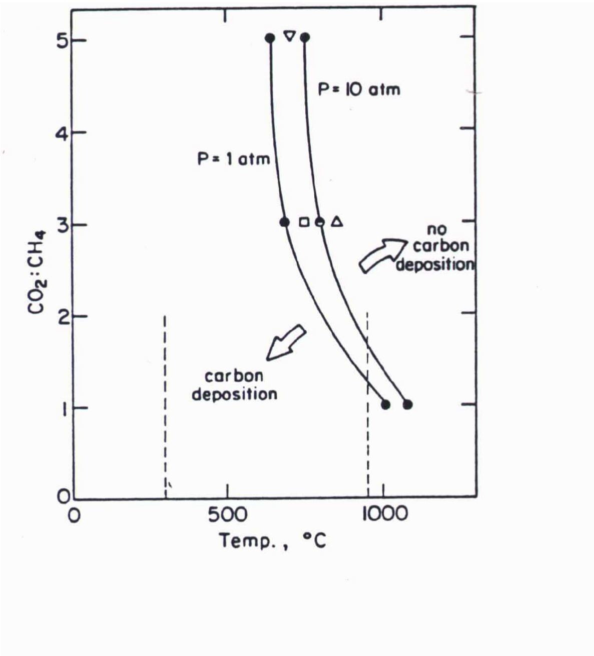


FIG. 5.29. Carbon deposition threshold for the CO_2/CH_4 system.

5.3.2 Catalysts

The CO_2 reforming of methane on regular nickel catalysts will result in the formation of carbon whiskers [39] via a mechanism, involving dissolution of adsorbed carbon atoms in the nickel crystal and nucleation of the whisker from Ni-surfaces. The rate of carbon formation was found to be far less on noble metals, which was ascribed to a smaller dissolution of carbon into these metals. Rhodium and ruthenium were found to be far more active than nickel [40].

Table 5-7 [23] lists some results of CH_4/CO reaction over group VIII metals-supported catalysts. Most of the group VIII metals (Rh, Ru, Ni, Pt, Pd, Ir, Co, Fe), except Os, are more or less effective for catalysis of the CH_4/CO_2 reaction. Using catalysts based on platinum group metals, Ashcroft, *et al.* [41] examined the stoichiometric CO_2-reforming reaction at 1050° K and the atmospheric pressure and achieved 90% yield of synthesis gas, without carbon deposition on Ru, Rh and Ir. Catalysts based on Ni, Ru, Rh, Pd, Ir and Pt were also compared for CO_2 reforming of methane. Ru and Rh showed high selectivity for carbon-free operation. The sequence of activity for the CO_2/CH_4 reaction or carbon-free formation was Ru,Rh>Ir>Ni,Pt,Pd [39]. For Rh and Ru, the conversion and deactivation tests showed that Rh was more stable [42].

Table 5-7: Summary of catalytic reforming of CO_2/CH_4

Catalyst	CO_2/CH_4	Conversion (%)	Temperature (K)
Ni/Al_2O_3	1:1	36.3	873
Ni/SiO_2	1:1	14.9	873
Rh/TiO_2	1:1	88.2	893
Rh/Al_2O_3	1:1	85.1	893
Rh/Al_2O_3	1:1	85	1073
Ru/Al_2O_3	1:1	83	1073
Ru/MgO	1:1	90	963
Rh/MgO	1:1	88	963
$Ni/CaO\text{-}Al_2O_3$	2:1	88	1211

Table 5-8 [23] illustrates the activity order for metals dispersed on various supports. It is seen that the combination of metal and support affects the resultant metal activity. Even for the same support, there are conflicting conclusions due to different operation conditions.

Rh is a superior element with respect to the high activity and the coking-free operation, if supported on Al_2O_3. Ru is better than Rh if MgO and SiO_2 are used as supports. Loadings of metals on supports also affect the activity of the catalysts. Low loadings (about 0.5−5 wt %) of the noble metals are usually sufficient because of their effective performances. Higher loadings are required for Ni and Co catalysts in which the metal-support interaction is stronger than in the case of heavier metals.

Table 5-8: Catalytic activities of metals on various supports

Group No.	**Metal activity**	Metal loading (wt %)	Temperature (K)
1	Al_2O_3		
	Rh>Pd>Ru>Pt>Ir	1	823
	Rh>Pd>Pt>Ru	0.5−1	823−973
	Ir>Rh>Pd>Ru	1	1050
	Ni>Co>Fe	9	773−973
	Ni>Co>Fe	10	1023
	Ru>Rh	0.5	873
	Ru>Ru	0.5	923−1073
2	SiO_2		
	Ru>Rh>Ni>Pt>Pd	1	973
	Ni>Ru>Rh>Pt>Pd>>Co	0.5	893
3	MgO		
	Rh>Ru>Ir>Pt>Pd	0.5	1073
	Ru>Rh>Ni>Pd>Pt	1	973
	Ru>Rh≈Ni>Ir>Pt>Pd	1	823
	Ru>Rh>Pt>Pd	1	913
4	Eu_2O_3		
	Ru>Ir	1−5	873−973
5	NaY		
	Ni>Pd>Pt	2	873

Perera, *et al.* [43] studied the effect of noble metal loadings. It was shown that in the range of 1−5 wt % loading and a temperature of 700°C the conversion of CH_4 in the CH_4/CO_2 system was increased from 70 to 80%.

Berman, *et al.* [44] studied the activity of the Ru catalyst on alumina for both reforming of CO_2/CH_4 and methanation of CO-rich hydrogen. Long-term testing of 1% Ru/Al_2O_3 showed that this catalyst is most suitable in the temperature range of 600−900°C and CO_2/CH_4 molar ratios of 1.3−5.0. The kinetics of CO_2/CH_4 reforming with Ru on alumina stars was investigated at temperatures of 600−800°C and pressures of 2−6 atm. It was found that the conversion rate of CH_4 to H_2 increases with the partial pressure of CH_4 (P_{CH_4}) and decreases with the increase in the partial pressure of CO_2 (P_{CO_2}). The activation energy was found to be 16±0.6 kcal/mole.

The rate of the RWGS reaction, which produces CO and H_2O, increases slowly with the increase of P_{CH_4} and considerably with the increase of P_{CO_2}. The H_2/H_2O ratio decreases sharply when the P_{CO_2} is increased, whereas the total CH_4 conversion exhibits almost no change.

Following Edroheyi, *et al.* [45], the kinetics scheme of CO_2/CH_4 reforming with the 1% Ru/Al_2O_3 was investigated by Berman *et al.* [44] for temperatures at the range of 600–900°C, pressure of 1–5 atm and CO_2/CH_4 ratio of 1–1.3.

The following reaction mechanism was assumed:

$$(CH_4) + 2(*) = (CH_3) + (H) \tag{9}$$

$$(CH_3) = (CH_2) + (H) \tag{10}$$

$$(CH_2) = (CH) + (H) \tag{11}$$

$$(CH) = (C) + (H) \tag{12}$$

$$(CO_2) + (H) = (CO) + (OH) \tag{13}$$

$$(C) + (OH) = (CO) + (H) \tag{14}$$

$$(H) + (OH) = (H_2O) \tag{15}$$

$$(H) + (H) = (H_2) \tag{16}$$

$$(CO) = (CO) + (*) \tag{17}$$

where (*) is the active center on the Ru surface.

The following kinetic equations were derived from the experimental data [46]. The rate of hydrogen formation in liters per hour per gram catalyst is:

$$W_{H_2} = \frac{k_H P_{CH_4}^2}{\left(b_0 P_{CH_4} + b_1 P_{CO_2} + b_2 \dfrac{P_{CH_4}}{P_{CO_2}} + b_3 P_{CO_2}^2 \right)^2} \tag{18}$$

The constants k_H, b_0, b_1, b_2 and b_3 were determined from the experimental data. The dependence of these constants on temperature is as follows:

k_H = $5 \cdot 22 \cdot 10^3 \cdot \exp(-E/RT)$ liter/h · gram catalyst · atm^{-2}
E = 9027 cal/mole for 600<T<700°C
E = 9027 – (T-273) · 22.6 cal/mole for 700<T<750°
b_0 = 1 atm^{-1}
b_1 = $3.686 \cdot 10^6 \cdot \exp(-32900/RT)$ atm^{-1}
b_2 = $5.43 \cdot 10^{-3} \cdot \exp(7570/RT)$
b_3 = $6.59 \cdot 10^{-6} \cdot \exp(17668/RT)$ atm^{-2}
R = 1.98 cal/mole · °K

The following kinetic equation was found for the RWGS reaction:

$$W_{H_2O} = \frac{k_W P_{CH_4} \cdot P_{CO_2}^2}{\left(c_0 P_{CH_4} + c_1 \cdot P_{CO_2}\right)^2} \tag{19}$$

where

k_W	=	$2900 \cdot \exp(9200/RT)$	liter/h · gram catalyst · atm^{-3}
c_1	=	$0.06 \cdot \exp(5400/RT)$	atm^{-1}
c_0	=	1 atm^{-1}	

Berman, *et al.* [44], based on experimental data, also reported the activation energies (E), calculated by Arrhenius equation, as follows:

For rhodium/alumina catalysts:

0.05%	Rh/Al_2O_3	E =	19±2	kcal/mole
0.1%	Rh/Al_2O_3		19±1	kcal/mole
0.5%	Rh/Al_2O_3		15±1	kcal/mole
0.05%	Rh+0.5% P_t/Al_2O_3		17±1	kcal/mole
0.1%	Rh+0.5% P_t/Al_2O_3		13±1	kcal/mole

It can be seen that the activation energies for the bimetallic catalysts are lower than those of monometallic catalysts.

For ruthenium/alumina catalysts:

0.5%	Ru/Al_2O_3	E =	20±1.5	kcal/mole
1%	Ru/Al_2O_3		16±1	kcal/mole
2%	Ru/Al_2O_3		16±1	kcal/mole

5.3.2.1 Effect of support on catalyst activity and carbon formation

The nature of the oxide support greatly affects the catalyst activity due to the varying active surface area and acid-base properties. Carbon dioxide reforming involves the adsorption and dissociation of CO_2 on catalysts. Since CO_2 is well known as an acid gas, adsorption and dissociation of CO_2 may be improved with a basic catalyst.

Nakamura, *et al.* [47] found that the effect of support was in the following order: Al_2O_3>TiO_2>SiO_2 for the CO_2/CH_4 conversion with ruthenium catalyst at 0.5% loading. For rhodium at 0.5% loading, Tsipouriari, *et al.* [48] found the following order: YSZ>Al_2O_3>TiO_2>SiO_2>>MgO. The addition of alkali promoters to catalysts was effective for preventing coke formation from methane during steam reforming. Similarly, for the reaction of CO_2/CH_4, the addition of basic promoters, such as alkali or alkali-earth oxides, changes the nature of the support, because CO_2 is adsorbed strongly on the surface of basic catalysts and reduces the CO_2 partial pressure near the surface, which will prevent carbon deposition on the catalysts. Yamazaki, *et al.* [49] developed a catalyst of Ni/MgO-CaO showing high basicity and lower coke-forming ability, which attributed to the addition of CaO.

Carbon deposited on the catalysts comes from two routes. Experiments showed that the deposited carbon is primarily derived from the CO_2 molecular route and, to a lessor extent, from the CH_4 molecular route [48]. The carbon species originally produced are believed to be atomic carbon. They are very reactive and an important intermediate in the CO_2+CH_4 reaction. In fact, it has been reported that CO_2 dissociated into carbon and two oxygen atoms on metal, and that it was possible that carbon and oxygen on the surface recombined to form CO. There are studies reporting that coke gasification by H_2 steam or CO_2 is promoted by nickel. Therefore, it is expected that water produced in the system may react with reactive surface carbon to give H_2, CO and CO_2 as follows:

$$C + H_2O \rightarrow CO + H_2 \tag{20}$$

$$C + 2H_2O \rightarrow CO_2 + 2H_2 \tag{21}$$

$$C + CO_2 \rightarrow 2CO \tag{22}$$

Zhang and Verykios [50] found that carbon originating from the CO_2/CH_4 mixture on Ni/La_2O_3 catalyst may be active, promoting the reaction via the participation of itself or by its interaction with a component migrated from the La_2O_3 carrier. Mark and Maier [51] studied the behavior of the surface carbon formed in CO_2/CH_4 reaction on Rh/Al_2O_3 catalyst and found that 90% of the surface carbon arising from the decomposition of methane on Rh catalyst is highly reactive and reacts extremely fast with CO_2 to give CO. They proposed that the decisive factor for carbon-free CO_2 reforming is a rapid reaction of the active carbon with CO_2, which avoids a high surface coverage of carbon. Such condition can only be achieved when both reactants are added simultaneously.

The stability of Ru/Al_2O_3 catalyst at high temperatures was investigated [52]. Results show that the activity of 2% Ru/Al_2O_3 catalysts promoted by 2–20 wt % Ce_2O_3 was significantly increased with a maximum at about 10% Ce content. Both γ-Al_2O_3 and α-Al_2O_3 were used as substrates. This activity was measured at high temperatures in the range of 800–1100°C. The results show that at 4 atm and CO_2/CH_4 ratio of 1.3, 1–2% Ru/Al_2O_3 catalysts were sharply deactivated at 1000–1100°C. However, when cerium (5 wt %) or lanthanum (19 wt %) were added, the activity increases after reaction at 1100°C for 8 hours compared to the fresh catalyst, followed by a slow deactivation process.

5.3.2.2 Catalyst poisons

Most of the catalyst development work for CO_2/CH_4 reforming was performed in connection with the closed loop concept for transportation and storage of solar energy. Working in a closed loop avoids the issue of catalyst poison. However, when this reaction is used for the production of syngas as a raw material for methanol or hydrogen or for direct burning as a fuel, the catalyst is exposed to various poisons contained in the feed. Catalytic activity is affected seriously, even by very low concentrations of catalyst poisons in the gases to be reformed. Such catalyst poisons are sulfur, arsenic, copper, vanadium, lead and chlorine, or halogens in general. Sulfur, in particular, lowers catalyst activity considerably and can be found in practically all gaseous feedstocks [53].

Zinc oxide desulfurization systems operating at 350–400°C are generally used today. They are reliable in absorbing hydrogen sulfide and, with limitations, sulfur compounds, such as carbonyl sulfide and mercaptans. To achieve optimal desulfurization conditions, certain space velocities and effective flow rates must be ensured. Sulfur pickup ranges from 15 to more than 30%, which is almost the complete conversion of zinc oxide to zinc sulfide, are possible. Cyclic organic sulfur compounds, such as thiophenes, normally require hydrogenation over cobalt-molybdenum or nickel-molybdenum catalysts. These catalysts are often arranged in separate vessels. Hydrogen or hydrogen-containing gas are added to the process feedstock at temperatures of 150–350°C. In some cases, when nickel is not used, part of the product syngas stream can be recycled and used for the hydrogenation. The cyclic sulfur compounds are converted to hydrogen sulfide, which then can be absorbed by the zinc oxide bed.

Nickel-based catalysts are also available today for low temperature desulfurization. Operation temperatures are 100–150°C. This catalyst can operate without the addition of hydrogen at relatively high concentration of sulfur compounds. The hydrogenation reactor can be used simultaneously to hydrogenate unsaturated hydrocarbons, especially olefins, in the row gas. Today, new catalysts, particularly suitable for natural gases, combine both the hydrogenation and the H_2S absorption in a single reactor.

5.3.3 Type of reformers and experience gained in solar reformers

In industry, the steam reformers are mostly of tubular type with top and wall-fired box-type units. In wall-fired reformers, rows of reformer tubes are heated mainly by the radiant sidewall. Top-fired reformers provide most of the heat through radiation of the burner flame and the hot flue gases. Therefore, top-fired reformers may have several parallel rows of tubes, whereas wall-fired reformers can have only one row. The reformer box consists of several layers of refractory insulation and insulating mats with exterior metal skin. The hot face refractory must be resistant to temperatures of over 1200°C. Burner geometry, flame length and diameter, tube-to-tube and row-to-row spacing, fire tube length, and distance from the flame to the reformer wall, determine the homogeneity of heat transfer to the tubes. Normally, the industrial tubes have inside diameters of 75-125 mm, wall thickness of 10-20 mm, and fire lengths of 9-15 m, depending on the reformer type.

In principle, the same type of reformers and tubes could be used for the CO_2 reforming. However, if the heat source is not a flame but a HTGR. The heat transfer mechanism is primarily convective between helium and the process gases, instead of a radiative mechanism. These types of reformers with helium flows along the reformer tube through an annular gap between a jacket tube and the catalyst tube, or in a combination of axial and cross flow through an arrangement of baffles and concentric tube bundles, which was developed at KFA-Jülich, within the framework of the Prototype Plant Nuclear Process Heat (PNP) project [54]. These reformers were developed for steam reforming of methane but, in principle, they could be used for the CO_2 reforming process as well. If the heat source is concentrated solar energy, the heat is transferred via direct or indirect radiation. So far, most of the research in solar reforming has focused on the concept of a closed loop for storage and transport of solar energy. The CO_2/CH_4 reforming has thermodynamic and practical advantages over the steam reforming for use with solar energy as an intermittent source of heat.

The two main solar reforming concepts are tubular reformer tubes filled with the catalyst, directly incorporated into the solar furnace where they are heated by concentrated solar radiation, and windowed or are volumetric reformers where the catalyst is heated directly by the solar beam, allowing very high volumetric reaction rates. As a result, the windowed reformer is compact and potentially less expensive. This technology requires good matching of flow and reaction rates with the solar flux, also requiring the development of a reliable window (usually made of quartz).

Several experiments are reported in connection with solar-driven CO_2/CH_4 reforming reactors. A 100-kW-windowed reformer, named Catalytically Enhanced Solar Absorption Receiver (CAESAR), which contains porous ceramic foam (mostly α-Al_2O_3) loaded with approximately 0.2 wt % Rh catalyst was tested successfully [36].

WIS operated a pentagon-shaped cavity receiver containing eight vertical reformer tubes (2-inch diameter, schedule 80), 4.5-m long, made of Inconel 617 and filled with 1 wt % Ru on alumina catalyst. It was capable of providing 480 kW of solar heat into the reaction [46]. The concentrated solar radiation entered through an aperture, 60-cm diameter, in the front side of the cavity enclosure. There were two banks of tubes connected in parallel, one on each side of the aperture (Figure 5.30) [55]. The tubes were directly heated by the solar radiation reflected from the walls of the enclosure. The tubes are placed and spaced in such a way that the illumination will be uniform. The CO_2/CH_4 feed ratio was 1.2 entered at about 500°C, 16–18 atm and exit temperature range of 800–830°C. The results show 77–82% CH_4 conversion. The activity of the catalyst was maintained stable for two years of operation. No carbon formation was detected.

Another type of CO_2/CH_4 reformer was integrated into a sodium reflux heat pipe and tested at the WIS solar furnace [56]. The receiver/reactor was a heat pipe with seven tubes inside an evacuated metal box containing sodium. The catalyst, 0.5 wt % Rh on alumina, filled the tubes. In operation, concentrated sunlight heated the front plate of the reactor and vaporized sodium from a wire mesh wick attached to the other side. Sodium vapor condensing on the reactor tubes released latent heat returning to the wick by gravity. The receiver system performed satisfactorily. The maximum power absorbed was 7.5 kW at a temperature of about 800°C.

The important feature of this technology is the decoupling of the reactor from the heating source by an intermediate heat transfer fluid. The receiver section is optimized in size and shape for solar absorption, while the reactor configuration is designed for maximum chemical performance. The sodium heat pipe is an approach for this solution. Liquid sodium contained in an evacuated chamber evaporates under heating of concentrated sunlight. The sodium vapor condenses on the reactor tubes in the chamber and liberates the heat of vaporization. The excellent heat transfer characteristics of evaporating and condensing sodium result in uniform temperatures throughout the chamber. Therefore, the reactor tubes remain isothermal and the energy flux is transformed from high values at the receiver to those required by the reaction. The flux profile in the reactor adjusts to the demands of the reacting system, which are affected by changes in catalyst activity, flow rate and reactor inlet temperature.

The isothermal reformer was also proposed for the future HTGR [57]. The HTGR can provide heat at a temperature range of 950–1000°C at the primary helium loop and 900–950°C at the secondary loop. The convective heat transfer from the helium gas to the reformer tube requires enhancement. The process working pressure in the reformer has to be slightly higher than the helium (usually, approximately 40 atm) for safety reasons, to avoid helium leaking out in case of failure in the reformer tube. These requirements are in contradiction with the thermodynamics of the reforming process, where a lower pressure in the reformer and higher heat transfer characteristics are needed. The isothermal reformer offers a possible solution to these problems. The helium in the primary cooling loop can be used to evaporate intermediate medium at a constant temperature. The vapors condense on the external surface of the reformer tubes (Figure 5.31), improving the heat transfer characteristics.

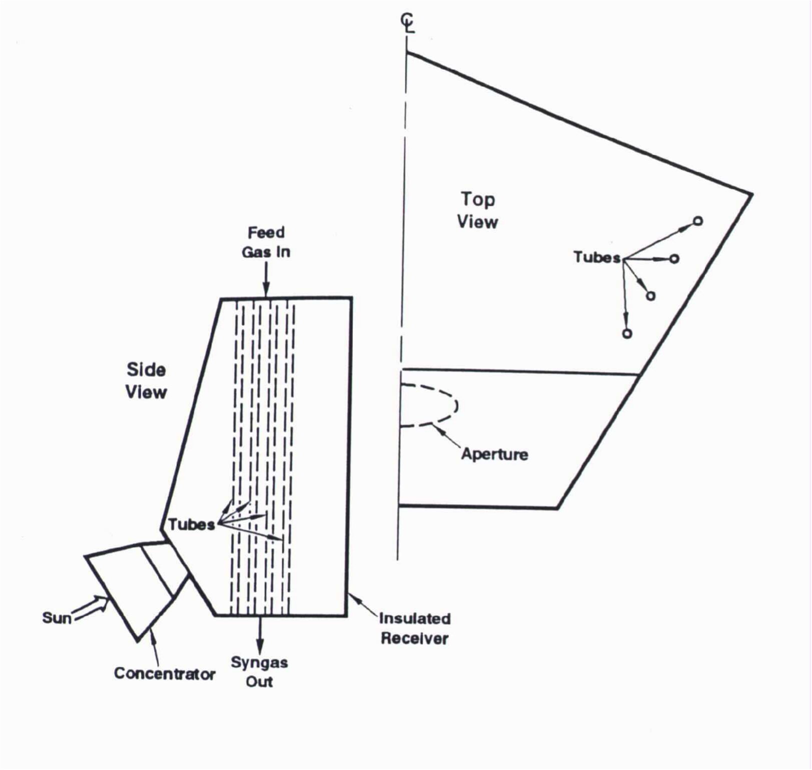


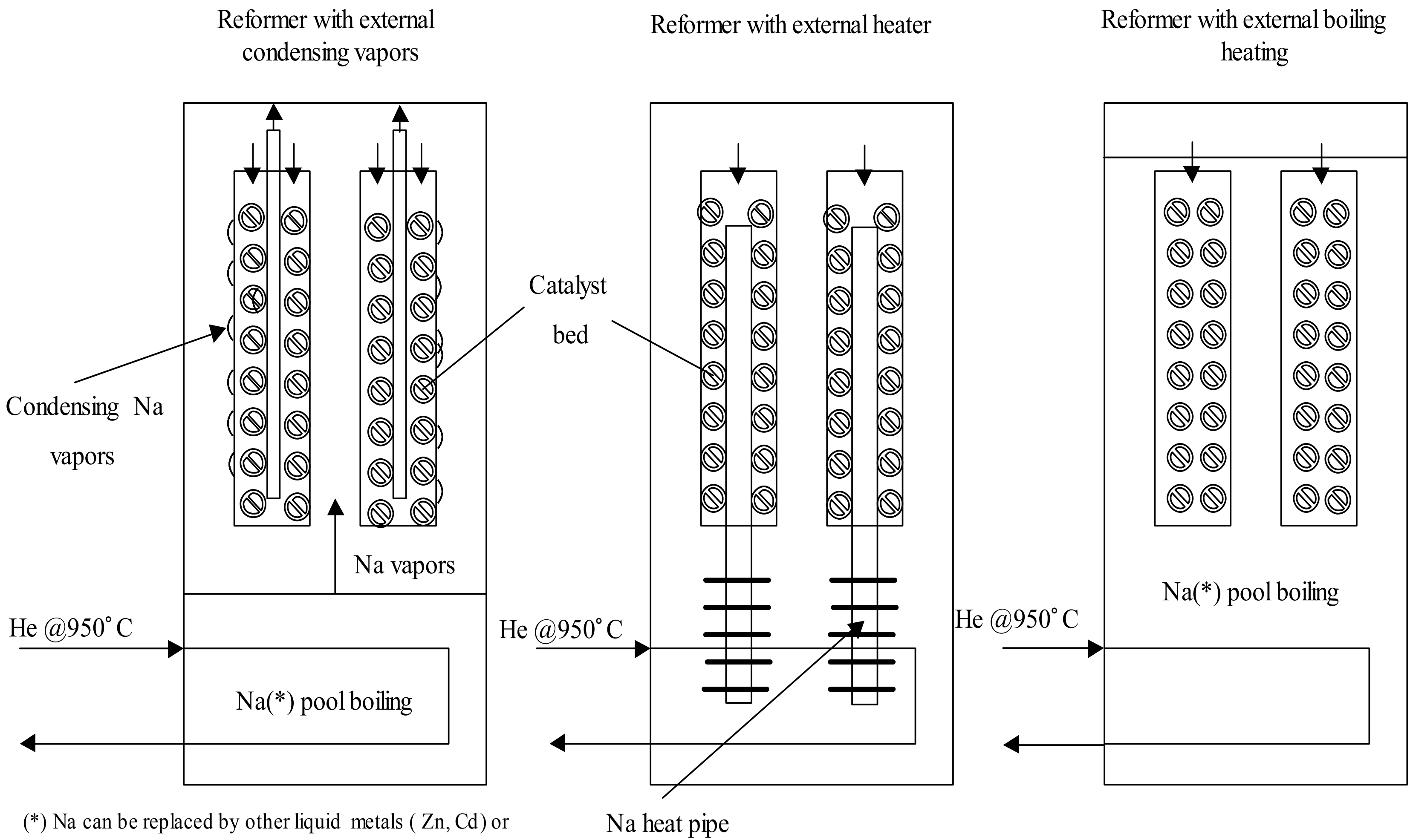
FIG. 5.30. The Weizmann Institute of Science 480-kW tubular reformer/receiver.

