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# Materials for the Very High Temperature Reactor (VHTR): A Versatile Nuclear Power Station for Combined Cycle Electricity and Heat Production

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*Abstract:* The International Generation IV Initiative provides a research platform for the development of advanced nuclear plants which are able to produce electricity and heat in a combined cycle. Very high-temperature gas-cooled reactors are considered as near-term deployable plants meeting these requirements. They build on high-temperature gas-cooled reactors which are already in operation. The main parts of such an advanced plant are: reactor pressure vessel, core and close-to-core components, gas turbine, intermediate heat exchanger, and hydrogen production unit. The paper discusses the VHTR concept, materials, fuel and hydrogen production based on discussions on research and development projects addressed within the generation IV community. It is shown that material limitations might restrict the outlet temperature of near-term deployable VHTRs to about 950 °C. The impact of the high temperatures on fuel development is also discussed. Current status of combined cycle hydrogen production is elaborated on.

**Keywords:** Gas-cooled reactor · Generation IV · Helium gas-turbine · High-temperature materials · High-temperature reactor · Hydrogen

## Introduction

Ten countries and the EU (represented by EURATOM) have formed the International Generation IV Initiative (GENIV, GIF) to develop advanced fission plants for co-generation of electric energy and process heat [1]. The very high temperature reactor (VHTR) is a next step in the evolutionary development of high-temperature gas-cooled reactors (HTR). The VHTR system is primarily aimed at relatively fast deployment of a system for electric energy production combined with high temperature process heat applications, such as coal gasification and hydrogen production, with superior efficiency. The efficiency is over 50% at 1000 °C, compared with 47% at 850 °C. Co-generation of heat and power makes the VHTR an attractive heat source for large industrial complexes. The VHTR can be deployed in refineries and petrochemical industries to substitute large amounts of process heat at different temperatures, including hydrogen generation for upgrading heavy and sour crude oil. Core outlet temperatures higher than 1000 °C would enable nuclear heat application to such processes as steel, aluminium oxide, and aluminium production as summarized in [2]. The main elements of an advanced VHTR co-generation plant are shown in Fig. 1. The plant consists of the gas-cooled reactor part, a direct cycle (Brayton cycle) helium gas-turbine driving the generator of electric energy, heat exchangers and different options for hydrogen production. The VHTR can produce hydrogen from only heat and water by using a thermochemical iodine-sulphur (I-S) process or from heat, water, and natural gas by applying the steam reformer technology to core outlet temperatures greater than about 1000 °C. A vision of a reference VHTR system that produces hydrogen is shown below. A 600 MWth VHTR dedicated to hydrogen production can yield over 2 million normal cubic meters of hydrogen per day.

Current state-of-the-art high-temperature gas-cooled reactors operate at maximum steady state temperatures of 850 °C. Although for the Japanese high-temperature engineering test reactor (HTTR) operation temperatures of 950 °C were reported [3] this temperature is currently considered as advanced VHTR technology. The variety of problems to be solved in the fields of materials for the gas turbine and hydrogen process give rise to expectations that this very advanced concept will be achieved in two steps. It might be speculated that the near-term VHTR (probably deployed around 2020) will operate at maximum gas temperatures of 900 to 950 °C with an indirect cycle (*e.g.* ANTARES programme of Areva [4]) using an intermediate heat exchanger as interface to the non-nuclear part of the plant. Temperatures of 1000 °C and higher together with a direct cycle helium gas turbine might be the generation of plants deployed beyond 2020.

## VHTR Concepts

The VHTR is a graphite-moderated, helium-cooled reactor with thermal neutron spectrum. It can supply nuclear heat

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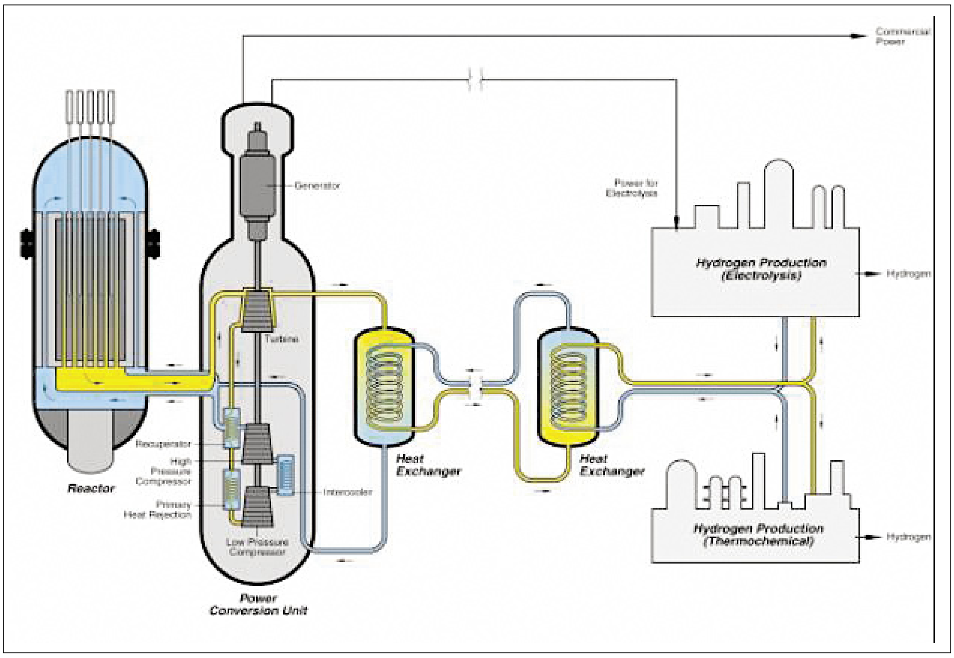