The pressure on the side of the intermediate medium can be as low as 1−2 atm, for instance, if sodium is selected. This affords opportunity for reducing the process pressure in the reformer, therefore, lowering the reforming temperature and increasing the CH_4 conversion yield.

In principle, industrial nuclear and solar reformers, which have been developed for steam reforming of natural gas, can be used for CO_2 reforming with minor mechanical modification, except for the catalytic system. These modifications include the inlet header and outlet manifold systems. The safety issues and solutions that are applicable for the nuclear steam reforming process can also be suitable for the CO_2 reforming process.

5.3.4 Potential applications for nuclear CO_2/CH_4 reformers

In general, the HTGR as a heat source for the reforming process offers the opportunity to use the nuclear power not only for the electricity sector, but also for the production of fuels for the transportation sector and chemicals for industry.



(*) Na can be replaced by other liquid metals (Zn, Cd) or molten salts ($ZnCl_2$, $SnCl_2$).

FIG. 5.31. Pseudo-isothermal reformers.

A few examples are listed below

(1) CO_2 reforming of methane to produce syngas as a fuel for direct combustion in a gas turbine or combined cycle. Nuclear heat contributes about 25% of the calorific value of the product gas compared to the feed. The conversion efficiency of heat to electricity in a combined cycle today reaches a value of 50%, compared to 40% efficiency in other nuclear reactors where steam is generated and conversion is performed by means of the Rankine cycle. This could be the immediate and simplest application for nuclear CO_2 reforming, since, in case of failure, the turbine can be operated in a hybrid mode and use a regular fuel, thus, the electricity production is not interrupted.

(2) Production of liquid fuels from the synthesis gas, such as methanol, synthetic gasoline or diesel oil through the Fischer-Tropsch process. Methanol is especially important not only as a direct fuel, but also as a starting material to other automotive fuel additives, turbine fuel, other alcohol substances and fuel for electric cars, which use fuel cells as converters of hydrogen to electricity. In this application, the interfacing between the methanol plant and the nuclear reformer is more complicated and requires a buffer storage and/or emergency means to operate the nuclear reactor smoothly, in case of failure in the methanol plant.

(3) Production of various chemicals.

(4) Production of hydrogen for hydrogenation processes, e.g., upgrading of heavy oil residues and for the production of chemicals like ammonia.

A particular application is the exploitation of the Natuna gas field in Indonesia, using the HTGR. The Natuna gas field, discovered in 1973, is located in the Natuna Sea, at a depth of 145 m, around 225 km northeast of the Natuna Island, which is situated 1,100 km north of Jakarta and 600 km northeast of Singapore. It is one of the largest natural gas reserves in the world, with a total gas volume of 210 TSCF or 6,000 BSCM. However, this gas contains about 67% mole carbon dioxide and 0.52% mole H_2S.

PERTAMINA, the national oil company of Indonesia, and ESSO Exploration and Production Natuna, Inc., an affiliate of EXXON Corporation, are planning the development of the Natuna gas field under an agreement awarded in 1980. The Natuna development concept includes offshore methane production facilities. The gas from the Natuna reservoir will be cryogenically separated into sales gas (mostly methane) and waste gas (mostly carbon dioxide). The facilities will include drilling and quarter platforms, in addition to the very large treating platforms. The hydrocarbon sales gas will be transported by a pipeline 225 km to Natuna Island, for further treatment and then liquefaction into LNG. Offshore and onshore waste gases will be piped to injection platforms located above very large high quality carbonate aquifers. Here, using proven reservoir gas injection techniques, the gas will be injected into the aquifers for permanent disposal. The high investment and operational costs for this concept result in double or triple price of the final LNG, compared to other gas fields.

An alternative proposal of the above concept could be the use of high temperature nuclear heat from an HTGR source, with direct CO_2 reforming of the methane or with the addition of H_2O. A cost estimate for the exploitation of the Natuna gas based on this concept was performed by Barnert [31]. Four different schemes have been analyzed. The result shows a potential for economical competitiveness because the synthetic product fuels that can be produced from the syngas are readily marketable as substitute for gasoline and diesel with relatively higher market values compared to regular methane, which has a lower market value. In addition, the transportation of methanol or other synthetic fuels overseas is cheaper and simpler than the transportation of LNG.

One major problem of the Natuna gas is CO_2 surplus, compared to the amount required stoichiometrically to convert all the methane to syngas. There are several solutions to this problem, among them, to mix methane-rich natural gas and create a molar ratio of CO_2/CH_4 in the range of 1–1.3. Another solution is to import hydrogen and convert the CO_2 surplus to different useful products (e.g., 4 moles of H_2 per one mole of CO_2 are required for conversion to CH_4, 3 moles for conversion to methanol and only 2 moles for conversion to acetaldehyde).

An additional example for HTGR utilization is to convert biogas to syngas. Biogas contains roughly 60–65% CH_4, and the balance is CO_2. In this case, it is necessary to add H_2O to balance the stoichiometry. The economical advantage of this process is the simultaneous handling of waste, and the HTGR contribution is in CO_2 mitigation, as well as in the ecological aspect.

5.4 GAS TURBINE FOR ELECTRICITY GENERATION

A promising approach for making good use of the high temperature capability of the HTGR is to use the primary helium coolant to drive a gas turbine in a direct closed cycle arrangement. In the 1970s, this was extensively studied in the U.S., Germany, the U.K. and France. At that time, the concept was based on enclosing a large (2000 to 3000 MW(th)) reactor core and the gas turbine power conversion system within a prestressed concrete reactor vessel. After nearly a decade of work, this concept was abandoned primarily because the system achieved only about 39% efficiency and would have required substantial development to resolve design and safety issues [58]. Subsequent technological advancements in the design and operation of magnetic bearings, compact plate-fin heat exchangers and turbomachines, coupled with international capability for their fabrication and testing, and the development of the annular core modular HTGR has resulted in renewed interest in this HTGR concept [59].

Tasks related to the helium gas turbine reactor conducted as part of the CRP included the following:

- An evaluation of the potential markets for different size plants
- To define and evaluate helium gas turbine reactor plant design requirements
- Evaluate the R&D base technologies of the gas turbine plant components
- Define R&D needs for interfacing technologies

Results of these activities were reported at the third Research Coordination meeting and included a re-confirmation of the international potential market for the gas turbine HTGR plant along with its technical feasibility. However, the potential value for conducting tests of gas turbine power conversion system components integrated with the HTTR was determined to be limited. This is due to the low power and pressure capabilities of the HTTR system.

It was recommended that modular HTGR related technology development be conducted at the HTTR (although, this would not be within the scope of the CRP) in areas including the reactor system, the reactor cavity cooling system, irradiation behaviour of graphite, of carbon/carbon composite control rods, fuel performance, etc. JAERI indicated that changes in the HTTR testing programme would be considered and were subject to approval and corresponding funding by the Japanese government.

Two specific closed cycle gas turbine HTGR systems are currently under substantial development for the generation of electricity. These include the Gas Turbine-Modular Helium Reactor (GT-MHR) and the Pebble Bed Modular Reactor (PBMR). A consortium of organizations within Russia belonging to the Russian Ministry on Atomic Energy (MINATOM) and companies from the USA (General Atomics Co.), France (Framatome) and Japan (Fuji Electric Co.) are in the process of developing the GT-MHR initially for the purpose of burning weapons plutonium, but with the long-term goal of commercialization utilizing a uranium fuelled HTGR.

Although these helium gas turbine reactor programs represent the most significant applications to date of development in this advanced nuclear power concept, international interest has also lead to the scientific investigation of other designs by Member States comprising the IAEA’s International Working Group on Gas Cooled Reactors. This section includes a general overview of the developmental aspects of the some of the helium gas turbine reactor systems with specific focus on the GT-MHR, as well as a review of the gas turbine programme and possible testing in conjunction with the HTTR and related facilities.

5.4.1 International GT-MHR programme — direct cycle

The GT-MHR retains the inherent safety characteristics of previous modular helium reactor designs to prevent meltdown and fuel failure even under the beyond-design-basis event scenarios. The GT-MHR also achieves lower capital power costs than previous designs through a combination of passive safety features, greater power conversion system simplicity and higher net thermal efficiency. The higher thermal efficiency also reduces thermal discharges and high-level wastes on a per-kilowatt basis. Low spent fuel plutonium content and the absence of GT-MHR spent fuel reprocessing capability, contribute toward making the GT-MHR fuel cycle highly proliferation resistant.

The GT-MHR represents an important option for resolving the proliferation risks of weapons-usable-plutonium. Over 90% of the initially charged weapons desirable plutonium-239 and over 70% of the total plutonium can be destroyed in a once through fuel cycle while achieving highly efficient energy production.

The GT-MHR couples the passively safe, high temperature modular helium reactor with a closed Brayton cycle (gas turbine) power conversion system to generate electricity at a net thermal conversion efficiency of ~ 47%. The reactor coolant, helium, directly drives a turbine-generator for the production of electricity. This improved nuclear power system has been made possible by new technology developments during the last decade. These include: large industrial gas turbines matching the thermal output of the modular reactor; large active magnetic bearings; compact, highly effective plate-fin heat exchangers; and high-strength, high temperature steel alloy vessels.

The high reactor outlet high heat rejection temperature capabilities of the GT-MHR also makes it well suited for process heat applications including district heating.

5.4.1.1 MHR plant description

The GT-MHR couples the reactor system contained in one vessel, with a high efficiency gas turbine energy conversion system contained in an adjacent vessel. The reactor and power conversion vessels are interconnected with a short cross-vessel and are located below grade in a cylindrical silo (Figure 5.32).

The reactor system employs a graphite moderator and features refractory fuel known as TRISO-coated particle fuel. The TRISO-coated fuel, shown in Figure 5.33, contains a spherical kernel of fissile or fertile material, as appropriate for the application, encapsulated in multiple coating layers. A low density carbon (buffer) layer surrounds the kernel to attenuate fission recoil atoms and provide void volume to accommodate fission gases. Surrounding the buffer is an inner pyrocarbon coating (IPyC), a silicon carbide (SiC) layer, and an outer pyrocarbon coating (OPyC). The IPyC, SiC, and OPyC layers together form a miniature, highly corrosion resistent pressure vessel and an essentially impermeable barrier to the release of gaseous and metallic fission products. Extensive tests in the US, Europe, and Japan have demonstrated excellent performance characteristics of this fuel.

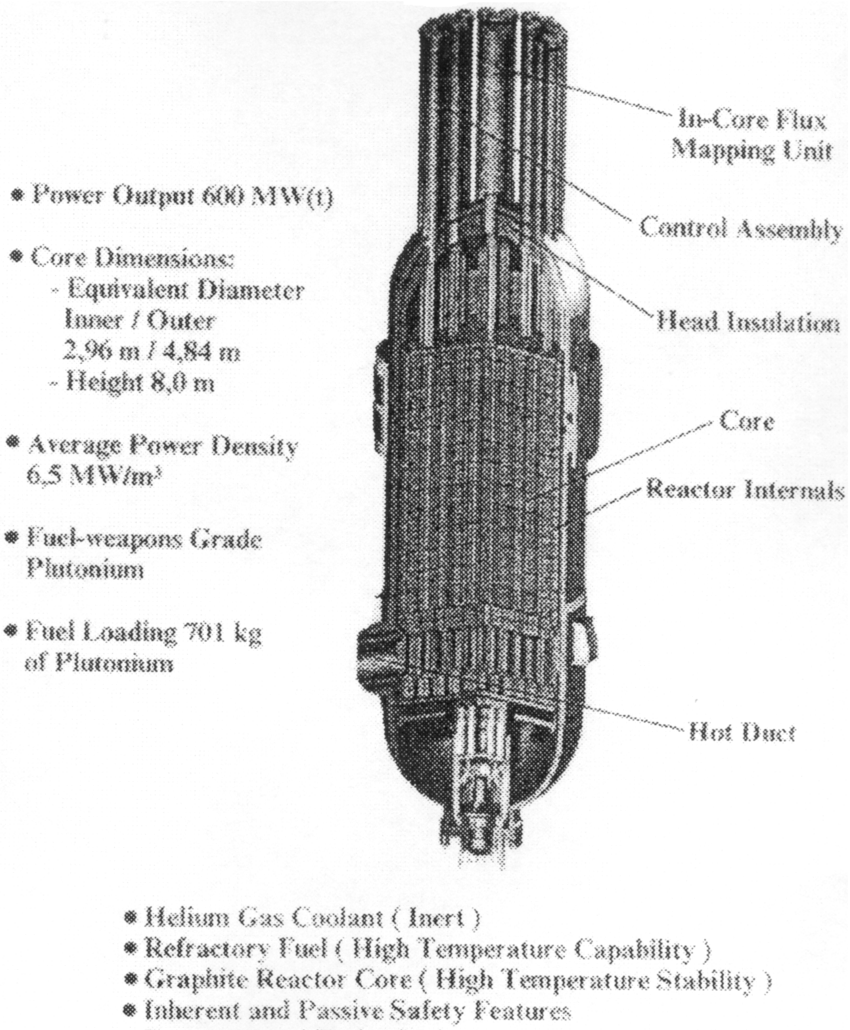


FIG. 5.32. GT-MHR reactor system as designed by OKBM.

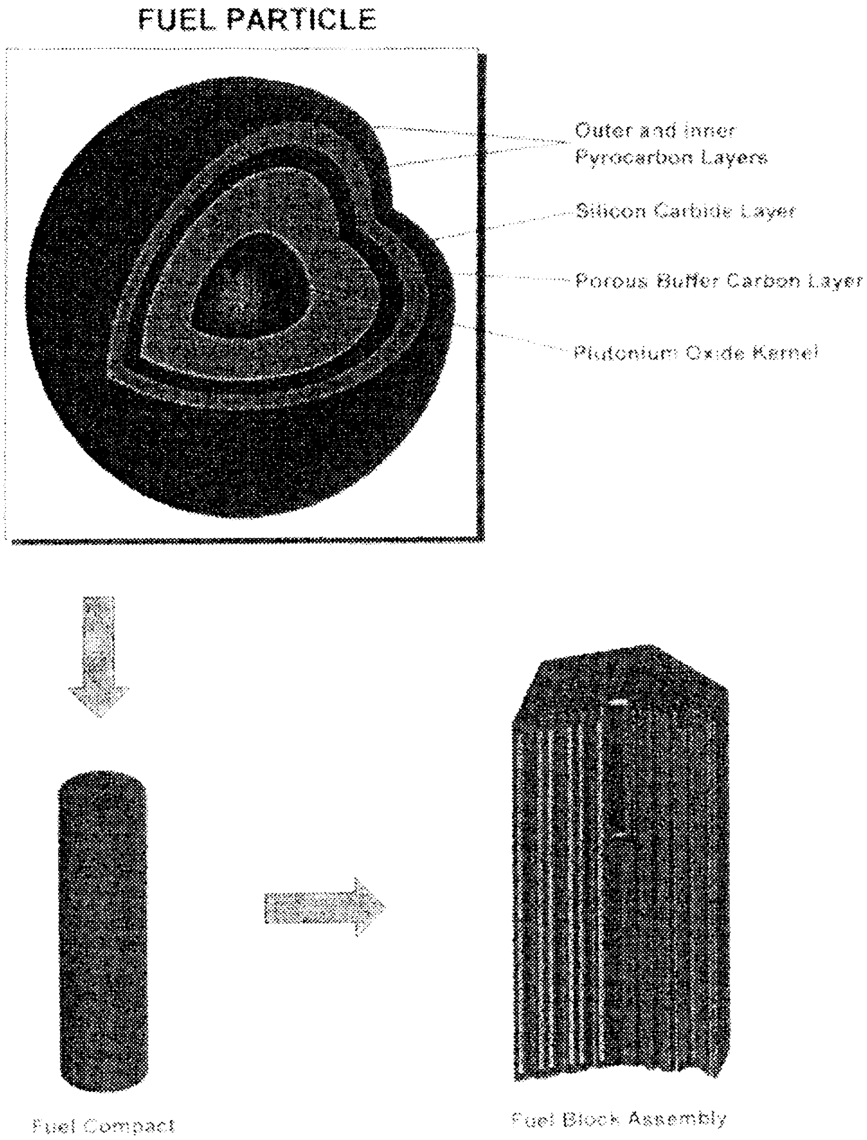


FIG. 5.33. GT-MHR fuel system.

The overall dimensions of standard TRISO-coated particles vary from about 650 microns to about 850 microns. For the GT-MHR, TRISO-coated particles are bonded with a graphitic matrix to form cylindrical fuel compacts approximately 12.5 mm in diameter and 50 mm long. Approximately 3000 fuel compacts are loaded into hexagonal graphite fuel elements, 800 mm long by 360 mm across flats. One hundred and two columns of the hexagonal fuel elements stacked 10 elements high are used to form an annular core. Reflector blocks are located inside and outside of the active core.

TRISO-coated particle fuel remains stable to very high temperatures. The coatings do not start to thermally degrade until temperatures approaching 2000°C are reached. Normal operating temperatures do not exceed about 1250°C and worst case accident temperatures are maintained below 1600°C. Thus, fuel failure and fission product release is prevented.

The coated particle fuel is also an excellent spent fuel waste form for both long-term interim storage and permanent geologic disposal. The refractory coatings retain their integrity in a repository environment for hundreds of thousands of years. As such, they can ensure that the long-lived radionuclides are contained for geologic time frames and do not migrate to the biosphere.

In the GT-MHR primary system, helium is heated in the reactor and expanded through a gas turbine to generate electricity. Helium from the turbine exhaust flows through the hot side of a recuperator transferring residual heat energy to helium on the recuperator cold side returning to the reactor. From the recuperator, the helium flows through a precooler where it is further cooled. The cooled helium then passes through low and high-pressure compressors with intercooling. From the compressor outlet, the helium flows through the cold, high-pressure side of the recuperator where it is heated for return to the reactor. Nominal full power operating parameters are given in Table 5-9.

Table 5-9: GT-MHR Nominal Full Power Operating Parameters

Reactor power, MW(th)	600
Core inlet/outlet temperatures, ^{o}C	491/850
Core inlet/outlet pressures, MPa	7.07/7.02
Helium mass flow rate, Kg/s	320
Turbine inlet/outlet temperatures, ^{o}C	848/511
Turbine inlet/outlet pressures, MPa	7.01/2.64
Recuperator hot side inlet/outlet temps, ^{o}C	511/125
Recuperator cold side inlet/outlet temps, ^{o}C	105/491
Net electrical output, MW(e)	284
Net plant efficiency, %	47

Figure 5.34 shows the GT-MHR turbo-compressor. The entire gas turbine power conversion system is a direct, recuperated and intercooled, closed Brayton Cycle.

The power conversion system is entirely contained within the primary coolant system. The turbomachine incorporates a 12-stage turbine and high and low pressure compressors on the same shaft with the generator. The turbomachine utilizes the same technology, materials and design methods as large aircraft and industrial gas turbines, except that blade designs are modified for the thermodynamic properties of helium. The size is equivalent to the largest open-cycle gas turbines currently being built.

The turbomachine is supported by large active magnetic bearings. The bearing loads are larger than currently demonstrated large bearing technology (e.g. natural gas line compressors), but speeds and control techniques for individual bearings are similar. Control of multi-bearing systems (i.e. more than two radial bearings) at speeds exceeding the critical speed has been demonstrated for magnetic bearing systems.

High-grade turbine exhaust heat is recovered by compact, highly effective plate-fin heat exchangers. Effectiveness equal to 95% or greater have been demonstrated in utility gas turbine applications. The precooler and intercoolers are based on conventional finned-tube heat exchanger designs. The GT-MHR employs a modular approach to these heat exchangers to facilitate maintenance and replacement.

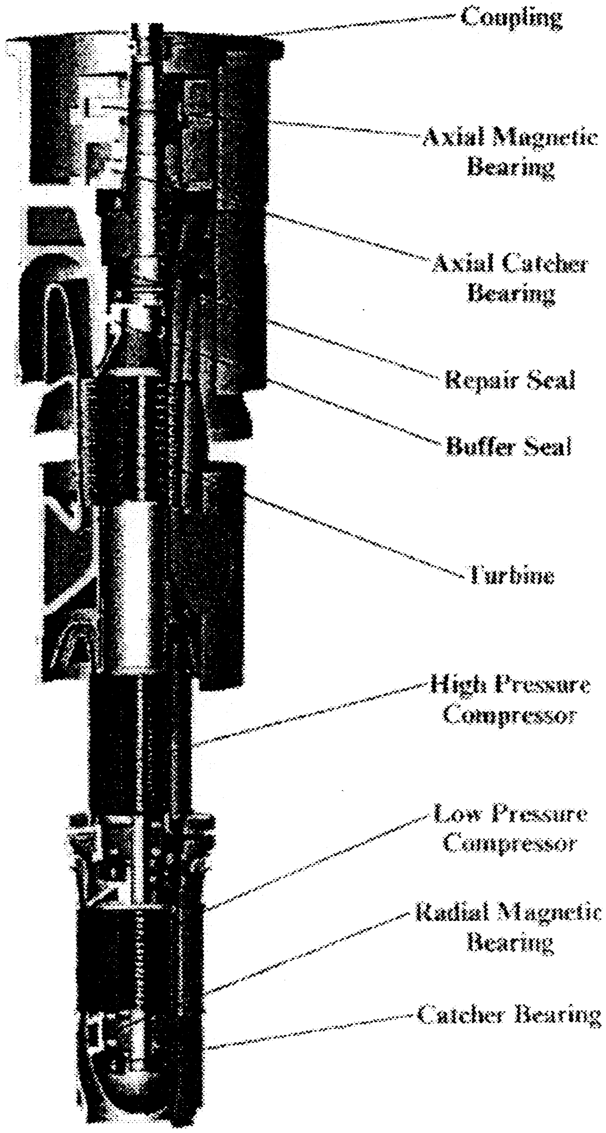


FIG. 5.34. GT-MHR turbo-generator arrangement.

5.4.1.2 GT-MHR safety

The GT-MHR is meltdown-proof and passively safe. This is achieved through a combination of inherent safety characteristics and design selections that take maximum advantage of these characteristics. These design selections and features include:

- Helium coolant, which is single phase, inert, and has no reactivity effects;
- Graphite core, which provides high heat capacity and slow thermal response, and structural stability at very high temperatures;
- Refractory coated particle fuel, which allows extremely high burnup and retains fission products at temperatures much higher than normal operation;
- Negative temperature coefficient of reactivity, which inherently shuts down the core above normal operating temperatures; and
- An annular, low power density core in an un-insulated steel reactor vessel surrounded by a reactor cavity cooling system, which enable passive heat transfer from the core to the ultimate heat sink while maintaining fuel temperatures below damage limits.

These safety design features result in a reactor that can withstand loss of coolant circulation or loss of coolant inventory and maintain fuel temperatures below damage limits.

5.4.1.3 GT-MHR plutonium disposition features

The design of the GT-MHR core and fuel enable the following benefits relative to the destruction of unwanted plutonium and prevention of material diversion:

***Effective plutonium destruction*: The GT-MHR provides the capability to consume more than 90% of the initially charged plutonium-239 and more than 70% of the initially charged total plutonium in a single pass through the reactor. By achieving this high level of plutonium destruction, the GT-MHR extracts a substantially higher portion of the useful energy content from the material than other reactor options without reprocessing and recycle. Because the plutonium fueled GT-MHR uses no fertile fuel material, all fissions in the core are plutonium fissions, and no new plutonium is produced by the operation of the reactor.

Diversion/proliferation resistance: The GT-MHR is particularly well suited for international deployment for plutonium disposition. Both the fresh fuel and the spent fuel discharged from the GT-MHR have higher resistance to diversion and proliferation than other reactor options for plutonium disposition. The plutonium content of the fresh fuel is very diluted within the fuel element graphite. In addition to having the self-protecting characteristics of other spent fuel (high radiation fields and spent fuel mass and volume), the amount of plutonium per GT-MHR spent fuel element is very low (0.15 kg of all fission isotopes per 115 kg of fuel block), there is no developed process and capability anywhere in the world for separating the residual plutonium from GT-MHR spent fuel, and the discharged plutonium isotopic mixture is severely degraded making it particularly unattractive for use in weapons.

Relevant conclusions concerning the GT-MHR are as follows:

- The GT-MHR is a promising nuclear energy source for the next century. The design addresses the current concerns with nuclear power with regard to safety, economics, proliferation resistance, and high level waste disposal;
- The GT-MHR is attractive for the disposition of weapons plutonium. High levels of plutonium destruction are achieved, a high portion of the useful energy content from the material is obtained without reprocessing and recycle, and the fuel is highly diversion and proliferation resistant;
- The GT-MHR's small modular size coupled with its safety, economic, environmental, and proliferation resistant characteristics make this plant an excellent candidate to meet the growing electricity demand in developing countries.

5.4.2 South Africa programme — direct cycle [59, 60]

The PBMR was first identified by the electric utility of South Africa, ESKOM, in 1993 as an option for expansion of their electrical generation capacity. Subsequently, ESKOM contracted with Integrators of System Technology (IST) to perform a technical and economic study of the feasibility of the PBMR for the generation of electricity. This study, which was completed in early 1997, supported the continued development of the PBMR. Reactor development follows the HTR-MODUL pebble bed that was previously licensed in Germany for commercial operation.

A series of internal reviews by ESKOM and subsequent independent reviews of this feasibility study by international entities have generally acknowledged that the PBMR is technically and economically capable of meeting the requirements originally set by ESKOM for commercialization. These requirements included: (a) New generation capacity capable of being located where the load growth is taking place in South Africa (coastal regions); (b) Small, modular increments of electrical generation capacity corresponding to system growth needs; (c) Reduced exposure to negative environmental issues such as carbon dioxide emissions and capable of providing a strategy for economic mitigation of greenhouse gas reductions; (d) Generating plants placed where there would be a limited need for extensive transmission system additions, and; (e) Cost of capital and plant operation to be within those costs presently being achieved at ESKOM's largest coal fired stations (with targets of capital cost less than US$ 1000/installed kW and overall generation, transmission and distribution costs of utility operations within 2.0 cents/kW·h.

The simplified schematic flow and Brayton cycle diagrams of the PBMR are provided herein as Figure 5.35 and Figure 5.36, respectively.

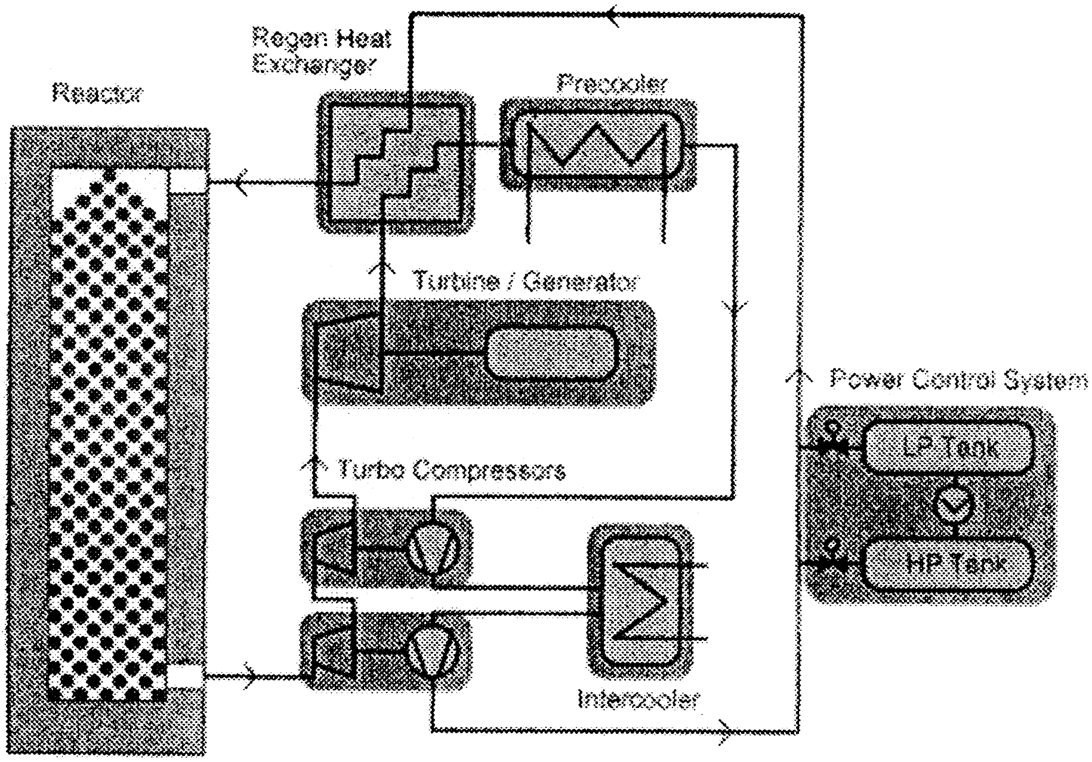


FIG. 5.35. Schematic layout of the P BMR.

A thermal power level of 265MW is calculated to provide a gross electrical generation of 117 MW(e). Helium, at a pressure of ~7.0 MPa and a temperature of 900°C. leaves the pebble bed reactor and flows through the high and low pressure turbines providing energy to drive their associated compressors. The helium, at ~721°C. and ~4.4 MPa, then enters the power turbine providing energy to drive the electrical generator. The helium at ~554°C. then enters the recuperator which provides heat exchange to the helium returning to the core. After leaving the recuperator (at ~140°C.), the helium passes through the precooler and exits this cooler at conditions of ~27°C. and ~2.6 MPa. It is then compressed by passing through the low pressure compressor, intercooler and then the high pressure compressor prior to entering the high pressure side of the recuperator. The recuperator heats the helium from ~104°C. to ~536°C. at a pressure of ~7.0 MPa before entering the top of the reactor. Table 5-10 lists key performance data and Figure 5.37 includes a layout of the system.

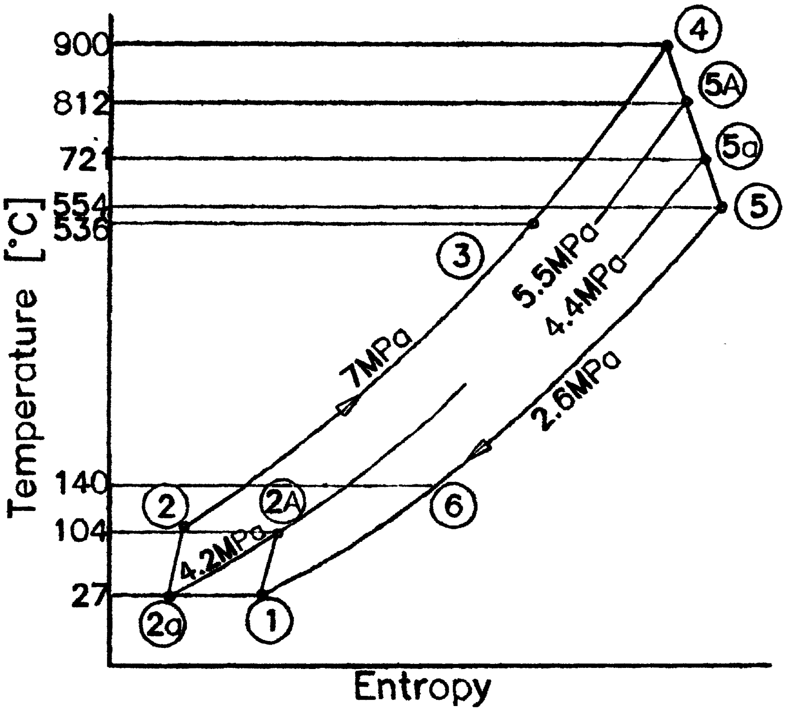


FIG. 5.36. Brayton cycle diagram of the PBMR.

Design of the PBMR is currently in progress with construction of the first unit tentatively scheduled to begin July 2001. ESKOM's intent is to utilize the first unit to demonstrate the operating and accident response characteristics of the PBMR as the means of obtaining full licensing of the design and to provide this plant as a commercial product for marketing to other countries/utilities. Commercial operation of the first unit is scheduled for 2005.

Table 5-10: PBMR Key Performance Data/Unit

Thermal power	265MW(th)
Electrical generation	117MW/114MW(net)
Core inlet pressure	7.0MPa
Core helium outlet temperature	900°C.
Avg. core power density	2.4MW/cubic meter
Ramp rate	10MW/minute
Load rejection w/o trip	100%
Plant life	40 years
General overhauls	30 days each 72 months
Plant staff level (for 10 module plant)	80 persons
Emergency planning zone	400 meters
Construction period	24 months
Capital cost	< \$100 million/module
Fuel cost	< \$4/MW·h

5.4.3 Chinese programme — indirect cycle

5.4.3.1 Systemic design

Considering the problems of the possible radioactivity deposition in the turbine blades in the direct GT-MHTGR, and the lower inlet helium temperature (250°C-300°C) in the indirect gas turbine cycle MHTGR, the indirect gas turbine and steam turbine combined cycles (GT-ST-MHTGR) were studied in China. The GT-ST-MHTGR can be used to generate electricity more efficiently. The reactor heat is transferred to the secondary loops by means of an Intermediate Heat exchanger (IHX) and a Steam Generator (SG). There are two patterns of GT-ST cycles, i.e. a parallel GT-ST cycle and a series GT-ST cycle. In the former pattern, the IHX and SG are directly connected with the reactor primary loop, the gas turbine cycle and the steam turbine cycle are arranged in parallel manner. In the latter pattern, IHX and the gas turbine cycle are arranged in the secondary loop, while the SG and the steam turbine cycle are arranged in the third loop. The system thermal efficiencies of a parallel GT-ST cycle and a series GT-ST cycle are 47% and 45.6 % respectively [61].

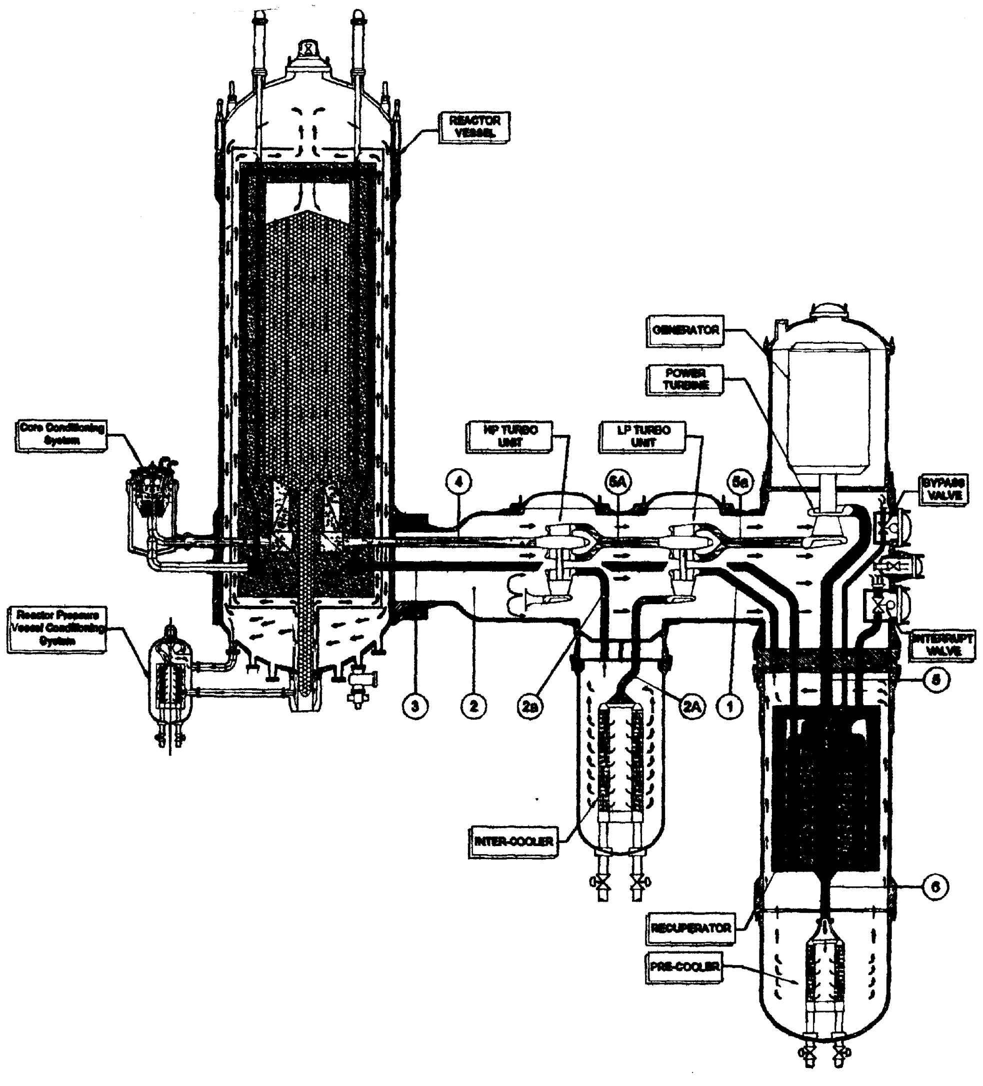


FIG. 5.37. PBMR system layout.

In China, a 10MW(th) high temperature gas cooled reactor test module (HTR-10) [62] is under construction at site of Institute of Nuclear Energy Technology of Tsinghua University (INET). There are two phases of the high temperature heat utilization in this project. In the first phase, the reactor core outlet temperature is 700°C and a conventional ST cycle is used in the secondary loop. In the second phase, the reactor core outlet temperature is increased up to 900°C and a GT-ST combined cycle is planned for demonstration.

The HTR-10 design features allow it to accept a gas/gas intermediate heat exchanger (IHX) in series with the steam generator, which gives the HTR-10 flexibility for multi-aspect applications. A gas turbine/steam turbine (GT-ST) combined cycle could be connected with the secondary gas circuit for a high efficiency power generation test. Figure 5.38 shows the schematic flow of the GT-ST combined cycle for the HTR-10. The main advantage of using the HTR-10 for the GT-ST combined cycle is that the basically well-developed HTGR design and associated experience provide a secure energy source. For example, the inlet temperature of the reactor core can be kept at ~ 300°C. Of course, with adoption of IHX and double cycles, the economic competitiveness is decreased and the system operation and control are more complex. The main design data for the GT-ST combined cycle are listed in Table 5-11.

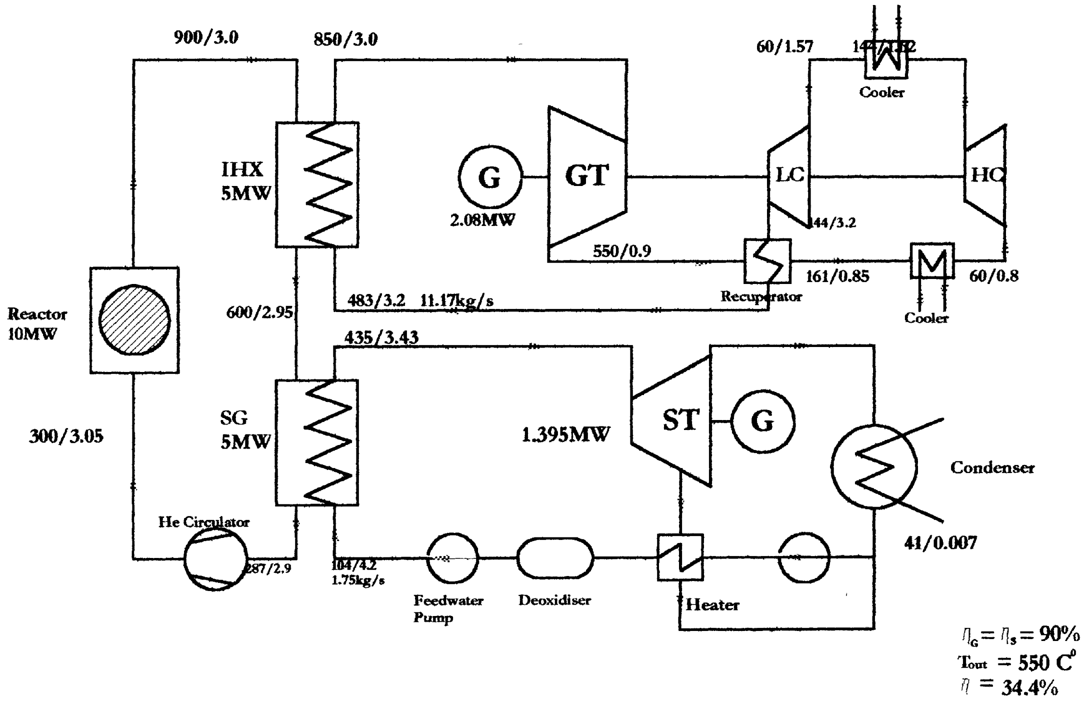


FIG. 5.38. Schematic flow diagram of the GT-ST combined cycle for the HTR-10.

The main purposes of the experiment of the GT-ST combined cycle with the HTR-10 are as following:

- Demonstration of gas turbine coupled with a nuclear reactor. Safety related test of the HTGR-GT-ST system, which would be valuable for development of commercial HTGR-GT plants.
- Thermodynamic simulation of the commercial HTGR-GT plant gas-turbine cycle. (Properly selected operating pressures and working fluids can meet the thermodynamic simulation conditions, such as, Mach number, Reynolds number, specific velocity, etc.).
- Verification of protection and control of HTGR-GT-ST.

- Optimization of the GT cycle.
- Verification of co-ordinate control of GT and ST cycles.
- Selected component development (seal, bearing, etc.).

Table 5-11: Main Data for the GT-ST Combined Cycle

Reactor		
Core thermal power	MW	10
Core outlet temperature	°C	900
Core inlet temperature	°C	300
Primary pressure	MPa	3.0
IHX		
Thermal power	MW	5
Primary helium inlet temperature	°C	900
Primary helium outlet temperature	°C	600
Primary pressure	MPa	3.0
Secondary nitrogen inlet temperature	°C	483
Secondary nitrogen outlet temp.	°C	850
Secondary pressure	MPa	3.2
Nitrogen flow rate	kg/s	11.17
SG		
Thermal power	MW	5
Temperature at helium side	°C	600/287
Temperature at water side	°C	435/104
Pressure at water side	MPa	3.43/4.2
Power		
Power for gas turbine	MW(e)	2.08
Power for steam turbine	MW(e)	1.36
Total efficiency	%	34.4

5.4.3.2 Conceptual design of the intermediate heat exchanger for the HTR-10 [63]

In order to provide a more compact arrangement and increase the power density of the IHX for the HTR-10, the multi-concentric helical bundle was taken into consideration. Compared the modular helical tube-bundle, the structure of multi-concentric helical bundle has distinguishing features as follows:

- The structure is more compact, and the power density and surface compactness is greater than that of the modular helical tube-bundle.
- The diameter of the helical tube is large, the hydraulic resistance at the tube side is lower.
- The rigidity of the tube structure is poor.
- The manufacture and installation are complex.

A compact structure can reduce both the diameter of IHX's tube-bundle and the diameter of integrated IHX and SG pressure vessel. Therefore a reduction in the cost of the component and the reactor building can be achieved.

Lower hydraulic resistance not only reduces the power consumption of the compressor, but also makes the pressure difference between the inside and outside of the tubes easy to be controlled. In the case of the IHX' operation at high temperature, the choice of materials with the necessary properties is limited. Even for the best available materials, the allowable stresses are small. In order to assure the safe operation of the IHX, one of the important means is to control the pressure difference between two sides of the IHX and to reduce the pressure load. The main parameters for the IHX with multi-concentric helical tube bundle are given in Table 5-12.

Table 5-12: Main parameters of the IHX for the HTR-10
(Multi-concentric helical tube bundle)

Thermal power	MW	5
Working medium at primary side		He
Working medium at secondary side		N_2
Inlet/outlet temperature at primary side	°C	900/600
Inlet pressure at primary side	MPa	3.0
Mass flow at primary side	kg/s	3.21
Inlet/outlet temperature at secondary side	°C	483/850
Inlet pressure at secondary side	MPa	3.1
Mass flow at secondary side	kg/s	11.17
Pressure loss at primary side	kPa	15
Pressure loss at secondary side	kPa	70
Diameter of the central pipe	mm	$\phi950 \times 20$
Diameter of the heat transfer tube	mm	$\phi22 \times 2$
Number of heat transfer tubes		127
Number of radial layers		7
The largest diameter of the helical tube	mm	1344
Heat transfer area	m^2	153
Tube material		Inconel-617
Inside diameter of the pressure vessel for integrated IHX and SG	mm	2500

5.4.4 Component testing programme

The development of technology for future MHRs (modular helium reactors) should be one of the key missions for the HTTR. This section discusses MHR developmental areas for (possible) HTTR support.

Before discussing specific developmental activities that could support MHR development, the importance of routine day-to-day HTTR operation must also be noted. The routine experience accumulated during long term operation of the HTTR will be most valuable to the commercial development of the MHRs. Success and any difficulties encountered during everyday operation, how those difficulties were solved, which HTTR design features performed well, and which features required attention will all be of great interest to future MHR development. In addition, HTTR operating data will not only be necessary for the validation of the HTTR design and analysis methods, but also can be very useful to future validation of MHR analysis methods.

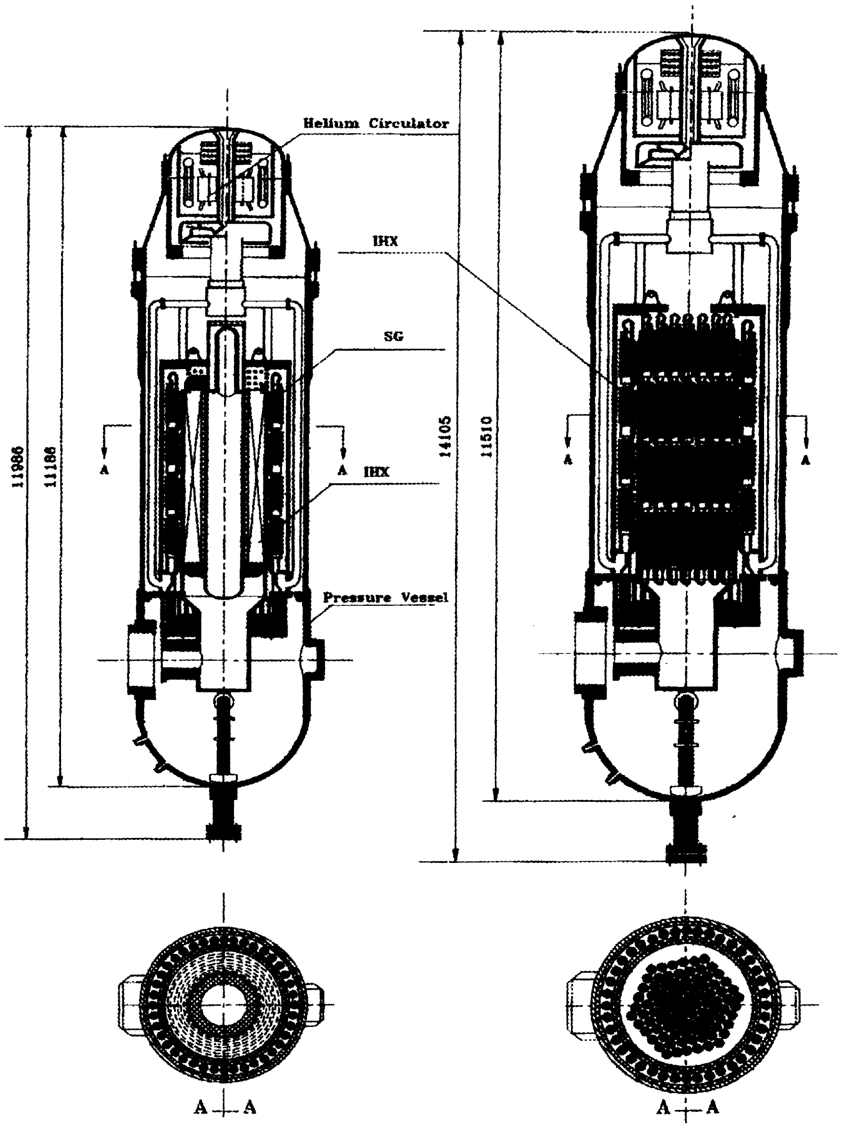


Fig.2. Integrated IHX/SG
(Multi–concentric helical tube bundle for IHX)

Fig.3. Integrated IHX/SG
(Modular helical tube bundle for IHX)

FIG. 5.39. IHX cross-section.