Fig. 1. Advanced VHTR plant for co-generation of electricity and hydrogen. According to the Technology Roadmap for Generation IV Nuclear Energy Systems [1], published with permission of GENIV.

with core-outlet temperatures of 1000 °C. The reactor core type of the VHTR can be a prismatic block core or a pebble-bed core. For electricity generation, the helium gas turbine system can be directly set in the primary coolant loop, which is called a direct cycle. For nuclear heat applications such as process heat for refineries, petrochemistry, metallurgy, and hydrogen production, the heat application process is generally coupled with the reactor through an intermediate heat exchanger (IHX), which is called an indirect cycle.

The VHTR evolves from HTR experience gained over 20–30 years and extensive international databases that can support its development. The ongoing 30-MWth HTTR project in Japan [5] is intended to demonstrate the feasibility of reaching outlet temperatures up to 950 °C coupled to a heat utilization process, and the HTR-10 in China [6] will demonstrate electricity and co-generation at a power level of 10 MWth. The former projects in Germany and Japan provide data relevant to VHTR development. Steam reforming is the current hydrogen production technology. The coupling of this technology will be demonstrated in large scale in the HTTR program but still needs complementary R&D for market introduction. R&D on the thermochemical I-S process is presently proceeding in the laboratory-scale stage.

Table 1 lists all HTRs which have been in operation or which currently are operated under test conditions. Pebble-bed types (*e.g.* PBMR in South Africa [7]) as well as prismatic types are under consideration and until now no clear preference for the one or the other concept can be given. As our discussion will not depend on the type of reactor core we will not further investigate this question in this paper. There are also plans in the USA [8][9] and in South Korea [10] to build VHTR-based demonstration plants.

In South Korea, KAERI runs a nuclear hydrogen development and demonstration project (NHDD) with the aim to run a demonstration plant by the year 2019. It is planned to include a direct cycle helium turbine and an iodine sulphur hydrogen plant. The decision whether a pebble bed or a prismatic core design is used has not yet been taken. A similar VHTR reactor is projected as a new generation nuclear plant (NGNP) in the USA. It is planned for co-generation of electricity and hydrogen until 2020. The plant is intended to demonstrate a full-scale prototype VHTR that is commercially licensed by the US Nuclear Regulatory Commission and to demonstrate safe and economical nuclear production of hydrogen and electricity.

## VHTR Materials Issues

The challenges for materials in VHTRs are manifold. The gas temperature shall be at least 1000 °C in the outlet and the backflow can reach 650 °C. Although helium is basically an inert gas it carries un-avoidable impurities ($CO$, $CO_2$, $N_2$, $O_2$, $H_2O$) which can lead to high-temperature corrosion degradation of the materials. In-core and close-to-core located components are subjected to neutron irradiation which is an additional degradation source. The lifetime of the non-replaceable components should be 60 years, which corresponds to about a 500'000 h design life which is far beyond usual laboratory testing times. New techniques for damage assessment and damage modelling must therefore be developed. The possible materials for different components are shown in Table 2. The different components will be discussed in the following.