During the conceptual design of the Modular Helium Reactors at GA, the developmental efforts required to support the design of the MHR were identified. The MHRs can be deployed in the near term without the need for fundamental research because of the availability of a considerable gas-cooled reactor technology base. This technology has been demonstrated by more than 50 gas-cooled reactors built and operated in the United Kingdom, Germany, France, Italy, Spain, Japan, and the United States since 1956 (including the Peach Bottom and Fort St. Vrain reactors in the US). The Japanese HTTR, scheduled to go on line in 1998, will also provide significant data directly applicable to MHRs.

The engineering development needed for the MHRs falls basically into two categories; (1) technology development which provides data for design methods and validation of computer codes, and (2) component or process verification including prototypical component testing. The requirements for engineering development data are specified for the MHRs, and the needed data and the recommended approach to obtain the data (e.g. test program) on a schedule consistent with the program planning has been developed.

The scope of engineering development for the MHRs includes the following areas:

- Fuel development
- Reactor physics
- Thermal hydraulics
- Structural materials
- Instrumentation and controls
- Service equipment
- Power conversion system integration
- Component development

Each of these MHR developmental needs are summarized below. Some of these developmental needs can be satisfied using the HTTR. Others may be met by the supporting HTTR facilities, e.g., HTTR fuel fabrication facilities. The developmental tests which could be performing using HTTR facilities are identified below. After review of these developmental tasks by JAERI, it is recommended that GA further define details of the tests for HTTR during next contract work on HTTR support.

5.4.4.1 Fuel development

The MHR development activities on fuel are focused on TRISO coated uranium or plutonium fuel. The MHR could be deployed either for commercial electricity production or for burning surplus weapon grade plutonium along with electricity production. The fuel development needs for both applications are divided into four areas as described below.

Fuel fabrication

The main fuel fabrication activities include the development of equipment and processes for the construction of fuel fabrication facility at the MHR site as follows:

- Unit processes and equipment development for the uranium or plutonium oxide kernel process, coating process and cylindrical compact fabrication. Plutonium scrap and waste recovery for plutonium requires automation and robotics technology for remote operation and shielding.
- Development of process steps for the fabrication of burnable poison erbium oxide kernels, PyC coating applications and compacting in a hands-on facility.
- Development of quality control (QC) methods for defect detection and contamination for uranium or plutonium fuel compacts. In the case of Pu, QC to be performed in remote glove boxes.
- Design, construction and operation of a lab-scale line for resolution of design concerns and issues in the fuel fabrication process and the fabrication of demonstration fuel for irradiation testing.
- Design, construction and operation of a full-size equipment demonstration facility for the fabrication of qualification test fuel and proof test fuel for irradiation testing.

Some of the above developmental tasks can be performed at HTTR fuel fabrication facilities.

Fuel performance

The fuel performance activities are aimed to qualify the use of uranium or plutonium fuels in the MHRs and validate fuel performance models and include:

- Qualification testing, in-reactor, of uranium or plutonium fuel particles fabricated with lab-scale equipment to achieve high burnup under peak MHR conditions for normal operation and postulated accidents.
- Single-effects testing, both in- and out-of-reactor, to develop fuel performance models for uranium or plutonium fuel particles under conditions for normal operation and postulated accidents.
- Integral testing, typically in-reactor, under representative reactor conditions to independently validate fuel performance models and codes under normal operation and postulated accidents.
- Proof testing, in-reactor, of uranium or plutonium fuel fabricated by full-size production equipment under near-real-time reactor conditions.
- Confirmation testing, in-reactor, of plutonium fuel fabricated with fuel fabrication equipment from the Fuel Fabrication Facility (FFF).
- Qualification and proof testing of fuel fabricated in comparable facilities for uranium fuel.

The MHR fuel developmental tests could be performed in the HTTR at approximate temperatures, burnup and fluence.

Radionuclide transport

The main fission product activities are in support of transport model refinement and core validation for uranium or plutonium fuel and include:

- Single effects tests on intact and designed-to-fail fuel particles to determine gas releases (Kr, Xe, I and Te) and metal releases (C_s, Ag and Sn) under normal operating conditions and postulated accidents including moisture ingress.
- Single effects test on unirradiated and irradiated compact matrix and graphites to determine diffusivities and sorbtivities of Cs, Sr, Ag and Pu.
- Out-of-pile fission product transport loop tests to characterize plateout, liftoff, and washoff, including dust effects of Ce, Ag, I and Te under representative PCS service conditions with special emphasis on turbine and recuperator metals.
- Post irradiation heat-up tests of fuel compacts approximating dry and wet core conduction cooldown accidents to measure fission product releases.
- Integral, in-reactor, loop tests of designed-to-fail fuel particles in fuel compacts contained in graphite bodies to provide radionuclide transport data in the power conversion system and graphite corrosion data.

Some of these tests could be performed in HTTR and related JAERI facilities. It is recommended that further details be developed during the next phase of the HTTR support work.

Core corrosion

The core corrosion activities address the development of improved analytical component models and material property correlations for describing the interaction of corrosive agents, principally water and air, with core materials under normal operating and accident conditions and include:

- Single effects tests to characterize the reaction kinetics for PyC-coated B_4C and Er_20_3 poison granules.
- Single effects tests to characterize both the transport of coolant impurities and corrosion products in a thermosetting-resin matrix material and its intrinsic reaction kinetics.
- Integral test data from fuel and fission product transport tests described above for independent code validation.

The HTTR operational data may be able to provide some of the information in satisfying part of the developmental needs on core corrosion.

5.4.4.2 Reactor physics development

The "reactor physics development" primarily addresses the unique features of a plutonium core and erbium burnable poison for added reactivity control. Specific data are needed for validation of the reactor physics codes including reactivity worth of plutonium fuel and erbium, reactivity effects of moisture in the core, and radial and axial neutron flux distributions. These data could be generally obtained from benchmark reactor core criticality calculations. Also, HTTR validated physics codes may help in confirming this physics data.

5.4.4.3 Thermal hydraulic development

The "thermal hydraulic development" describes basic thermal hydraulic data needed by the reactor core designers for design assessment and validation of computer codes. These include primarily flow distributions and pressure drop data for core components, and thermal mixing at the core outlet.

These data are obtained generally from component test programs performed in air including a power conversion system/shutdown cooling system air flow test which investigates the flow and hot streak attenuation along the flow paths to the turbine and shutdown heat exchanger inlets. Also, HTTR validated thermal-hydraulic codes can help in further confirming the MHR thermal-hydraulic data.

5.4.4.4 Structural materials development

The "structural materials development" addresses the needs for test data on material properties and strength of materials selected for use in MHR components, including environmental effects. Material data are needed in the following areas:

- Reactor core graphites
- Reactor metals
- Reactor ceramics
- Vessel materials
- Heat exchanger materials

Some of these developmental needs can be satisfied by testing in the HTTR under helium conditions comparable to MHR operation. After review of the following developmental tests by JAERI, it is recommended that GA further define details of tests for the HTTR during the next phase of the HTTR support work.

Reactor core graphite development

Reactor core graphites include H-451 and HLM: Although an extensive database already exists for these graphites, additional data are needed for the MHR to cover specific MHR design conditions, increase the statistical basis, and satisfy more stringent requirements for experimental data for safety related components. Tests on irradiated and unirradiated graphite specimens will be performed to determine strength, fracture, thermal properties, and dimensional change and creep as well as tests to characterize the oxidation effects on graphite from air and water. In addition, new sources of raw materials, the screening to select one or more candidate cokes which will be used to produce H-451 graphite, can be completed.

Reactor metals

Reactor metals include Alloy 800H for the reactor internals and hot duct: Since Alloy 800H is an ASME code qualified material, and a significant data base already exists, only supplemental data on environmental effects (i.e., irradiation and helium impurities) on material properties covering the range of MHR specific service conditions are needed.

Reactor ceramics

Reactor ceramic materials include Aluminosilicate: Grade AD-85 insulation pads for the core support structure are the same material employed in Fort St. Vrain (FSV). Since the pads in the MHR are larger than those used in FSV, additional testing to establish material properties is needed because the properties of ceramics are dependent on the size of the manufactured billet.

Vessel materials

Vessel materials for reactor vessel, cross duct and power conversion vessel include modified 9Cr-1Mo-V ferritic steel, SA-387 Grade 91, Class 2 plate and SA-336 Grade F91 forging. Bolting material is high temperature Alloy 718 austenitic nickel-iron-chromium-molybdenum-niobium alloy. 9Cr-1Mo-V is an ASME code material but not currently approved for use at MHR design temperatures of 1000°F for Section III, Division 1, Class 1 components. Additional property data particularly on heavy sections, are required to include approval for 9Cr-1Mo-V in Code Case N-47 currently being pursued by Oak Ridge National Laboratory (ORNL) and ABB-CE. Tensile and charpy V-notch tests are also needed to determine the neutron-induced, nil-ductility transition temperature (NDTT) shifts for 9Cr-1Mo-V plate, forgings, weldments, and weld heat-affected zones. Tests on the Alloy 718 bolting material are needed to characterize the effects of irradiation induced changes in high temperature creep and irradiation induced creep on stress-relaxation properties at temperatures up to 1000°F.

Heat exchanger material

Heat exchanger materials include Type 316L stainless steel for the recuperator, 1/2Cr-1/2Mo steel for the precooler/intercooler, and 2-1/4Cr-1Mo steel for the shutdown heat exchanger (SHE). These materials are ASME code qualified, and extensive laboratory testing for a large range of temperatures and helium impurity levels (H_2, H_20, CO, CO_2 and CH_4)

have been carried out in US, Europe and Japan. However, additional environmental tests, including the effects of helium impurities and water, fission products (e.g., Cs, Te, I) and impurities released directly from the fuel (e.g., Cl, S) on material properties are needed for 316L stainless steel (and braze joint material) and 2-1/4Cr-1Mo at specific PC-MHR service temperatures. The effect of helium impurities or water on 1/2Cr-1/2Mo steel precooler and intercooler materials is not expected to be a concern because of the low temperature of operation. In addition, a review of candidate materials resistant to higher than anticipated temperatures for the SHE shroud/support components can be performed.

5.4.4.5 Component development

The "component test development" describes development requirements in the following areas:

- Fuel handling system
- Neutron control system
- Shutdown cooling system circulator
- Shutdown heat exchanger
- Reactor cavity cooling system
- Reactor internals and hot duct

Development in these areas mainly consists of subcomponent design verification testing and prototype testing with a significant focus on power conversion system components and assemblies. While HTTR cannot provide many of these developmental needs directly, one of the heat utilization systems planned under IAEA Coordinated Research Programme (CRP) is demonstration of Brayton cycle. Some of the power conversion system related developmental needs can be partly demonstrated using HTTR high temperature, high pressure helium.

Fuel handling system

The MHR fuel handling system (FHS) equipment has evolved from the technology developed for Peach Bottom and FSV. Newer automated machinery and simultaneous operations need component testing to verify mechanical, electrical and electronic hardware to meet plant performance and reliability requirements. These include fuel handling machine (FHM) and fuel transfer cask (FTC) operations with simulated core elements in helium, element hoist and grapple assembly robot (EHGA) accuracy and endurance, fuel handling equipment positioner (FHEP) axes kinematics and accuracy, fuel handling equipment support structure (FHESS) demonstration in air with reactor isolation valves and inflatable seals.

Neutron control system

Neutron control system (NCS): NCS component tests are needed to ensure reliable and repeatable operations and include design verification of control rod drives (CRDs), reserve shutdown control equipment (RSCE), In-core flux mapping units (IFMUs), and flow induced vibrations and seismic qualification of the neutron control assembly (NCA). While these tests may not be able to be conducted directly in HTTR, it can help test ceramic control rods developed for modular helium reactors under appropriate irradiation environment.

Turbomachine development

Turbomachine: The bulk of the turbomachine technology comes from the open cycle gas turbine industry. Although no known feasibility issues exist, several areas need design development and verification. An experimental investigation will be undertaken to determine the effect of fission product (e.g., silver and tellurium) plateout on the blade cast nickel-base alloy. Component verification tests include full-scale magnetic bearing assembly tests in helium including thrust, journal and catcher bearings with selected control system, power supply, instrumentation etc. to demonstrate overall system performance, full-size seal system tests to determine leakage rates and seal integrity, and flow distribution tests to characterize flow maldistributions in the compressors and turbine inlets and outlets. Verification tests on generator windings/insulations also need to be performed under temperatures and pressures experienced under steady state and cyclic reactor conditions including depressurization testing to determine the effect of sonic outgassing on insulation integrity. The power conversion vessel (PCV) instrumentation and electrical penetrations need to be tested under helium pressure and temperature to demonstrate structural integrity and insulation resistance. The heat utilization program currently being planned for HTTR under the IAEA Coordinated Research Programme can address some of these developmental needs for MHR.

SCS circulator development

SCS circulator: The approach taken in the development of the shutdown cooling system (SCS) Circulator will follow that for the British AGR circulators. Prototypical component testing will be performed to confirm SCS impeller aerodynamic forces and establish acoustic properties, and confirm life cycle integrity of the shutdown loop shutoff valve (SLSV). A prototype SCS circulator test needs to be performed in a helium high pressure test facility (HPTF) to provide final design confirmation. HTTR reliable circulator operation will be an asset to the developmental needs of SS circulator.

Recuperator development

Recuperator: The PCS recuperator is considered state of the art technology with little development necessary. Component testing is needed to verify the structural integrity of the recuperator brazed plate-fin type construction for high differential pressure loads, flow maldistribution tests to determine performance effectiveness, leakage tests to characterize "leakage signature" and leakage degradation, and heat transfer and pressure drop performance tests in air.

Precooler/intercooler development

Precooler/Intercooler: No major design uncertainties have been identified for the precooler and intercooler. The component service temperatures are low and the shell side (external) pressure is higher than the internal water pressure. Component testing is needed to verify fabrication and coiling methods for finned tubing and to develop a method for inspection and detection of tube wall flaws. Air flow tests will be conducted to determine inlet flow distributions, acoustic response, heat transfer, and flow induced vibration response.

Shutdown heat exchanger development

Shutdown heat exchanger (SHE): The SHE design will, to a large extent, be based on data from steam generator tests conducted on the NPR program including attenuation of flow maldistribution, flow induced vibrations, tube restraint device (TRD) development and fabrication and installation of large helical coils. Additional development and testing required include SHE inlet flow distribution tests, and shroud and seal performance tests.

Reactor cavity cooling system development

Reactor cavity cooling system (RCCS): Testing of the RCCS aims to verify analytically based performance predictions including heat transport phenomena. Single effects testing will be performed to determine the effective conductivity of the graphite core, buoyancy-induced fluid mixing in the enclosures along the core and emissivities of metal surfaces including the RCCS panels, reactor vessel and metallic reactor internals. Final verification of the RCCS concept is accomplished by a combination of analysis and performance testing of the gas loaded heat pipe heat exchanger arrangement, and the RCCS natural draft stack. Extensive testing planned at the HTTR on the vessel cooling system (VCS) will definitely help the developmental needs of RCCS for modular helium reactors.

Reactor internals and hot duct development

Reactor internals and hot duct: reactor internals and hot duct component tests are conducted to verify their structural integrity during normal operation and postulated accidents. The selected component testing is substantially based on the experience from test programs conducted on graphite core elements and assemblies, control rods, and the thermal barrier for prior HTGR concepts. Additional test requirements for the MHR internals and hot duct mainly account for changes in component configuration and loading conditions and include tests to determine core column vibration data, core support strength, control rod vibration characteristics and strength, control rod shock absorber effectiveness, assessment of core column flow induced vibrations, and to confirm integrity of hot duct thermal barrier cover plates and selected insulation material. HTTR operation can clearly help in satisfying the developmental needs of reactor internals and hot duct for modular helium reactors.

5.4.4.6 Instrumentation and control development

The “instrumentation and control development” describes the development of instrumentation and controls for neutron detection, helium mass flow, reactor protection and fission product monitoring. Although the instrumentation and control design is substantially based on FSV, additional development is required to account for changes in physical configuration and performance requirements. Specific development tests include verification of neutron detectors and cabling in the reactor environment, helium mass flow and core inlet/outlet helium temperature instrumentation, conduction cool down temperature instrumentation, and plate out probe performance. The HTTR is an excellent environment to test the MHR instrumentation and control system components.

5.4.4.7 PCS integrated test

The power conversion system integrated test development describes the need for integral testing of the power conversion vessel (PCV) components to verify the key design features prior to operation of the first reactor module. Prototypic PCV component assemblies will be tested in the first module power conversion vessel with a hot helium supply from a natural gas fired heater to simulate steady state module operation and key transient conditions specified in the plant duty cycle. The major design elements to be addressed include primary coolant bypass, remote removal and replacement of PCV components, the dynamic response and structural integrity of the turbine assembly, effects of sound pressure loads from the turbomachinery, and the uncertainty in predicting component performance (e.g., relative thermal expansion, stresses) when assembled and operated as a system. While the HTTR cannot be used for PCS internal tests, the heat utilization test with the gas-turbine concept currently being evaluated can provide invaluable guidance to the PCS integrated test of modular helium reactors.

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Chapter 6

ACHIEVEMENTS OF OTHER HIGH TEMPERATURE ENDOTHERMIC TECHNOLOGIES AND SUPPORTING ACTIVITIES

This chapter includes a discussion of additional heat utilization systems which were evaluated within the scope of the CRP and judged to either require further technological development prior to consideration for future coupling with the HTTR, or were found to not be feasible for application with the HTTR.

6.1 THERMOCHEMICAL WATER SPLITTING FOR HYDROGEN PRODUCTION; THE IODINE-SULFUR (IS) PROCESS

6.1.1 Major process parameters

Figure 6.1 shows the basic flow diagram and the representative process temperatures of the IS process. The Bunsen reaction produces hydriodic acid and sulfuric acid. The acids are separated by liquid-liquid phase separation phenomena appearing in the presence of an excess amount of iodine. Main components of the heavier and the lighter solutions are HI, I_2, H_2O and H_2SO_4, H_2O, respectively. As for the heavier solution, the HIx solution, it is purified to remove unseparated sulfur compounds. Then, HI is separated from the purified HIx solution by distillation and thermally decomposed. As for the lighter solution, sulfuric acid, it is also purified, concentrated and decomposed.

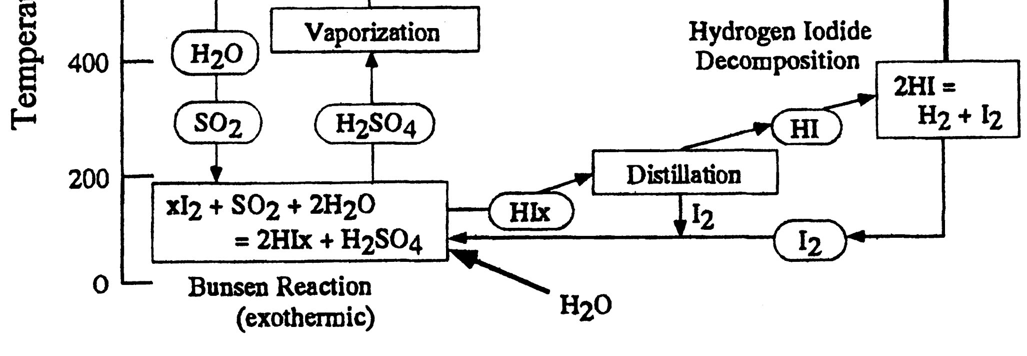


FIG. 6.1. Scheme of the "IS" process.

Relative to the processing of HI, there are presently several candidates and the selection remains to be made in the future. This should be carried out by taking the process efficiency, the availability and the cost of the construction materials into consideration. As for the separation of HI from the HIx solution, in order to reduce the vaporization of water, an extractive distillation using phosphoric acid [1] and a reactive distillation under pressurized condition [3] have been proposed. JAERI is pursuing an application of electrodialysis as outlined in section 6.1.2. The HI decomposition reaction can be carried out in the liquid phase using solid or metal ion catalysts [1,4], or in the gas phase using solid catalysts [1,5]. Since the equilibrium conversion of the reaction is rather low (ca. 20% at 400^{o}C), an application of a membrane reactor is under study at JAERI as outlined in section 6.1.2.

Therefore, process conditions for a pilot scale test of IS process has not yet been established. However, an engineering flowsheet presented by General Atomic Co. (GA) [5] should be a basis of the future study. In this flowsheet, the extractive distillation of the HIx solution and the liquid phase decomposition of HI have been adopted. The representative process conditions may be summarized as follows:

(a) Bunsen reaction step
    Bunsen Reaction: SO_2 conversion; 100%, 368 K, 1.85 bar
    Liquid-Liquid phase separation: 368 K, 1.85 bar

(b) HI decomposition step
    Extractive distillation: top; 401 K, feed; 418 K, bottom; 431 K, 1 bar
    HI decomposition reaction: HI conversion; 52%, 393 K, 50 bar

(c) H_2SO_4 decomposition step
    Concentration: from 57wt% to 98wt%, 393 – 634 K, 2 bar
    Vaporization: 686 K, 8.6 bar
    SO_3 decomposition reaction: SO_3 conversion; 73%, 1144K, 5.2 bar

In the GA flowsheet, an HTGR with the primary helium (He) loop of 49 bar and 1255 – 773 K, was assumed as the heat source. The nuclear heat is transferred to three secondary helium loops. The high temperature loop provides heat for the H_2SO_4 decomposition step. The recovered heat from the step was used in the HI decomposition step. The heat of the intermediate and the low temperature loops are used mainly to generate power for the HI decomposition step. The power is required for steam recompression in the concentration operation of phosphoric acid that is used as an extracting agent for the distillation. Figure 6.2 provides a summary of the energy flow [6].

The thermal efficiency of hydrogen production, defined as the ratio of the higher heating value of hydrogen and the net heat input to the process, was reported to be 47%.

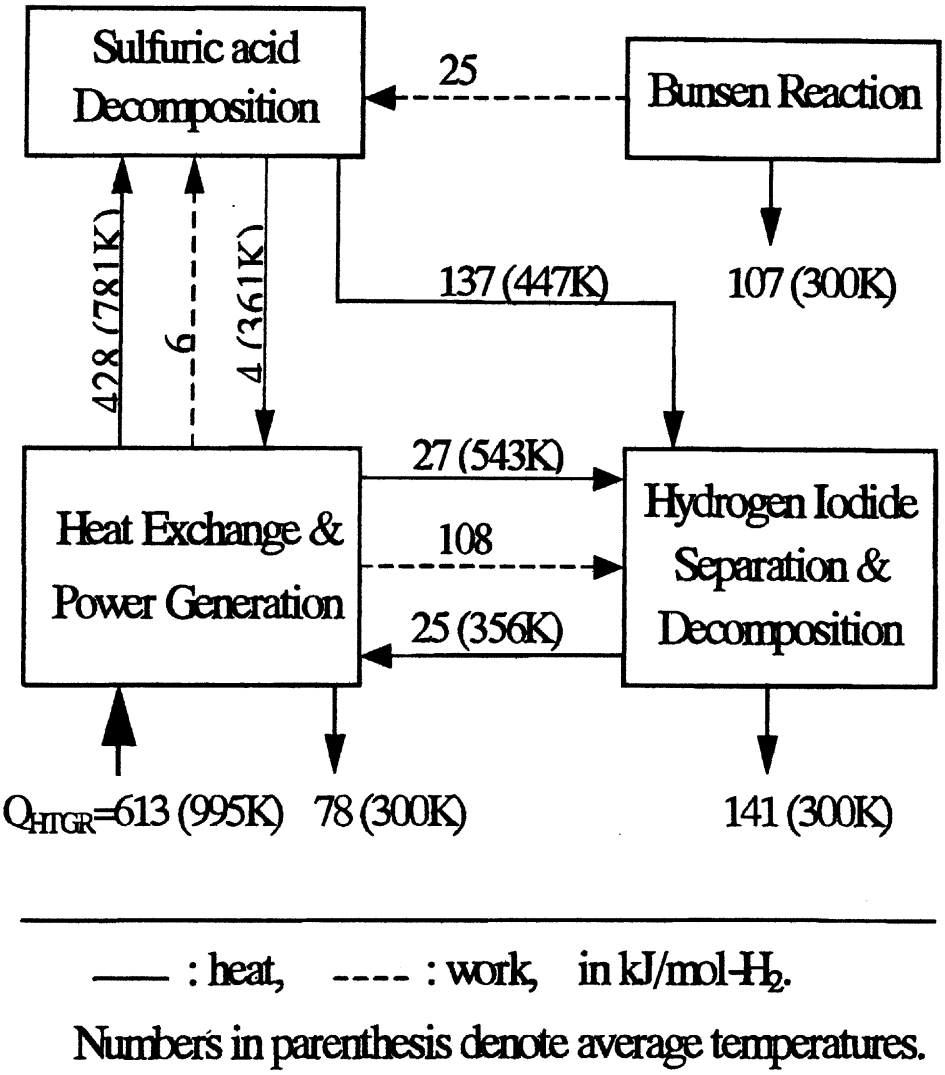


FIG. 6.2. Energy flow in the GA flowsheet [6].

Several modifications of the GA flowsheet have been proposed concerning the H_2SO_4 decomposition step. A group lead by Prof. Knoche at RWTH Aachen presented a modified flowsheet aimed at improving the energy balance [7], where SO_3 decomposition was carried out at 1120 K and 20 bar. A group of Prof. Bilgen at University of Montreal presented a scheme that could avoid a critical problem of the selection of the materials of construction for the corrosive environment [8]. In this modification, using oxygen as a heat carrier, the vaporization of sulfuric acid is carried out in a conventional adiabatic reactor. The SO_3 decomposition is carried out at 1034K and 9 bar with the conversion of 46%.

6.1.2 Achievements — laboratory scale

JAERI has been conducting a study on the IS process in three research fields: the laboratory-scale demonstration of continuous hydrogen production, the modification of the HI processing scheme, and the materials of construction. Recent progress on the former two is described in this section.

The laboratory-scale demonstration study aims to show the chemical feasibility of continuous hydrogen production with recycling of the process materials except water, which is the very specific characteristic of thermochemical process. An experimental apparatus made of glass and Teflon has been used in the study, in which the principal unit operations were included as shown in Figure 6.3. As for the HI decomposition step, an atmospheric pressure distillation of HIx solution and a gas phase thermal decomposition using a Pt catalyst, were adopted by use of conventional methods. The main chemical and technical target of the study was on the Bunsen reaction step. In order to secure the stable liquid-liquid phase separated state, it was required to accumulate knowledge on the reaction and the phase equilibria [2,9]. Owing to the knowledge and also to a newly devised pump for quantitative transportation of melted iodine, the continuous hydrogen production could be carried out for ca. 50 hours with the hydrogen production rate of 1 liter per hour. Stable rate of hydrogen production, stoichiometric production of hydrogen and oxygen, and stability of the composition of the phase-separated solutions were successfully demonstrated in the experiments [10,11,12]. At present, the acquisition of thermochemical data on the Bunsen reaction step is continuing [13,14] and a demonstration under more efficient process condition is scheduled.

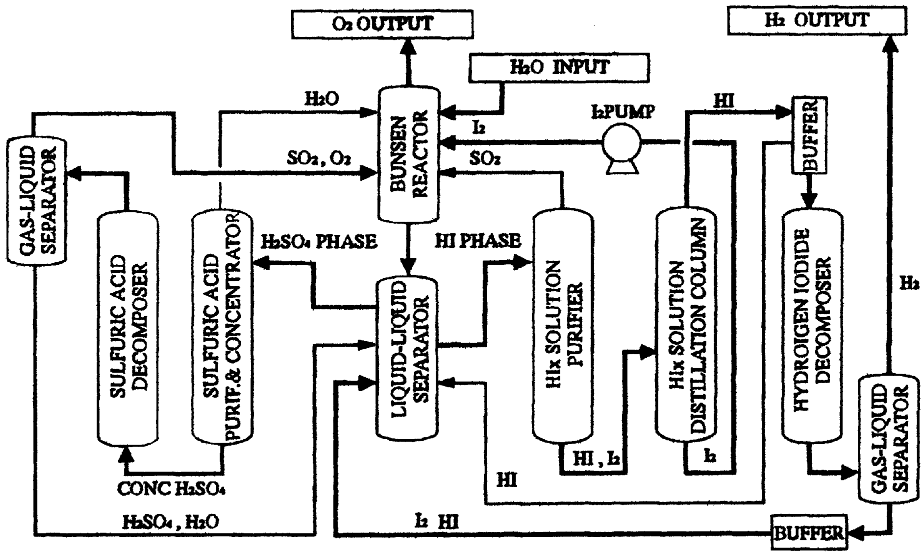


FIG. 6.3. Flow diagram of a laboratory scale demonstration experiment of the IS process.

In a study on the modification of the HI processing scheme, an application of innovative membrane technologies has been pursued. Figure 6.4 is a flow diagram of the proposed scheme. In this scheme, first, an electrodialysis is applied to concentrate the HIx solution fed from the Bunsen reaction step. By concentrating it over quasi-azeotropic composition, it is possible to obtain pure HI even with the atmospheric distillation [15]. The following performance is the target of the electrodialysis: (1) concentration of the feed solution with HI molality of ca.10 mol/kg and I_2 molality of ca.40 mol/kg, (2) operation temperature of ca. 100°C, (3) unit cell voltage lower than 0.5V. So far, promising electrodialysis behaviors were found such that the water permeation through the ion exchange membranes was strongly prohibited in the presence of iodine due to the iodine adsorption on the membrane [16], and a novel electrodialysis using only cation exchange membranes was effective at elevated temperatures [17].

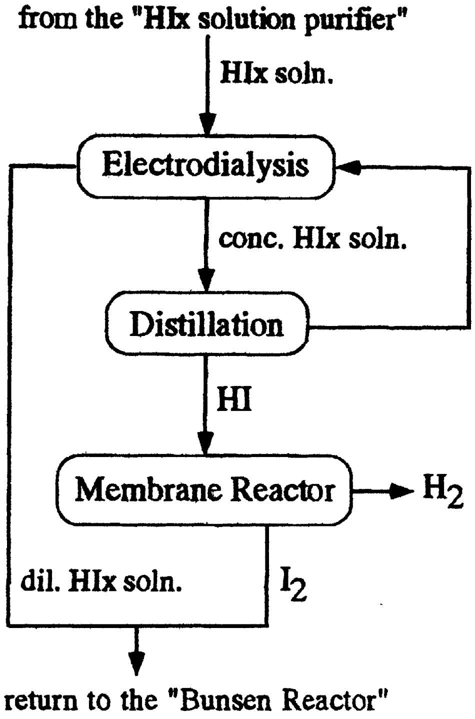


FIG. 6.4. Flow diagram of the proposed HI processing scheme.

The second theme on the HI processing scheme is a membrane reactor equipped with hydrogen permselective membranes to enhance the one-pass conversion of HI. In the membrane reactor, a novel membrane is required for hydrogen separation from H_2-HI-I_2(-H_2O) gaseous mixture at 300-500°C. Therefore, focusing on thermal and chemical stability of ceramics, a fabrication of hydrogen permselective ceramic membranes has been studied. Chemical vapor deposition of silica on porous alumina substrate was applied for the fabrication [18,19,20]. The modified membranes showed a permeance ratio of H_2 and HI higher than 150 [20], and the H_2 permeance of the order of 10^{-7} mol/(Pa m^2 s). The H_2 permeance was stable in a 24-hours separation experiment of H_2-HI-H_2O gaseous mixture at 450°C [21]. A preliminary simulation of the membrane reactor based on these figures suggested that the maximum attainable one-pass conversion of HI exceeded 90%, whereas that of equilibrium was ca. 20%.

6.1.3 Industrial scale up — materials development

The IS process uses rather corrosive chemicals such as sulfuric acid, hydriodic acid and iodine. Therefore, careful consideration is required on the materials of construction for the scaled-up plant. GA presented results of corrosion tests carried out using commercially available materials in the representative process environments of the SO_3 decomposition [22] and of the HIx solution [23]. Concerning the sulfuric acid decomposition step, corrosion data were also presented by Westinghouse Co. [24] and by JRC Ispra establishment [25]. As for the decomposition environment of gaseous hydrogen iodide, the former National Chemical Laboratory for Industry, Japan, published a report on the corrosion resistance of various metals and ceramics [26]. JAERI also carried out a preliminary screening test of materials in the representative process conditions of the IS process [2].

Based on these studies, JAERI has initiated a detailed evaluation of the candidate materials, and, if necessary, a development of new materials. So far, the work has concentrated on the environments of the sulfuric acid decomposition step. In the boiling of sulfuric acid, one of the most corrosive environments of the IS process, corrosion resistance of the candidates, Fe-Si alloys and ceramics, have been examined deeply.

As for Fe-Si alloys, the critical Si content for the passivation in boiling 95wt% sulfuric acid was found to lie in the range of 9-10 wt% Si [27]. In the alloys with higher Si content, the corrosion rate drastically decreased and, above 12wt% Si, no evidence of corrosion was detected even after an immersion test of 300 hrs. The corrosion resistance was due to a formation of compact passive film composed of SiO_2 and SiO. However, the alloys with high Si content are brittle and thus it is desired to add a certain amount of ductility for the concerned service. For this purpose, a hybrid material composed of a ductile metal substrate and a corrosion resistant surface layer was examined [28]. The material was test-fabricated by a chemical vapor deposition (CVD) using Fe-3wt%Si as the substrate. Using $SiCl_4$ as the reactant gas, a Si-enriched compositionally graded layer was formed on the surface of the substrate. The Si content on the top surface was 14 wt% and the thickness of the layer was ca. 70 μm. An immersion test in boiling 95 % sulfuric acid for 300 hrs demonstrated good corrosion resistance of the surface layer. However, a microscopic analysis of the surface layer identified corrosion grooves developed in micro-cracks, which were formed during the CVD treatment. Improvement of the CVD condition is required to prevent the formation of cracks.

As for ceramics, Al_2O_3, ZrO_2, SiC, Si_3N_4, and SiSiC have been examined focusing on the effects of environment on their chemical stability and also on the mechanical properties [29, 30, 31, 32]. In the former four materials, binding components were added in their fabrication while in SiSiC, Si worked as the binder. In 1000 hrs corrosion tests in boiling 95% sulfuric acid, no significant weight change was detected of the ceramics except ZrO_2 that was severely corroded. However, dissolution of binding components was observed in Al_2O_3, SiC and Si_3N_4. In accordance with this observation, the bending strength of these ceramics changed by increasing the immersion time. Here, it should be noted that the SiC alone showed an increase of the bending strength due to the formation of the protective surface layer composed of amorphous silica. The bending strength of SiSiC did not change. These results confirmed the excellent corrosion resistance of SiC and SiSiC, and identified the origin of the corrosion resistance. These ceramics can be used in equipment such as valves and pumps, and also used as lining materials for the reactors and pipes. Other ceramics including Si_3N_4 are unsuitable for the concerned service due to weight loss and/or the decrease of mechanical strength.

The SO_3 decomposition reaction constitutes the highest temperature environment in the IS process. The corrosion behavior of the candidate materials for this environment are under careful investigation including the effect on important mechanical properties such as creep behavior.

6.1.4 R&D programme

Figure 6.5 provides an outline of JAERI's R&D programme on the IS process. For the coming years, the present R&D concerning the closed-cycle process operation, the modification of HI processing scheme and the materials of construction will be continued.

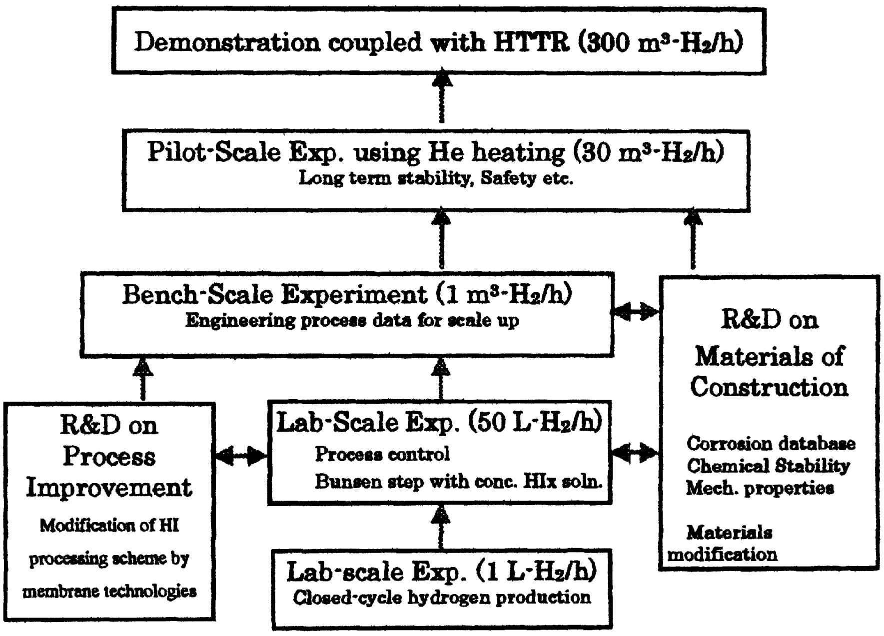


FIG. 6.5. R&D plan for the IS process.

As for the first theme, the next target is a demonstration of closed-cycle hydrogen production using liquid-liquid phase-separated solutions of higher iodine concentration. In the former demonstration with a hydrogen production rate of 1-liter per hour, the iodine concentration was equal to the solubility limit at 0°C, and the mutual solubility of HI and H_2SO_4 in the separated solutions was about 10%. By increasing the iodine concentration, better separation and higher acid concentration are expected, which results in lowering the heat duties in the following purification and concentration of the acids. Actually, with the iodine concentration equal to the solubility limit at 95°C, the mutual solubility is lower than 1%. The technical target lies in controlling the process condition while handling the corrosive and heavy HIx solution. For the experimental investigation, a scaled-up glass plant is under construction.

Concerning the modification of the HI processing scheme, the following basic studies are further required to clarify their prospects. As for the electrodialysis of the HIx solution, an experimental verification under the concerned process condition should be carried out. As for the hydrogen separation membranes for the membrane reactor, the remaining targets are the increase of the hydrogen permeance and verification of long term stability of the modified layer. R&D for scale up will begin after these targets are cleared.

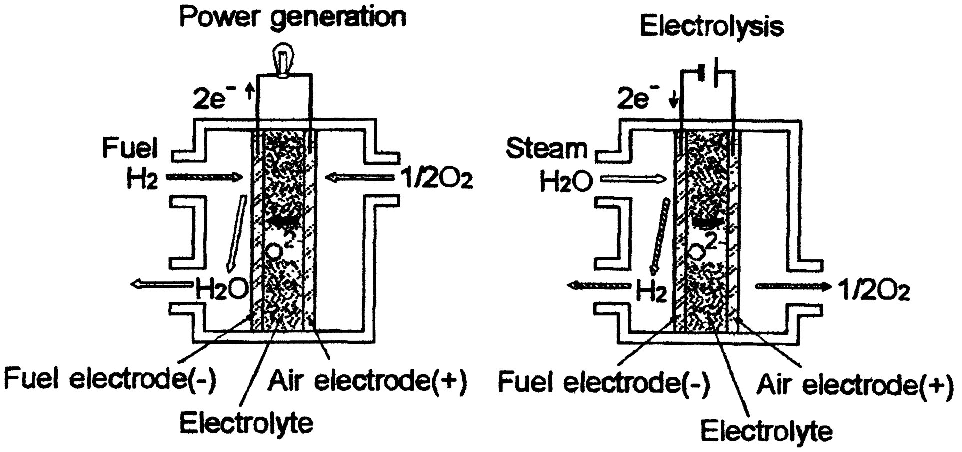
The results of the above-mentioned studies will be utilized for the detailed evaluation of the process performance. Then, the scaled-up experiments will be carried out stepwise aiming at demonstration of the process coupled with the HTTR.

Study on the materials of construction is divided into two categories and will be carried out in parallel with the process development. One is to acquire a corrosion database, and the other is concerned with development of materials that will be carried out by surface modification of commercial materials. The results of the former study under atmospheric pressure will be reflected in the bench scale experiment. The corrosion data under pressurized conditions and most of the results on the materials modification will be utilized and tested in the pilot scale experiment.

6.2 HYDROGEN PRODUCTION BY HIGH TEMPERATURE ELECTROLYSIS OF STEAM

The high-temperature electrolysis of steam (HTES) using ceramic electrolysis cells is one of the advanced technologies of the hydrogen production process. The HTES is a reverse reaction of the Solid-Oxide Fuel Cell (SOFC) which is presently being vigorously developed around the world. The latest technology of the SOFC (i.e. the electrolysis cell) can be applied to the HTES. From the viewpoint of the energy demand, the HTES could potentially suppress the electric energy required to decompose steam to be much lower than that of water electrolysis. Figure 1 shows the energy demand for the water and steam electrolysis. The total energy demand (ΔH) is the sum of the Gibbs energy (ΔG) and the heat energy (TΔS). The electric energy demand, ΔG, decreases with increasing temperature as shown in Figure 6.6; the ratio of ΔG to ΔH is about 93% at 100°C and about 70% at 1000°C.

The HTES, however, is at a very early stage of technology development, and thus, it is necessary to take extensive efforts in order to make the HTES feasible. As a first step, laboratory-scale experiments are being carried out to examine the effectiveness of the HTES for hydrogen production and to improve the HTES technology. This report presents typical experimental results obtained using practical electrolysis cells.



	Power generation	Electrolysis
at fuel electrode	$H_2+O^{2-} \rightarrow H_2O+2e^-$	$H_2O+2e^- \rightarrow H_2+O^{2-}$
at air electrode	$1/2O_2+2e^- \rightarrow O^{2-}$	$O^{2-} \rightarrow 2e^-+1/2O_2$
Total reaction	$H_2+1/2O_2 \rightarrow H_2O$	$H_2O \rightarrow H_2+1/2O_2$

FIG. 6.6. Principle of high temperature electrolysis of steam (Reverse reaction of the solid oxide fuel cell).

6.2.1 Experimental results obtained with tubular cells

6.2.1.1 Test apparatus and test conditions for electrolysis tube

Figure 6.7 shows a structural drawing of an electrolysis tube, and Photo 6.1 an outer view of the tube. The electrolysis tube was composed of 12 electrolysis cells of 19 mm in effective length, labeled as “the banded-cell structure”. Each cell was connected in series electrically with a thin layer of interconnection working as an electric conductor. An electrolyte of the cell was made of zirconia stabilized with 8mol% yttria (YSZ), which is a popular material in the SOFC. The electrolyte layer was sandwiched between a porous cathode layer made of a

Ni cermet (fuel electrode) and an anode layer made of $LaCoO_3$ (air electrode). At both ends of the electrolysis tube, copper layers were coated to work as electric terminals, which were connected with thin layers of the electric conductor. These layers were formed on a porous calcia-stabilized ZrO_2 (CSZ) tube (a support tube or a ceramic tube) of 22 mm in outer diameter and 3 mm in thickness by using a plasma spraying method. The thickness of each layer was in the range of 0.1mm to 0.25 mm.

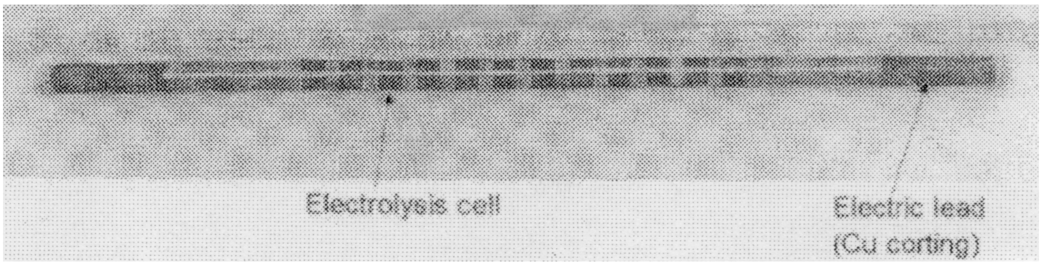


Photo 6.1. Outer view of the electrolysis tube.

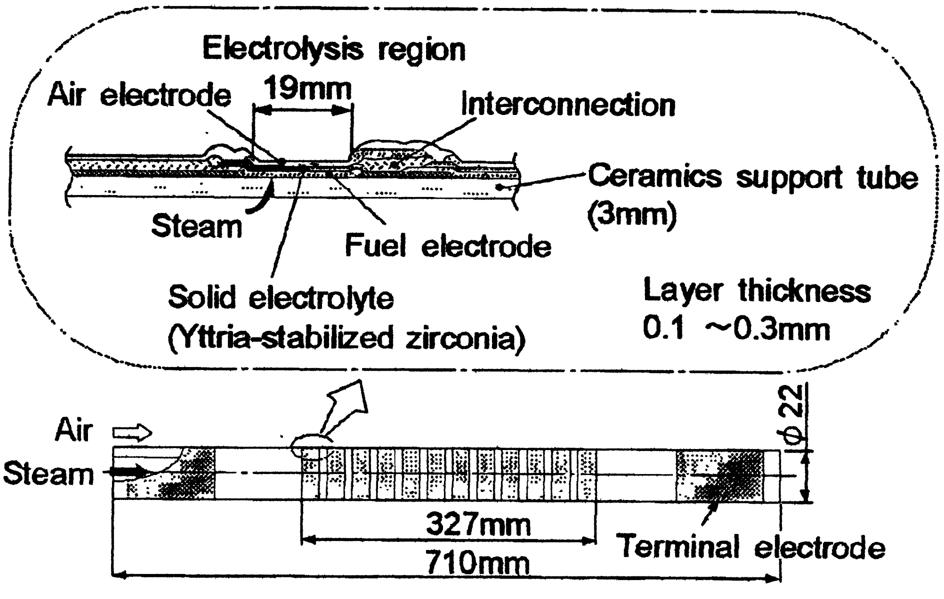


FIG. 6.7. Schematic drawing of the electrolysis tube (banded-cell structure supported by ceramic porous tube).

In the experiments, the electrolysis tube was installed in an electric furnace to control the electrolysis temperature. Steam was mixed with argon carrier gas from gas cylinders with a humidifier, and was supplied to the fuel electrode inside of the electrolysis tube. Steam concentration was detected by dew point monitors both at the inlet and the outlet of the electrolysis tube. Dry air from an air compressor was supplied to the air electrode outside of the electrolysis tube in order not to decompose the anode compound of $LaCoO_3$ under low partial oxygen pressure. The electric power necessary for the electrolysis was applied using a direct-current (DC) power supply through platinum (Pt) wires welded on the copper layers. The hydrogen concentration was measured by a gas chromatograph at the outlet of the electrolysis tube. Electrolysis voltage and current were also measured.

Experiments were carried out under the mixed gas temperatures of 850°C, 900°C, and 950°C. Other test conditions were as follows:

Argon flow rate	2.2 Ndm^3/min
Dew point at the inlet of the electrolysis tube	40~56°C
Steam content at the inlet of the electrolysis tube	0.13~0.32 g/min
Air flow rate	4~5 Ndm^3/min (dew point < -20°C)
Inlet pressure	0.11 MPa (1.1 bar(abs))

In the start-up and shut-down of the electric furnace, an increasing and decreasing velocity of the furnace temperature was set below 20°C /h in order not to generate a large difference of the thermal expansion among the thin layers and the support tube. Before applying electric power to the tube, the cathode material (the Ni cermet) was reduced with hydrogen mixed with an argon carrier gas for several hours in order to work as an electrode.

6.2.1.2 Test results obtained by electrolysis tube

Figure 6.8 shows a relationship between the current and the hydrogen production densities. The hydrogen production density increased with the current density and the electrolysis temperature. The maximum hydrogen production density was recorded at 25Nml/cm^2h (the hydrogen production rate of 3.9Ndm^3/h) under an applied power of 22.3W and the electrolysis temperature of 850°C, 28Nml/cm^2h (4.4Ndm^3/h) under 22.5W and 900°C, and 44Nml/cm^2h (7.0Ndm^3/h) under 26.8W and 950°C, respectively. Apparent electric resistances including resistances of electric leads and interconnections were around 11Ω at 850°C and 900°C, and around 9Ω at 950°C.

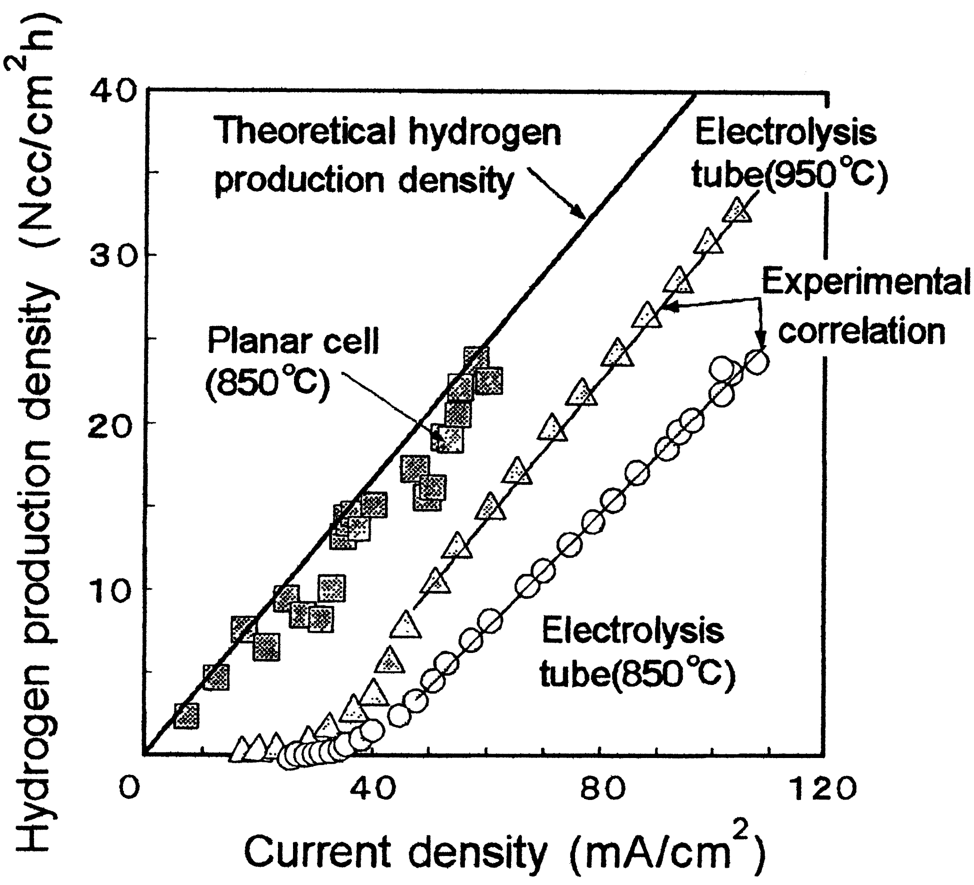


FIG. 6.8. Relationship between hydrogen production and current densities.