## Pressure Vessel

The pressure vessel of a HTR must be made of steel that can withstand stresses for temperatures up to 400 °C in current designs and up to at least 500 °C in the currently considered future designs. At these temperatures the stresses upon the pressure vessel can lead to creep and/or relaxation. However, to design a pressure vessel with creep taken into account would require

Table 1. HTR reference plants

| **Reactor Type** | **Power** | **Operation** |
|---|---|---|
| **Pebble bed reactors** | | |
| AVR Germany | 46 MWth/15 MWel | 1966–1988 |
| THTR-300 Germany | 750 MWth/296 MWel | 1985–1989 |
| HTR-10 China | 10 MWth | Since 2000 |
| PBMR South Africa | 400 MWth/125 MWel | Expected 2010 |
| **Prismatic core** | | |
| Peach Bottom 1 US | 40 MWel | 1966–1974 |
| Fort St Vrain US | 842 MWth/330 MWel | 1976–1988 |
| HTTR Japan | 30 MWth | Since 1998 |
| GT-MHR US/Russia | 600 MWth/293 MWel | Point Design |

Table 2. Materials for current and future gas-cooled reactors (T refers to gas temperature)

| **Component** | **T ≤850 °C** | **850 °C< T ≤950 °C** | **T >950 °C** |
|---|---|---|---|
| Reactor Pressure Vessel | LWR-RPV (ferritic) | 2 ¼ Cr 1 Mo (ferritic), 9-12% Cr-steel (martensitic) | 9–12% Cr steel (martensitic) |
| Control Rod | Ni-base superalloy, IN-800H | Ni-base superalloy, C‘C, SiOC, SiC/SiC | C/C, SiC/C, SiC/SiC |
| Graphite, ceramic internals | Graphite | Graphite (new grades), SiC/C, SiC/SiC structural parts | Graphite (new grades), SiC/SiC structures, (super plastic) ceramics |
| Metallic internals | Steels | Steels, Ni-base superalloys, ODS | Steels, Ni-base superalloys, intermetallics, ODS |
| Piping/IHX/valves | Ni-base superalloys | Advanced Ni-base superalloys (eventually with TBC-coatings) | Advanced Ni-base superalloys with TBC-coatings, cooled designs, ceramics, intermetallics, composite structures |
| He-Gas Turbine: | | | |
| Blades/Vanes | Ni-base superalloys (gamma prime hardening) | Ni-base superalloys (DS.SC) | Ni-base superalloys (DS,SC), cooled designs, ODS, intermetallics, refractory alloys, composites |
| Rotor | Ferritic-martensitic steels (cooled designs) | Ferritic-martensitic steels (cooled designs), Ni-base superalloys (gamma prime hardening) | Ni-base superalloys (gamma prime hardening), advanced cooling technology, composites |

a lot of additional design data and curves (including creep strain data, multiaxiality and creep, creep of welds, notch sensitivity *etc.*) as well as procedures for surveillance under creep conditions. The avoidance of creep needs design measures and highly creep-resistant materials. Current light water reactor pressure vessel steel can be used for temperatures up to 350 °C [11]. The classes of ferritic (NiCr)MoV steels and the more creep resistant 9–12% martensitic chromium steels are very well established creep resistant materials for a temperature regime of 400–550 °C. They have been used in chemical plants, in boilers, in steam- and gas turbines and in jet engines. Temperature extensions to 600 °C have been tried for different applications (*e.g.* large steam turbine rotor forgings). A summary of these developments is given in [12]. As a result of its low activation and its high thermal conductivity this class of steels is also very interesting for fusion reactor applications [13]. The development has now reached a stage where no significant improvements of the stress rupture behaviour are expected anymore by changing the chemical composition. Only a change of the matrix (from ferritic martensitic to austenitic) or reinforcements of the martensitic matrix (*e.g.* by oxide dispersion strengthening) could lead to significant improvement of the creep properties. However due to the difficulties in producing large components, reliable welds and due to expected difficulties in non-destructive testing it can be stated that according to current knowledge, ODS materials cannot be considered for RPVs.

The choice of materials for a VHTR depends on the design of the vessel and the design rules. There are claims that a 2 1/4 Cr 1 Mo-steel would be sufficient but the majority of researchers propose an advanced steel of the 9–12% Cr-class. It should however be noted that the use of 9–12% martensitic chromium steels for RPVs represents a significant challenge for complete through-section heat treatment, fabrication, welding, and post weld heat treatment. It should also be noted that 9–12% martensitic chromium steels are not currently included in the ASME Boiler and Pressure Vessel Code but there are plans to include the 9Cr-1Mo (T91) steel in future revisions.

## Reactor Internals

The reactor internals of a current HTR mainly consist of a graphite core, control rods (superalloy Hastelloy XR) and steel support structures. A fundamental problem of graphite in nuclear reactor cores is the deterioration of mechanical and other properties as a result of the neutron irradiation. The primary source of degradation is the stresses that develop during irradiation [14]. Graphite will remain the core part of VHTRs and therefore the irradiation behaviour of different graphite properties is to be investigated in different international research projects, *e.g.* [15]. It also might be worth considering designs in which bricks or core parts are replaced by composite structures filled with graphite for use as a moderator. The advantage of composite structures is that their structural integrity is maintained even when locally cracked. This is the reason why black composite ceramics of C/C, SiC/C, and SiC/SiC (where the matrix material (M) is filled into a woven fibre structure (F) to form the (F/M) composite) are currently being investigated as future materials for smaller structural parts and liners. A possible near-term application for black ceramic compounds consists in SiC/SiC or SiC/C as material for the control rod components [1] [11]. Different properties of C- and SiC-type materials are therefore currently being investigated.