It was seen that the hydrogen production density Q_{H2} [Nml/cm^{2}h] had a linear relation with the current density i_d [mA/cm^2] in a range of more than 45mA/cm^2. The following experimental correlation [33] is shown in the figure:

$$Q_{H2} = 984.7 - 1.722Te + 7.427x10^{-4}Te^2 - (6.744 - 1.136x10^{-2}Te + 4.502x10^{-6}Te^2)\ i_d$$

where Te is the electrolysis temperature [K]. This equation is valid for more than 45mA/cm^2. The hydrogen production density had no linear relation in the range of less than 45mA/cm^2. This was because of a leakage of current which could not contribute to the steam decomposition.

In the case of 7.0Ndm^3/h at 950°C, an energy efficiency (a quotient of the combustion heat of generated hydrogen / the applied DC power) was only about 80%. This low energy efficiency could be caused by high electric resistance losses (Ohmic losses) at interconnections and electric lead layers. On the other hand, a steam conversion rate (an efficiency of steam utilization for the electrolysis) had a very low value of less than 40%. This was probably because that the steam could not permeate smoothly through the support tube to the fuel electrode (cathode), since the support tube had a rather low porosity of around 38%. If the steam reaches the cathode sufficiently, the hydrogen production density could increase more than the presently observed results. A self-supporting cell structure without the support tube, therefore, was considered to be much better in the HTES than the banded-cell structure supported by the ceramic tube.

After one thermal cycle test ranging from room temperature up to 950°C, it was observed that a large part of the layers of the air electrode (anode) peeled off from the electrolyte layers as shown in Photo 6.2. As a result, the durability of the cell against thermal cycles was found to be the key issue of the HTES technology. The plasma spraying method should be improved so as to raise an adhesion of the anode layer on the electrolyte layer against the thermal stress generated by the thermal expansion difference between these layers.

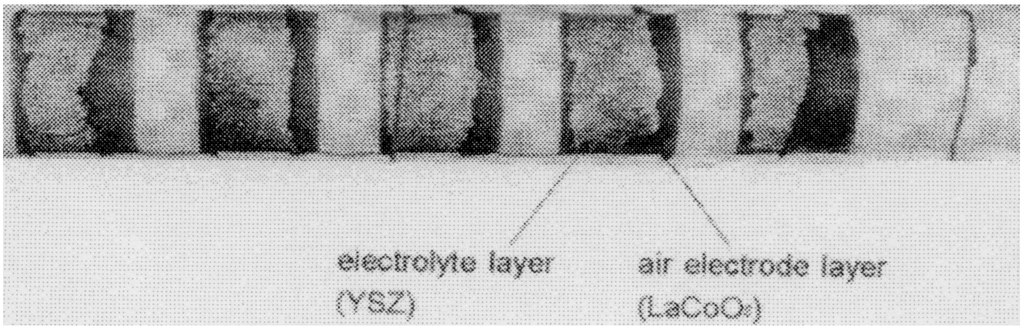


Photo 6.2. Outer view of the supported electrolysis tube after one thermal cycle (Air electrode layers were separated from the electrolyte layers).

6.2.2. Experimental results obtained with planar cell

6.2.2.1.1 Structure of self supporting planner cell

On the basis of test results described above, JAERI had tried to fabricate a self-supporting electrolysis tube. Its yield, however, was very low, and the fabrication cost was much higher than that of the banded-cell tube, because it was very difficult to manufacture electrolysis cylinders of 0.3mm thick and to connect segmented cells in series with gastight interconnections.

JAERI then focused on the planar cell, particularly a self-supporting planar cell, from the viewpoint of the mass production of the cell. Quality control in the course of its production was determined to be much easier than the banded-cell tube, and the production technology has progressed rapidly under the SOFC development. The production cost of the self-supporting planar cell was 1/7 or less lower than that of the banded-cell tube and will be reduced further by mass production. In addition, since the volume of an electrolysis reactor using the planar cells can be reduced much lower than that using the electrolysis tubes, the initial plant cost will decrease and maintenance procedures will be simplified.

Figure 6.9 shows a structural drawing of the self-supporting planar cell made by Fuji Electric Co. The cell was composed of an electrolyte plate of YSZ and thin layers of porous electrodes. The YSZ plate was a 100 mm square plate with a thickness of 0.3 mm, and the electrodes were coated on the area of 80 × 80 mm with a thickness of less than 0.03 mm. Material used for the fuel electrode (cathode) was the Ni cermet, and the air electrode was a strontium-doped $LaMnO_3$ (anode) which had better performance on thermal expansion than that of $LaCoO_3$ used in the electrolysis tube.

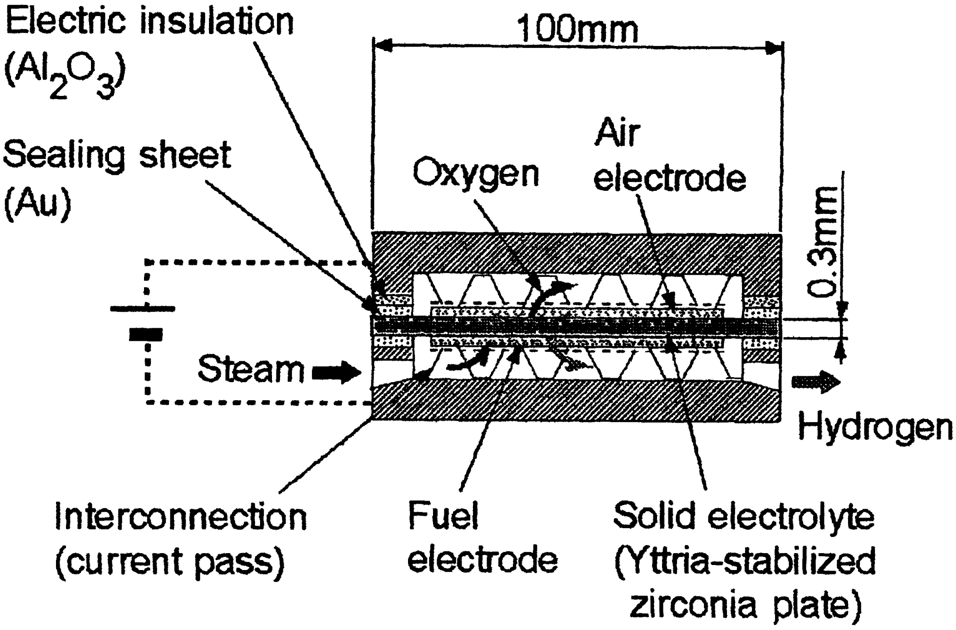


FIG. 6.9. Schematic drawing of self supporting planar cell.

The cell was sandwiched with metal housings which had a metal rod for an electric lead, and inlet and outlet piping for gases, which were made of SUS-310S. A Platinum (Pt) sheet of 0.1 mm thick was welded on the inner surface of each housing opposite to the electrodes of the cell in order to keep a good electric path under high temperature conditions. The wavy electric lead plate made of a Pt mesh was installed in each electrode compartment, which also worked as the support of the cell against the pressure difference between electrodes. The DC power for the steam electrolysis was supplied through the metal rods, housings, Pt sheets and wavy electric lead plates to the cell. Compression seals by using gold (Au) sheets were done at the edge of the cell plate: the compression load was up to 20kg. Then, an Al_2O_3 sheet of 0.3mm thick was inserted between the cell plate and the housing in the anode side in order to prevent current leakage through the cell plate edge.

The cell coupled with housings was installed in an electric furnace as shown in Photo 6.3. Electrolysis steam was supplied to the cathode compartment, which was mixed with argon and hydrogen gases through a steam generator instead of the humidifier used for the former experiments with the electrolysis tube. Argon gas was the carrier gas of the steam, and hydrogen, a reduced gas, was used to keep Ni of the cathode material from oxidation. Dry air was supplied to the anode compartment. Measurements and operation procedures were the same as those described previously.

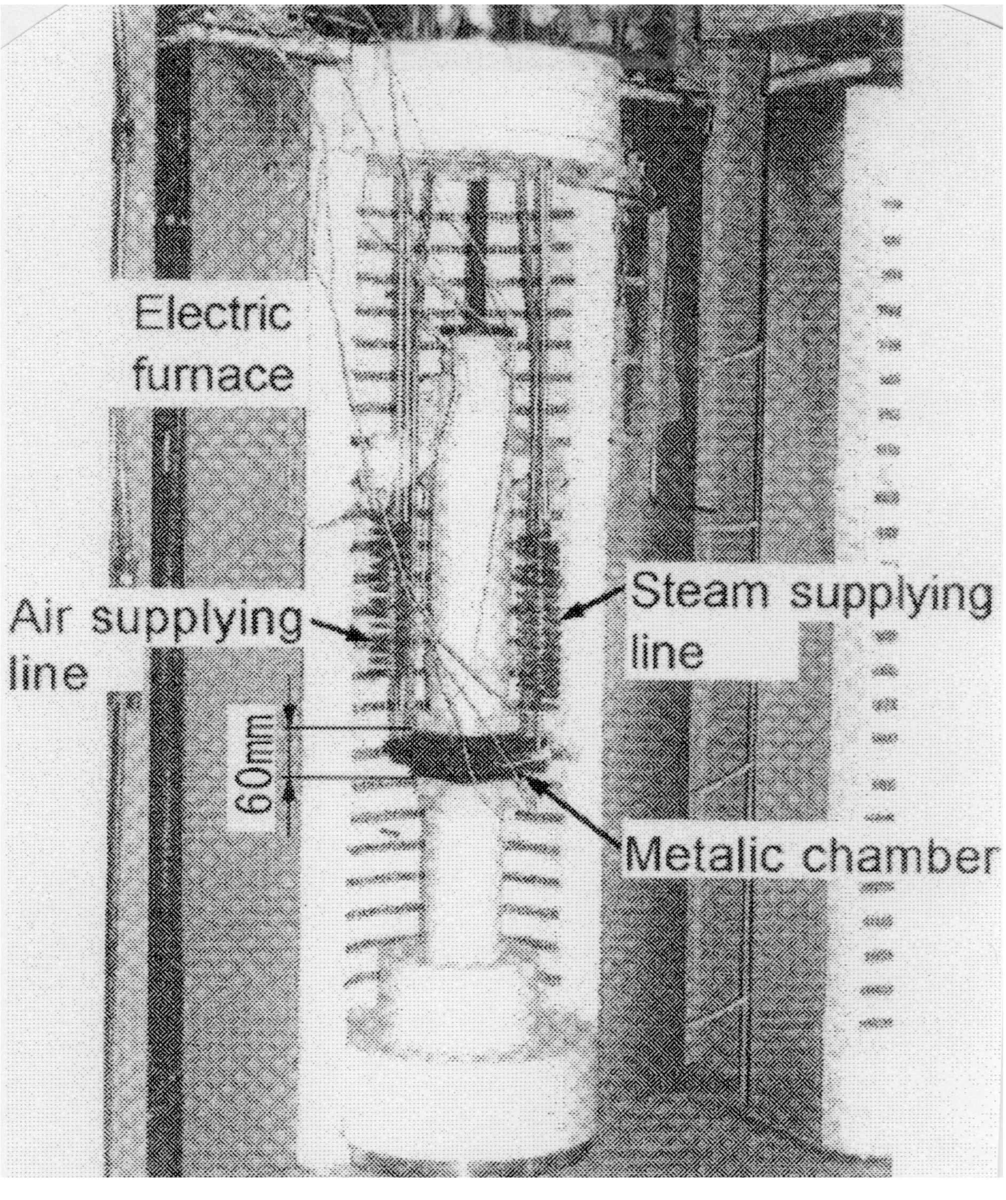


Photo 6.3. Test apparatus for the planar cell.

6.2.2.2. Experimental results obtained with planar cell

Experiments were carried out under the following conditions:

Electrolysis temperature	850°C
Inlet gas flow rate	0.2 Ndm^3/min (argon gas) 0.1 Ndm^3/min (H_2)
Steam flow rate	0.3 g/min
Air flow rate	1 Ndm^3/min (dew point is less than -20°C)
Inlet pressure	~0.14 MPa (~1.4 bar(abs))

In this condition, the maximum hydrogen production rate was 2.3Ndm^3/h at the applied power of 10.8W: applied voltage and current were 2.7V and 3.7A, and the apparent electric resistance was around 0.7Ω. The hydrogen production density was 24Nml/cm^2h and is about 3 times higher than that obtained with the electrolysis tube at 850°C as shown in Figure 6.8. The energy efficiency at 2.3Ndm^3/h of the hydrogen production rate was around 0.7, which was also higher than that obtained at 850°C when using the electrolysis tube.

Mixed gas leakage from the edge of the cell was less than 10% under a low pressure operation. It is necessary to develop high-temperature seals for the high-pressure operation (up to 4MPa) in order to incorporate the HTTR heat utilization system. After one thermal cycle test, we observed cracks as shown in Photo 6.4. This would be caused by a thermal expansion difference between the cell and the metallic housings.

To improve cell strength against the thermal expansion difference described above, a new type planar cell supported by a porous metallic plate was fabricated by Fuji Electric Co.

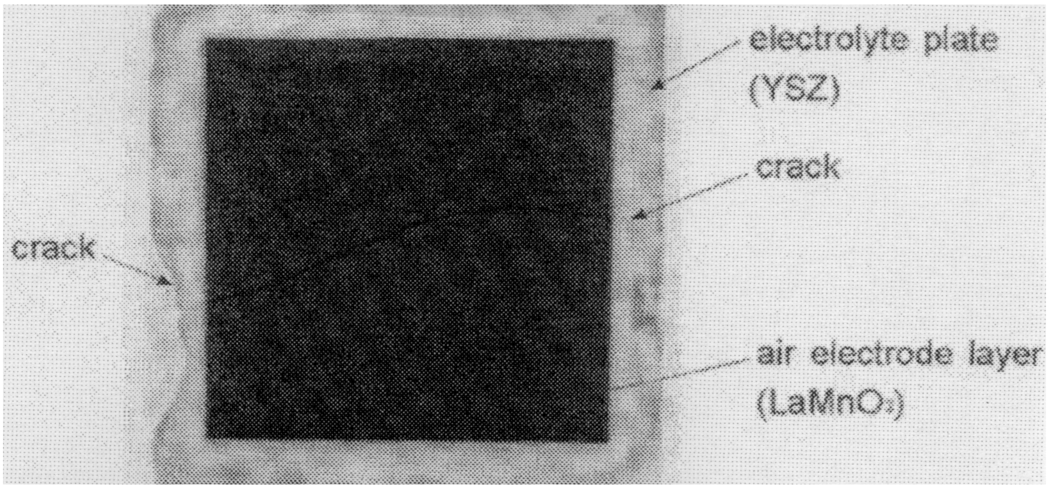


Photo 6.4. Outer view of supported electrolysis tube after one thermal cycle.

Figure 6.10 shows the schematic drawing of the supported planar cell. The cell was composed of the YSZ of the electrolyte and thin layers of porous electrodes made of the Ni cermet (fuel electrode) and the strontium-doped $LaMnO_3$ (air electrode), which was structured on a porous metallic plate made of NiCr serving as the electrode and the support of the cell. The electrolysis area was 90mm in effective diameter.

A preliminary test was conducted under 950°C of the electrolysis temperature. The maximum hydrogen production density was 33.6Nml/cm^{2}h at 146mA/cm^2. The energy efficiency was around 0.35, which was much lower than that of the electrolysis tube obtained at 950°C. However, this type of cell was very sound without any damage such as cracks after one thermal cycle test, though other cells were broken after only one thermal cycle.

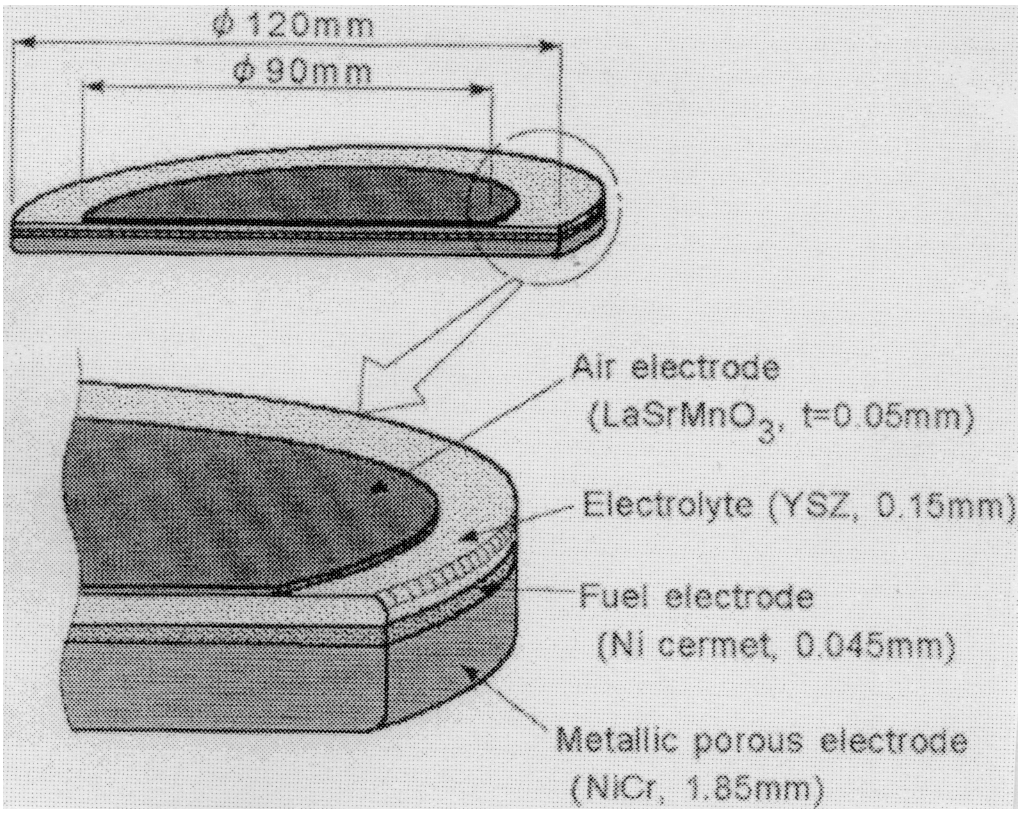


FIG. 6.10. Schematic drawing of supported planar cell.

6.2.3 Concluding remarks

The test results of the HTES have been introduced, which were carried out by using the electrolysis tube and the planar cells. The engineering problems of the HTES to make it feasible as a hydrogen production system were clarified from these test results, system. The main engineering problems are as follows:

- To increase the energy efficiency

  It is necessary to reduce the current leakage and ohmic loss of the cell by means of thinning electrolyte and electrode layers yet keeping their mechanical strength. For the planar cell, it is necessary to decrease the contact electric resistance between the electrode and the lead plate.

- To increase the cell durability against thermal cycles,

  It is necessary to select cell materials, especially electrode materials so as to lower the thermal expansion difference against the electrolyte material such as YSZ while still keeping high electron conductivity. Also, there is a need to improve the adhesion performance of the electrodes' layers to the electrolyte. Further efforts are required to improve the electrode coating techniques taking into consideration the yield and the coating cost for mass production. The improvements of the planar cell supported by the porous metallic plate would solve the durability problem.

Although more effort is needed to solve the above problems, resumption of research of the HTES is expected to broaden the field of nuclear heat utilization.

In acknowledgment, gratitude is expressed to Y. Miyamoto, the manager of the Department of the Advanced Nuclear Heat Technology, who has always supported the HTES. Gratitude is also expressed to F. Okamoto of Fuji Electric Co. for supporting the planar cell tests.

6.3 COAL CONVERSION

The fact that the world reserves of coal (hard and brown) significantly exceed the known resources of oil and natural gas makes it obvious that coal will play an important role in the world fuel and energy balance.

However, in view of a number of difficulties in using coal for power production associated with transportation problems, high cost of power plants using solid fuel and pollution of the environment, the consumption rate of coal decreased in the last 40–50 years, and was supplanted by oil and gas that are more convenient to use. The share of coal in the fuel and energy balance (FEB) of a number of large industrial countries was more than 60–70% back in 1920’s–1930’s, and presently it does not exceed 20–30%.

In view of the increasing deficit in oil and gas resources and reduction of their use, the structure of the FEB in many countries of the world will undergo changes in the direction of increasing the share of nuclear power and the extent of using coal even in the first half of the next century.

However, the use of solid fuel in the initial form will be hindered due to a number of important factors (economical, environmental, regional, etc.). Besides, different branches of industry have been developing by using gas and liquid fuels for many decades. Technically, many of the processes in metallurgy and chemical industries, as well as the use of fossil fuel in the power industry, transport and for household purposes were based on the use of gas, oil and oil refining products. The share of these kinds of fossil fuels in power consumption, for example, in the USA and Russia, averages about 70%.

This leads to the necessity to develop processes for conversion of coal into other high-quality fuels which are more convenient in use.

The physics fundamentals of coal gasification processes were developed in the 1920's–1930's, but their implementation in practice has been hindered for a long time by the availability in the market of cheap oil and gas. The purpose of the gasification processes is to convert solid fuel, primarily coal (brown and hard), including low-carbon content fuel (peat), into high-quality kinds of fuel.

Due to the progress made in practical implementation of the HTGR in recent years, a number of countries are pursuing research into using the high-potential thermal power produced by these reactors for gasification of brown coals, hard coals and shale. The research carried out in Germany, Russia, USA, China, Japan and other countries has demonstrated that reforming of coal using HTGR produced thermal power offers significant advantages over conventional gasification processes in terms of more efficient utilization of the available primary energy resources. This technique increases production efficiency allowing a nearly 40% reduction in coal consumption as compared to existing methods with the same output of synthesis gas This results in lower (by 20-30%) production costs of the final product and essentially improves the environmental situation.

Due to employment of inert helium coolant and graphite as the structural material of the core, an outlet temperature of ~ 950° is now attainable in HTGR. This temperature level suffices for efficient implementation of gasification processes.

Two coal gasification processes are presently being considered as applied to employment of thermal power produced in the HTGR:

- hydrogasification

$CH_4+H_2O = 3H_2 + CO - 60$kcal/mole;
$C + 2H_2 = CH_4 + 20.9$ kcal/mole

- steam gasification

$C + H_2O = H_2 + CO- 28$ kcal/mole

The specific feature of the gasification process based on the use of HTGR thermal power consists in external heat supply.

6.3.1 The lignite hydrogasification process

The lignite hydrogasification process consists in a reaction between coal and hydrogen that produces methane which can be used in different industries, in power production and for domestic purposes. Hydrogen needed for coal gasification is produced through catalytic steam conversion of methane followed by the stages of carbon oxide conversion and the removal of impurities.

This process is highly endothermic and requires heat to be supplied from a reactor. Helium at temperature of ~950°C is delivered to a reformer where it is cooled down to 750°. It then enters a steam generator where steam is produced for a steam turbine drive. From the steam generator, the helium at 350°C arrives at the reactor inlet. Part of the steam produced in the steam generator enters the steam reformer to accomplish the chemical process of steam conversion of methane. The steam reformer is made up of coaxial pipes, and its operation is based on the counterflow principle. In the presence of a catalyst, the steam conversion of methane commences at 600°C. Nickel oxide is used as the main active constituent of the catalyst. The most economically favorable reaction rate is attained at 800°C and can be increased by increasing the temperature. In order to have an economically and technically advisable industrial process of methane steam conversion, the helium temperature level at the reformer inlet and outlet should be 950 and 750°C, respectively. For reliable operation of the helium circulator and in-reactor systems, the temperature of the cold gas should not exceed 350°. The pressure in the primary circuit is 40 bar for the safety reasons.

In order to enhance the cycle efficiency, the mixture of methane and water/steam is heated in the recuperator by converted gas up to ~600°C prior to its supply to the steam reformer. While being cooled down to 500°C, this gas passes through several stages of CO_2 and H_2O cleaning, and separation into CH_4 and H_2. After that, hydrogen is directed to coal gasification. The hydrogasification process of lignite shown in Figure 6.11.

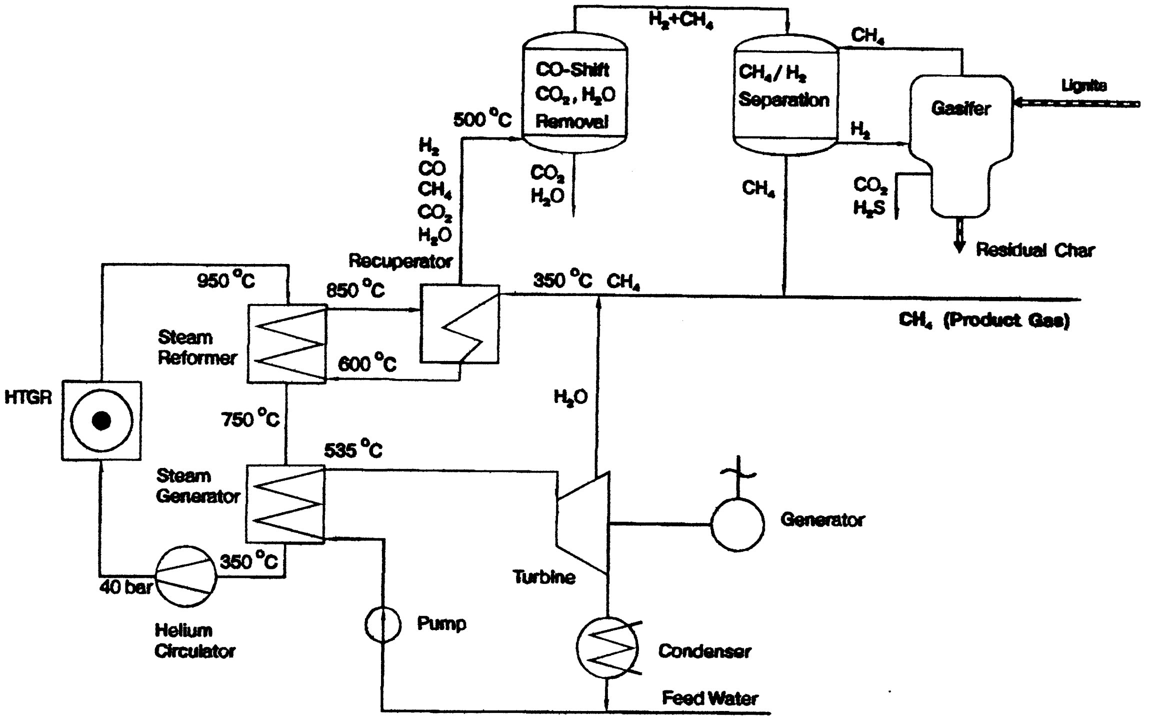


FIG. 6.11. Hydrogasification process of lignite.

The power level of the reactor or several modular reactors for a commercial coal gasification plant may be 2000–3000 MW(t). To avoid building large facilities, a gasification plant may contain several parallel loops of the same type connected to the reactor primary circuit. The power level of each loop may be as high as ~ 500 MW(t).

A coal gasification plant with a reactor of 3000 MW(t) power level can process 200 tons of coal an hour. The plant output will be 2.5 billion nm^3/year for methane production or 4 to 4.5 billion nm^3/year for hydrogen production with the capacity ratio of ~ 0.8.

6.3.2 Nuclear steam gasification of hard coal

Research and development work on process heat applications for the high temperature reactor, HTR, have been carried out in Germany in the past under the framework of the project "Prototype plant nuclear process heat, PNP" for different chemical processes such as steam gasification of hard coal, hydrogasification of lignite, steam reforming of methane etc., whereby the production of synthesis gas, hydrogen or methanol was investigated.

Theoretical and experimental work has shown that the process of steam gasification of different varieties of hard coal with different swelling behaviour is technically feasible and its further development to industrial scale is also possible. Different design and process parameters have been varied to improve the economical competitiveness of the products of the steam gasification of hard coal such as hydrogen in comparison with other conventional methods. A short review of these results have been described in the following.

The concept of the steam gasification of hard coal is based in the principal of providing the reaction heat through transporting the hot gas helium from the HTR to the heat exchanger tubes immersed in a fluidized bed of small coal granulates and steam. Investigation has been made with the gas generator directly arranged in the primary circuit of the HTR to improve the economic conditions of the steam gasification, whereby the intermediate He/He-heat exchanger, as well as the secondary helium circuit can be avoided, as shown in Figure 6.12. However this design concept must also fulfill the same safety requirements as in case of with secondary helium circuit.

Process conditions have been further changed to optimize this process. The helium temperature at the inlet of gas generator has been increased, system pressure has been reduced and the degree of coal conversion has been varied. Moreover, investigation has also been carried out with other possible improving potentials such as changing the size of the coal granulate and through the application of a catalyst to increase the reaction rate as well as coal throughput and at the same time to reduce the consumption of steam, so that more electricity can be produced with the spared steam. Some of the optimized data of the steam gasification process have been shown in the flow sheet of Figure 6.12.

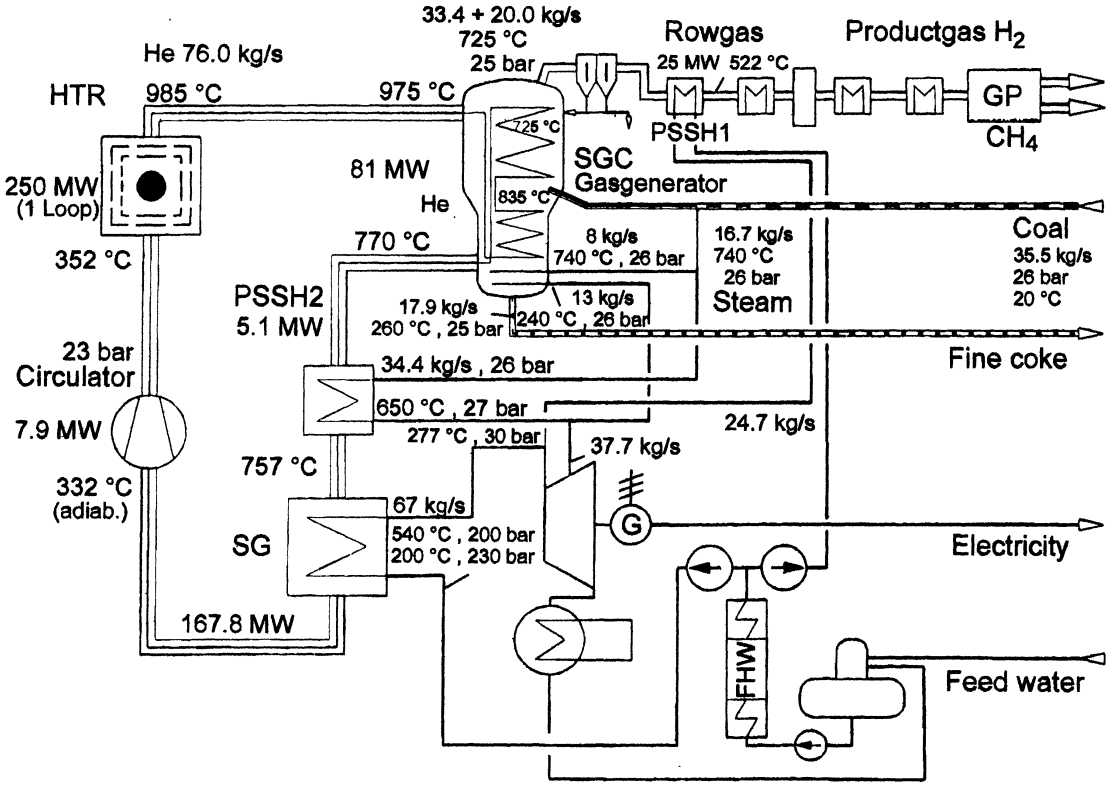


FIG. 6.12. Diagram of steam gasification process utilizing the HTR.

The results of increasing the helium temperature and the change of the coal conversion degree are shown in Figure 6.13. It can be seen that with the increase of helium temperature of about 100°C, the coal throughput and, therefore, also the coal conversion can be increased by about 50%. Moreover, it can also be shown that the coal gasification to the degree of about 95% is possible, however, the coal throughput and also the gas production is thereby reduced.

Futher improvement in the design is made through the new conception of the vertical gas generator instead of its previous horizontal design. With this concept, it is possible to arrange the pyrolysis and gasification zones above each other as shown in Figure 6.14. It is the possible to realize the heat transfer from primary helium to the gas generator in a counter-flow, whereby the increment of coal throughput as well as the gas production can be realized and at the same time the steam requirement for the process can be reduced.

In this design, feed coal powder is introduced in the pyrolysis section of the fluidized bed in the upper part of the gas generator through gas stream inlets. During the reaction process, the reacted coal granulates sink downward to the lower part of the fluidized bed, where the higher temperature gasification zone prevails. Residual coke after the gasification of the coal is discharged at he bottom outlet.

Heat exchanger tubes immersed in the fluidized bed are made of Inconel 617 material (2.4663) The construction of these tubes allows for inservice inspection, because this gas generator is arranged in the primary helium circuit and, therefor, these provide the barrier between the primary and secondary circuit in respect to safety requirements.

Investigation into the economics have shown that with these design and process improvements, hydrogen produced via nuclear steam gasification of hard coal is economically competitive with hydrogen produced through the conventional Texaco-gasification process.

6.3.3 Development of nuclear coal conversion technology

6.3.4.1 R&D programme on nuclear-coal conversion technology

As a long term programme, the R&D work into nuclear-coal conversion technology can be divided into three phases:

- The feasibility study phase: The aim of this phase is to develop a technical scheme of nuclear-coal conversion that would be suitable from the point of view of both technologies and the economy for the Chinese condition. The scope of feasibility study will be:

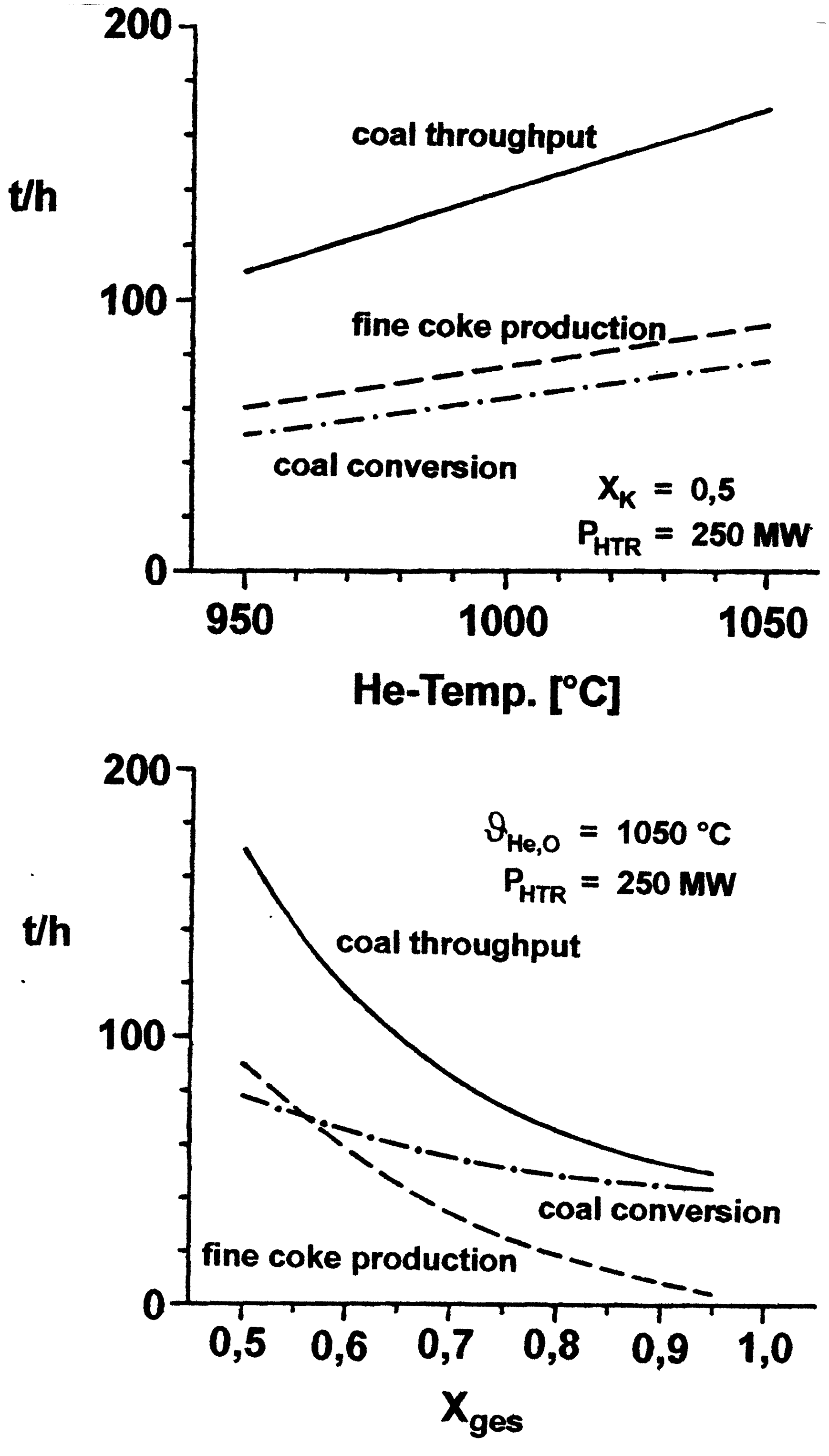


FIG. 6.13. Relationship between coal throughput and coal conversion as a function of helium temperature and coal conversion degree.

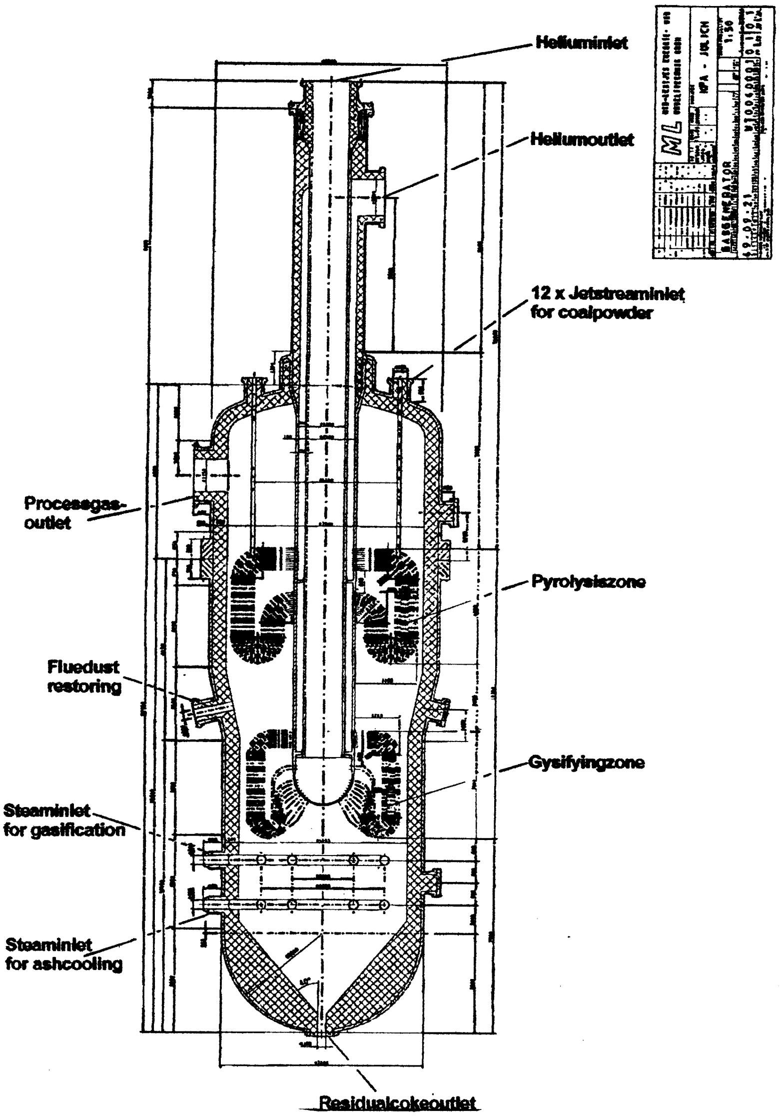


FIG. 6.14. Vertical gas generator.

Evaluation of the technologies and economy of coal gasification and liquefaction development

Evaluation of domestic technical conditions

Research on the technology scheme of nuclear-coal conversion

- Simulation experiments of coal gasification by hot helium gas in laboratory scale. The aims of this phase are:

To develop coal gasification technologies in helium heating conditions

To determine the optimum technical parameters

To obtain operating experience

- Nuclear-coal gasification experiment using the HTR-10 as a heat source. The HTR-10 will operate at temperatures up to 950°C and supply high temperature process heat for coal gasification tests. The aim of this phase is to obtain design and operating experience on nuclear-coal conversion technologies.

6.3.4.2 Simulation experiment for steam reform in laboratory conditions

The schematic diagram for the simulation test of methane steam reforming is shown in Figure 6.15. This includes two parts: the methane steam reformer heated by hot helium gas from the helium test loop and the methane conversion chemical reactor. The two parts form a closed test cycle. The closed cycle configuration is not only convenient for experimental study, but can also be used to study both methane reforming and conversion at once.

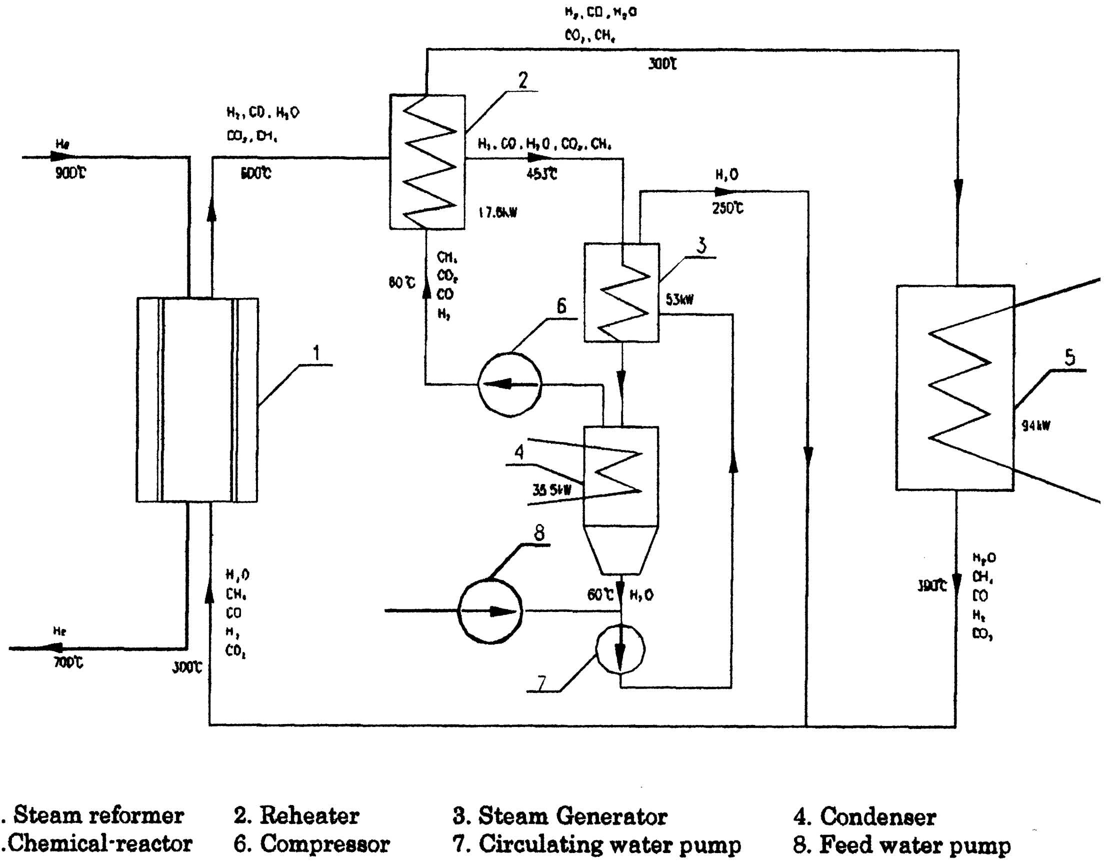


FIG. 6.15. Schematic diagram for the simulation of methane steam reforming.

6.3.3.3 Option selection of nuclear-coal conversion

The transformation method can be classified into two categories: gasification and liquefaction. Technology of direct liquefaction from coal is still in development. Some problems, such as loss of catalyst activity; separation of solid reminds and materials for apparatus etc. are still to be resolved. Therefore, final realisation for the industrialisation of this technology will require a relatively long time. For the time being, production of liquid fuel from coal adopts the indirect method, that is to first gasify the coal into methane and then to compose it to methyl-alcohol.

Technology of coal gasification is mature. Gasification with hydrogen or steam are both widely applied in industry. According to estimates, the coal resources which can be used for gasification is about 39 % of the total coal reserves in China. There are two methods for the coal gasification:

(1) Nuclear-coal conversion with steam. The main advantage of this method is the application of two independent helium heat transfer lines. It provides for separation of the reactor part from the gasification portion, to avoid permeation of hydrogen from the gasifier into the reactor and to avoid permeation of tritium from the reactor into the gasifier.

The main problem is that the efficiency of gasification is low. There are two ways to increase efficiency, a) increase the helium temperature at the reactor outlet to higher than 1200^{o}C (however, this is currently not possible for the HTGR) and, b) addition of oxygen as a catalyst to increase the reaction efficiency. For this an oxygen-production system is required. This makes the process scheme complicated and decreases the economic competitiveness.

(2) Nuclear-coal conversion with hydrogen addition. The main advantage of this method is that the helium temperature at the reactor outlet does not need to be higher than 950^{o}C for gasification of lower reactivity coal. Because of this, the heat exchanger is working at lower temperatures and atmospheric conditions and material selection becomes easy. Also, as the heat does not go directly to the fluidised bed, there is no problem with scale deposition. Thus, safety is ensured and the intermediate heat exchanger can be eliminated. Consequently, higher temperature heat can be supplied and the economic competitiveness can be improved.

Considering Chinese conditions, the industry base is not sufficiently developed. There is a lack of experience in the construction and operation of the HTGR, so it would be very difficult to increase the helium temperature at the reactor outlet. From the point of view of Chinese practice, the technical line of the gasification with the addition of hydrogen is more feasible and realistic.

6.3.3.4. Nuclear-coal gasification experiment using the HTR-10

In order to obtain design and operating experience on nuclear-coal gasification, it is planned that a pilot nuclear-coal gasification experiment facility will be built which uses the

HTR-10 as the heat source. The reactor will operate at temperatures up to 950°C to supply process heat for coal gasification.

Coal gasification with hydrogen is a conventional technology. Hence the goal of the HTR-10' high temperature process heat (HTPH) application test is only to demonstrate the use of nuclear heat in a steam reformer. The flow scheme of the HTR-10 HTPH experiment is shown in Figure 6.16. The reformer, which is located in secondary loop, is heated by the reactor through an intermediate heat exchanger. The goal of the indirect cycle is to provide a feasible design option that involves fewer licensing steps. The methane gas and steam mixture go to the steam reformer which produces hydrogen and carbon-oxide gas. The product gas is then cooled to about 50°C by a recuperator, a feed water preheater and a water cooler. The condensed water is separated from the gas in a water separator and the dry product gas is circulated by a gas compressor to the recuperator. There the hydrogen and carbon oxide gas mixture is reheated to 300°C . The reheated gas re-combines to form methane gas in a multistage methane conversion device. The heat of reaction is removed by water producing steam which is added to the flow entering the steam turbine cycle and the chemical cycle.

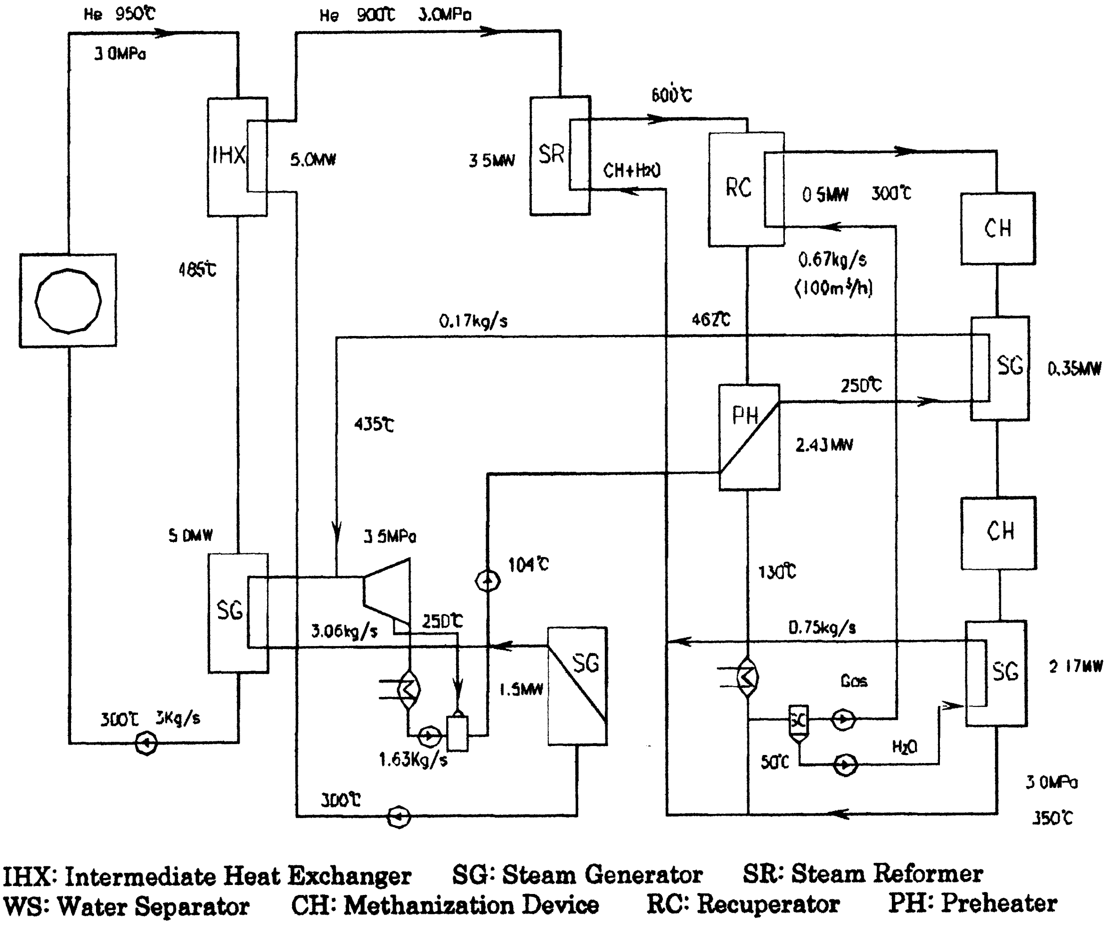


FIG. 6.16. Schematic circuit of the HTR-10 HTPH application.

6.4 NATUNA GAS USING NUCLEAR HEAT

6.4.1 Background

The Natuna gas field, discovered in 1973, is located in the Natuna Sea, Indonesia. It is one of the largest gas fields in the world, both in terms of total gas-in-place and of recoverable hydrocarbons. The total gas in the Natuna reservoir, including CO_2 and other impurities, is estimated to be 210 trillion cubic feet (6000 billion cubic meters), consisting of about 60 trillion cubic feet hydrocarbon and 150 trillion cubic feet carbon dioxide and other waste gases. Recoverable hydrocarbon reserves are estimated to be about 75% of the total or 45 trillion cubic feet (1270 billion cubic meters).

A typical composition of the Natuna gas is given below:

C_1	25.1	mole % (w)
C_2-C_4	0.7	mole % (w)
C_{5+}	0.3	mole % (w)
CO_2	66.5	mole % (w)
H_2S	0.52	mole % (w)
N_2	0.38	mole % (w)
H_2O	6.55	mole % (w)

The conventional concept for processing the Natuna gas comprises offshore gas production and gas treating, onshore gas treating and LNG production, and waste gas disposal into underground aquifer formations of porous rocks. Offshore gas will be produced directly from the reservoir. The produced gas will be cryogenically separated into sales gas (mostly methane) and waste gases (mostly carbon dioxide). The offshore facilities will include drilling and quarters' platforms, in addition to the very large gas treating platforms.

The treating platforms' offshore processing facilities will provide the bulk extraction of CO_2 for inlet cooling and separation of the produced gas. Raw gas from the reservoir will enter the inlet treating section at 91°C and 86 bar. The raw gas is cooled and condensed elements removed. A two-step process will be used to condense and separate the bulk of CO_2 and H_2S from the methane. First, a high-pressure stripper operating at -22°C and 55 bar will remove about 70% of the inlet CO_2. Then, a cryogenic stripper operating at -56°C and 47 bar will remove additional CO_2, in order to produce a treated gas stream containing approximately 80% methane, 18% CO_2, 940 ppm H_2S and 1% N_2. This hydrocarbon-rich stream will be warmed and compressed for pipeline transport to the Natuna Island. A 225-km pipeline to the Natuna Island for further treating and then liquefaction into LNG will transport the hydrocarbon sales gas.

6.4.2 Nuclear heat for upgrading the Natuna gas

The use of high temperature heat in the range of 880−900°C in the secondary helium loop of HTGR for direct CO_2 reforming of the Natuna gas was proposed [34, 35]. The product of the reforming process is synthesis gas (a mixture of CO and H_2), which can be further processed to produce methanol or other liquid fuels. The major problem with the Natuna gas is that the content of CO_2 is substantially higher than the stoichiometric quantity required for the reaction with CH_4. One option is to perform the first separation step offshore and create a mixture with a molar ratio of CO_2/CH_4 = 1:4. This mixture can be utilized in 'wet' CO_2 reforming as follows:

$$3/4\,CH_4 + 1/4\,CO_2 + 1/2\,H_2O + HTGR_{heat} \Leftrightarrow CO + 2H_2 \qquad (1)$$

The product synthesis gas, with a molar ratio of CO/H_2 = 1:2, can be used to synthesize methanol as follows:

$$CO + 2H_2 \rightarrow CH_3OH \qquad (2)$$

The surplus of CO_2 after the cryogenic separation from the raw gas can be injected into the underground carbonate formations for permanent disposal.

An additional process is 'dry' CO_2 reforming. No separation step of the CO_2 is needed and only purification of the gas to remove primarily H_2S is required. The Natuna gas is reformed directly according the following reaction:

$$CH_4 + CO_2 \Leftrightarrow 2CO + 2H_2 \qquad \Delta H^0_{298} = 247\ kJ/mole \qquad (3)$$

This reaction is highly endothermic and is performed at 850–900°C. The energy for the process can be supplied by the HTGR.

In this process, the surplus of CO_2 (about 2/3 of the volume) is separated and removed after the reaction takes place. If methanol is the desirable end product, 2 moles of H_2 can be imported from an external process such as electrolysis on a thermochemical process (i.e., the IS process), in order to create a ratio of CO/H_2 = 1:2, as expressed in Eq. (2). In addition, 3 moles of H_2 are needed for each mole of the separated CO_2, in order to convert the surplus of CO_2 as follows:

$$CO_2 + 3H_2 \rightarrow CH_3OH + H_2O \qquad \Delta H^0_{298} = -49\ kJ/mole \qquad (4)$$

Between these two options, there are several intermediate alternatives. The common requirement is the external addition of hydrogen. Alternatives that do not require addition of hydrogen are, for instance, the synthesis of formic acid according to the following expression:

$$3CH_4 + 9CO_2 + 6H_2O \rightarrow 12CH_2O_2 \qquad (5)$$

CO_2 can also be converted to methane according to the following reaction:

$$CO_2 + 4H_2 \rightarrow CH_4 + H_2O \qquad \Delta H^0_{298} = -165\, kJ/mole \qquad (6)$$

In this case, 4 moles of external hydrogen are required. The CO_2 in the Natuna gas is converted to CH_4. The content of the CH_4 in the original gas is enriched. The gas is then liquefied and can be transported as LNG.

6.4.3 Economic considerations

Barnert [36] describes the results of cost estimates for the exploitation of the Natuna gas field in Indonesia using the high temperature reactor (HTR). He shows that there is a potential for economic competitiveness for the exploitation of the Natuna gas, using high temperature nuclear energy from the HTR, primarily because of the relative high market value of methanol and synthetic fuels as substitutes of gasoline and diesel oil. The economic analysis has been performed for the following processes:

(1) Steam and CO_2 reforming, and additional electrolytic hydrogen and synthesis of methanol.

(2) CO_2 reforming and synthesis of methanol from the gas reformer, separation of the CO_2 surplus and addition of electrolytic hydrogen to synthesis of methane.

(3) Addition of electrolytic hydrogen to the Natuna gas to convert all the CO_2 to methane.

(4) Separation of methane, reaction of CO_2 with external electrolytic hydrogen to synthesis methanol.

The results of the cost estimates of methanol and methane from the Natuna gas, based on their calorific value, are summarized in the table below [36], referring to the above four processes.

Cost Estimate of the Natuna Gas Conversion to Methanol (*l*) and Methane (g) Using HTR (according to Barnert [36])

Where:

1: Methanol; min., middle, max.	High S: 1500$/kWe for the HTR 1000$/kWe for ELY
2: Methane:enrichment + methanol	
3: Methane	Low S: 50% of the high S
4: Methane: separation + methanol	NA: Natuna Gas

4 Processes: Process 1 in 3 Variants							
Process		1			2	3	4
Variant		a	b	c			
Methanol (*l*) (%)		100	100	100	51	0	68
Methane (g) (%)		0	0	0	49	100	32
NA CO_2 (%)		15	25	100	100	100	100
Ratio CO_2/CH_4		0.4	0.67	2.67	2.67	2.67	2.67
Estimated Cost: C: $/GJ							
Variation of S for HTR+ELY	High S	4.9	7.5	15.1	16.5	16.9	15.2
	Low S	4.1	5.5	9.4	10.2	10.1	9.2

6.4.4 Economic assessment

The use of high temperature heat from the HTR for the conversion of the Natuna gas to methanol is economically feasible. It has the potential to provide for mass production of HTR modules, which can lower the specific cost of the investment. Process (1) is the most attractive candidate; however, more detailed analyses and optimization are required.

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Chapter 7

SUMMARY AND CONCLUSIONS

The primary focus of this CRP was to perform detailed investigation of the high temperature industrial processes that are attainable through incorporation of an HTGR, and for their possible demonstration in the HTTR. The HTGR has the capability to achieve a core outlet temperature approaching 1,000° C in a safe and effective manner. These attributes, coupled with the offer by JAERI to utilize the HTTR, resulted in the initiation of this CRP by the IAEA.

7.1 HIGH TEMPERATURE ENGINEERING TEST REACTOR

The HTTR utilizes a 30 MW(th) HTGR comprised of 30 fuel columns of hexagonal pin-in-pin graphite block type fuel elements. The fuel consists of UO_2 TRISO coated particles with an enrichment of ~ 6%wt. Relative to the demonstration of high temperature heat applications, the HTTR will be capable of producing 10 MW(th) of heat at 950°C. However, the thermal power for these applications has the potential to be increased up to 30 MW(th) in the future, which may be required for demonstration of gas turbine system components. The HTTR reached initial criticality in November, 1998. Initial operational plans includes a series of rise to power tests followed by tests to demonstrate the safety and operational characteristics of the HTTR.

In addition to completion of the HTTR demonstration tests, it was recommended that the R&D given in Appendix "A" be performed within the HTTR project. JAERI is encouraged to publicize the results of the HTTR tests and "lessons learned" from their experiences including potential capabilities of the HTGR for heat applications. This could be in the form of JAERI/HTTR personnel arranging for international technical seminars, conferences and publications to industries, utilities and financing organizations.

7.2 PRIORITIZATION OF HEAT UTILIZATION SYSTEMS

Paramount among the recommendations of the Chief Scientific Investigators (CSIs) was the prioritization of individual heat utilization systems for demonstration using the HTTR. This prioritization was influenced by the significance of the application, the current state of technology development for each application, and the capability to couple it to the HTTR.

It was determined that the major focus should be on high temperature applications of nuclear power which would result in the production of hydrogen. The need to depart from the burning of fossil fuels was considered to be a priority worldwide requirement.

Production of hydrogen as an energy carrier for the future through the reforming of methane was selected as the highest priority heat utilization application. Reforming of methane with steam and carbon dioxide were investigated and, although the primary goal was the production of hydrogen, both processes have the proven ability to result in the final production of methanol (or syngas) through subsequent synthesis. This chemical conversion of natural gas with the HTGR offers the added benefits of a substantial decrease in CO_2 emissions and an increase in calorific value of the products with a corresponding greater fuel versatility.

The next priority application was determined to be the generation of electricity through the use of the gas turbine. Application of the Brayton Cycle utilizing high temperature helium from a modular HTGR was chosen for development because of its projected benefits as an economic and efficient means for the production of electricity.

Evaluation of the remaining high temperature heat utilization applications chosen for investigation by the CSIs resulted in the prioritized selection of hydrogen production through thermochemical water splitting, followed by the conversion of coal into higher quality fuels. These processes are to be demonstrated by out-of-pile tests prior to coupling to the HTTR.

The main findings and conclusions for each of the systems evaluated within this CRP are as follows:

- Reforming of methane for hydrogen production and synthesis

  Steam reforming of methane for the production of hydrogen is planned to be the initial heat utilization process demonstrated with the HTTR. This reforming process for hydrogen production is well known industrially and is technologically mature. The hydrogen production performance with a heat utilization ratio (i.e. ratio of the product energy to total input energy) of up to 78% in the reforming system is expected to be demonstrated with the HTTR at a thermal power level of 10 MW. The integrated control system of the HTTR with the steam reforming system is determined to be technically feasible and will also be demonstrated in the HTTR.

  Significant experience in out-of-pile tests and design studies associated with steam reforming of methane exist in Germany, China and the Russian Federation. The R&D activities to be conducted prior to demonstration of this process in the HTTR include out-of-pile testing and additional studies associated with design and safety such as the establishment of safety standards associated with the explosion of feed and product gases and determination of the tritium permeation rate.

- CO_2 reforming of methane for hydrogen

  Large resources consisting of a mixture of CO_2 and natural gas exist worldwide which have the capability to be converted into usable synthesis gas. Also, bio-resources (CO_2/CH_4) can be used for conversion into synthesis gas with no net generation of CO_2. Although not as highly developed as the steam reforming process, CO_2 reforming of methane has been proven experimentally.

  As with steam reforming of methane, the R&D needs, design, safety assessment requirements such as the tritium permeation rate and explosion of feed and product gasses and goals of the carbon dioxide reforming process have many common similarities and the same facility can be used for the demonstration of both reforming systems. In this regard, the initial design work has been completed by JAERI for the HTTR heat application systems of steam and CO_2 reforming of methane and out-of-pile demonstrations of both processes will be performed prior to coupling to the HTTR.

- Gas turbine for electricity production

  The generation of electricity through the use of the gas turbine was determined to be a priority application (of similar status as steam reforming of methane) for demonstration with the HTTR. The CSIs determined that investigating the coupling of the HTGR with a gas turbine energy conversion system is an important milestone in the development of advanced nuclear power. This system eliminates the constraints imposed by the use of steam via the Rankine Cycle, which is predominant in the current generation of nuclear power plants.

Significant interest exists worldwide in this concept and several plant designs are currently in progress. Both the direct and indirect cycle systems are determined to be technically feasible. Out of pile testing experience exists in Germany on the helium gas turbine [1], and the potential exists for demonstration of associated power conversion system components with the HTTR.

Based on the understanding that further evaluation and consideration is necessary by JAERI prior to proceeding with the gas turbine system, the CSIs proposed connection of both the direct and indirect cycle systems to the HTTR. Besides the integrated HTTR demonstration test, this could include evaluation of component performance and material testing within the HTTR Project.

- Thermochemical water splitting for hydrogen production, iodine-sulfur (IS) process

  Of the many chemical reactions that have been evaluated utilizing the HTGR as the heat source, the IS process is considered one of the most attractive for thermochemical water splitting to achieve hydrogen. This process provides several significant features, including the capability to produce hydrogen from naturally abundant water, freedom from carbon emission thereby helping prevent environmental issues such as global warming, direct conversion of nuclear heat into chemical energy and the ability to provide a relatively (>40% has been evaluated) high thermal efficiency.

  The basic concept of this process was developed by GA and demonstrated at JAERI in the course of this CRP on laboratory-scale experiments attaining continuous and “closed-cycle” hydrogen production. Because of this achievement, a larger scale test was initiated in 1999 to develop closed-cycle operation techniques with modified Bunsen reaction conditions.

  In addition to this test, studies are underway on the membrane technologies for establishing efficient processing of hydrogen-iodide and on the materials of construction for a bench-scale plant.

  It was concluded by the CSIs that this thermochemical process is worth continuing as a future candidate for potential demonstration in the HTTR following bench- and pilot-scale experiments.

- Coal conversion

  The world reserves of high and low grade coal greatly exceed the known resources of oil and natural gas. Significant investigation has been undertaken by Member States into the processes for conversion of coal into higher quality, more convenient to use, fuels. Conversion of these reserves provides the benefits of easier fuel product transportation and improved environmental conditions, compared to the direct burning of coal.

Gasification of coal using external heat from the HTGR can have a 150 to 180% yield compared to conventional conversion processes. Research carried out within the scope of this CRP by Germany, Russia, China and the USA has demonstrated that coal reforming via HTGR produced thermal power can be technically feasible and is expected to be economically competitive. This nuclear heat process offers advantages over conventional gasification processes in terms of more efficient and environmentally friendly utilization of available primary resources. Use of the HTGR allows a nearly 40% reduction in coal consumption compared to existing methods for the same output in syntheses gas. Converted coal can be used as a fuel in conventional fossil plants, and hydrogen produced by an HTGR can be utilized for hydrogasification of coal and subsequent conversion into hydrocarbons.

The conventional technology of coal gasification is mature. Although the use of high helium temperature as achieved in the HTGR has been demonstrated for this process in out-of-pile tests, evaluation to determine the most advantageous process to be utilized (steam gasification, hydrogen addition, etc.), including further investigation into the associated safety and economic issues, and component/material development is required prior to coupling to a nuclear heat source.

- Heavy oil recovery

  Investigation within the scope of this CRP into the feasibility of using the HTGR in the recovery of heavy oil concluded that HTGRs are capable of producing the high temperature and high pressure steam necessary for this process and could be used as the need arises with current technology. Also, through the ADAM/EVA tests, Germany has investigated and demonstrated the capability for chemical energy transport over long distances in oil fields. However, it was determined by the CSIs that research and development specific for oil recovery was not a feasible application for demonstration with the HTTR and this heat utilization application was deleted from the CRP.

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Appendix A

RESEARCH AND DEVELOPMENT NEEDS FOR THE FUTURE

HTTR Steam Reforming System

- Need to Establish Design of Commercial Plant
- HTTR Out-of-Pile Test
  (1) Development of a dynamic simulation computer code including the reactor dynamics
  (2) Establishment of normal start-up and shut-down operation technology
  (3) Establishment of emergency shutdown operation technology, considering issues such as structural integrity of the reforming tube
  (4) Optimize the design of a reformer from the view point of size, thermal efficiency, economics, easier removal of the used catalyst, lower stresses of the reformer tubes, etc. based on the HTTR out-of-pile test and the EVA-ADAM project
  (5) Optimization of feedgas composition (CH_4, H_2O, and $C0_2$) for methanol production
- Component Tests
  (1) Evaluation of the mechanism for permeation of the hydrogen isotope through the IHX and SR tubes
  (2) Development of an analytical code for tritium and/or hydrogen transport
  (3) Obtaining corrosion data of the reforming tubes

Commercial HTGR process heat application system

- First generation plant (steam reforming system)
  (1) Compact heat exchanger with high temperature and high thermal efficiency
  (2) High temperature isolation valve
  (3) Material development for reforming tube (high thermal and high pressure resistance)

- Future advanced plant
  (1) IS process
    (1). (a) Development of closed-cycle operation techniques under efficient process conditions of the Bunsen reaction
      (b) Establishment of the HI process scheme
      (c) Obtain corrosion database of the construction materials for the Bench-scale experiment
    (2) (a) Bench-scale experiment
      (b) Materials of construction for the Pilot-scale experiments
    (3) Pilot-scale experiment by He gas heating
    (4) Demonstration by nuclear heat
  (2) Development of new processes
    • Co-production of hydrogen and electricity
      - Fuel cell type reactor
      - Partial oxidation of methane
    • New thermo-chemical process for hydrogen production
    • Effective utilization of carbon in fossil fuels

- Advanced components
  Heat storage with high temperature
    - Enhancement of heat transfer

Further R&D for consideration utilizing the HTTR CO_2 reforming system

- Development and modification of the catalyst system to meet specific feedstock like Natuna gas, biogas, etc.
- Purification of the feed gas.
- Development of startup and shutdown procedures which may different for the various feedstock (e.g. if addition of steam is needed)
- Reducing the operating pressure in the reformer.
- Increasing the operating temperature of the reformer.
- Developing of an isothermal reformer.
- Developing of a buffer thermal storage to smooth the coupling of the chemical plant and the HTTR.
- Definition, design and demonstration of the coupling of the reformer with the end-user (gas-turbine, methanol plant, etc.)
- Demonstration of a “small” size HTTR coupled with CO_2 reformer for biogas feedstock for inherently safe, zero CO_2 emission nuclear plant.

Appendix B

ABBREVIATIONS

ACS	auxiliary cooling system
AEC	Atomic Energy Corporation
AHX	auxiliary heat exchanger
AOO	anticipated operational occurrence
ATWS	anticipated transient without scram
AVR	Arbeitsgemeinshaft Versuchsreaktor
BATAN	National Atomic Energy Agency of Indonesia
BNFL	British Nuclear Fuels plc
CAESAR	catalytically enhanced solar absorption receiver
CC	combined cycle
CCS	core conditioning system
CR	control rod
CRDM	control rod drive mechanism
CSS	cold shutdown system
CV	containment vessel
DBA	design basis accident
ECN	Netherlands Energy Research Foundation (now NRG)
EIA	environmental impact assessment
EPRI	Electric Power Research Institute
FA	fuel assembly
FC	fuel cell
FEB	fuel and energy balance
FHS	fuel handling system
FHSS	fuel handling and storage system

FSV	Fort St. Vrain
FZJ	Forschungszentrum Juelich GmbH (previously KFA)
GA	General Atomics
GCR	gas cooled reactor
GHTRN	global high temperature reactor network
GT	gas turbine
GT-MHR	gas turbine modular helium reactor
GT-ST	gas turbine/steam cycle
HENDEL	helium engineering demonstration loop
HEU	high enriched uranium (fuel)
HRB	hochtemperatur-reaktorbau
HKG	Hochtemperatur-Kernkraftwerk GmbH
HPC	high pressure compressor
HPTF	high pressure test facility
HSE	health and safety executive
HTGR	high temperature gas cooled reactor
HTR	high temperature reactor
HTES	high temperature electrolysis of steam
HTTR	high temperature engineering test reactor
HTR-10	high temperature reactor test module
HX	heat exchanger
IC	intercooler
IDC	Industrial Development Corporation
IHX	intermediate heat exchanger
INCOGEN	inherently safe nuclear COGENerator
INET	Institute of Nuclear Energy Technology

IS	iodine-sulfur process
ISFSI	independent spent fuel storage installation
IST	integrators of system technology
IWWGGCR	International Working Group on Gas Cooled Reactors
JMTR	Japan Materials Testing Reactor
JAERI	Japan Atomic Energy Research Institute
LEU	low enriched uranium (fuel)
LPC	low pressure compressor
LWR	light water reactor
MCS	main cooling system
MHR	modular helium reactor
MHTGR	modular high temperature gas cooled reactor
MINATOM	Ministry of Atomic Energy of Russia
MIT	Massachusetts Institute of Technology
MPS	main power system
NNR	national nuclear regulator
NPP	nuclear power plant
NRC	Nuclear Regulatory Commission
NRG	ECN Nuclear Research
OECD	Organisation for Economic Cooperation and Development
OKBM	Experimental Machine Building Design Bureau
PBMR	pebble bed modular reactor
PC	precooler
PCRV	prestressed concrete reactor vessel
PCS	power conversion system
PCU	power conversion unit

PCU	power conversion unit
PCV	power conversion vessel
PPWC	primary pressurized water cooler
PRA	probabilistic risk assessment
PTG	power turbine generator
PyC	pyrolytic carbon (coating)
QC	quality control
RAHP	research association of HTGR plant
RCCS	reactor cavity cooling system
RCS	reactivity control system
RCSS	reactivity control and shutdown system
RPV	reactor pressure vessel
RS	reactor system
RSS	reserve shutdown system
SAFSTOR	safe storage or safe enclosure decommissioning
SAS	small absorber sphere
SC	steam cycle
SCS	shutdown cooling system
SHE	shutdown heat exchanger
S/G	steam generator
SiC	silicon carbide (coating)
SINTER	safety related nuclear reactor technology elements-R&D
SCHP	solar chemical heat pipe
SOFC	solid oxide fuel cell
SPWC	secondary pressurized water cooler
THTR	thorium high temperature reactor

T/M	turbomachine
TRISO	coated fuel particle with PyC, SiC and PyC coatings
VCS	vessel cooling system
VHTR	very high temperature reactor
WIS	Weizmann Institute of Science

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RESEARCH CO-ORDINATION MEETINGS

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