In the event of higher gas outlet temperatures, the gas inlet temperatures will rise as well and so the temperature level in the reactor will also increase. As well as the core, possible materials for support structures, boltings and fixtures must also be able to accommodate higher temperatures. For such components oxide dispersion strengthened (ODS) materials, intermetallic phases and (superplastic) ceramics are considered as possible material candidates. Intermetallic phases are ordered structures (*e.g.* TiAl, NiAl, $Fe_3Al$, $MoSi_2$, $Si_2Zr_3$ *etc.*). A level of high energy is needed for the movement of dislocations in the lattice of ordered structures, which leads to a high yield strength (and low toughness) of such materials up to high temperatures. Titanium aluminides and nickel aluminides are already in use today for conventional structural applications [16]. Other intermetallics are still in the development phase. Research for the reinforcement of intermetallics in terms of fibres or dispersoids to improve their very high temperature properties is currently being pursued worldwide, *e.g.* [17][18]. The influence of point defects created as neutron irradiation damage within ordered intermetallic structure [19][20] will be a main topic to be investigated for future VHTR applications. Another scientific question to be clarified concerns the effect of impurities in the reactor coolant gas helium on the long-term behaviour of these materials.

For temperatures in the range of 1000 °C and higher ceramic materials could be used, however complex shaped parts are difficult to machine out of solid ceramics. $ZrO_2$-based fine grained super plastic ceramics can be shaped into complex structures much easier. Feasibility studies are currently underway to demonstrate the suitability of this class of materials for core internal applications in future VHTRs [21].

### Pipings and Valves

Solid solution strengthened nickel base superalloys like IN 800 and IN 800 H have already been investigated thoroughly for piping and other balance of plant applications for today's HTR-technology [22]. In Section IIC of the ASME Boiler and Pressure Vessel Code which deals with Pipings and Valves, it is indicated that alloy 800H can be used for temperatures up to 950 °C. However, no guidance for use of this material at 950 °C is currently included in the code. Also, future revisions of this code to include design information that would include the service temperature for Alloy 800H are not planned. Temperatures of up to 950 °C can probably better be reached with higher creep resistant, advanced nickel base superalloys like Haynes 230 or IN 617. A further temperature increase (up to 1000 °C) also pushes these materials towards their temperature limits where strength and creep properties drop very quickly. For heavily stressed parts subjected to very high temperatures, reinforcement like dispersoids (ODS) or intermetallics could be alternatives. Double walled piping with cool gas moving through the outer section can help to cool the hot gas ducts. Thermal barrier coatings can further reduce the material temperature. For piping sections this concept would allow operating temperatures in excess of 1000 °C. However, as soon as no possibility for the removal of heat exists, thermal barrier layers will not help and then ceramic concepts (reinforced) have to be considered.

### Helium Turbine

The key components of concern are: blades, vanes, and the rotor. Gas temperatures exceeding 1000 °C are common in conventional gas turbines. The high temperatures are accommodated by an appropriate choice of materials and the use of advanced cooling systems, bringing the metal temperature down to ~900 °C. Advanced nickel base superalloys produced either with columnar grains (directionally solidified DS) or even as single crystals (SC) are currently employed. These materials are based on an austenitic, solid solution strengthened NiCr-matrix reinforced with coherent $\gamma'$-particles, which are intermetallic compounds of the type $Ni_3Al$. At a material temperature of 1000 °C these superalloys operate at more than 80% of their incipient melting temperature, which means that their temperature capacity has been reached. Such material temperatures can be avoided by appropriate cooling concepts. In addition the surface of the blades can also be coated with a thermal barrier layer (usually $ZrO_2$) providing a further allowance in gas temperature increase of ~150 °C [23]. The application of such gas turbine material concepts to a helium turbine should be quite straight-forward as only the impurities in the He-atmosphere needs further consideration. If material temperatures in excess of 1000 °C are envisaged, superalloys will be at their limits and new blade/vane materials must be considered. Austenitic ODS steels, refractory materials (Mo, W, Nb-based), intermetallic silicides (fibre reinforced or bulky) or SiC/SiC ceramics would then be a solution.

Another critical component is the turbine rotor which has to carry the centrifugal forces from the blades. The situation for the helium turbine is the same as for conventional large land-based turbines. Either a cooled concept is used which allows a 9–12% Cr martensitic steel solution or an uncooled concept with a high temperature resistant material. In this case the ferritic martensitic steels are at their temperature limits, as already discussed in the pressure vessel section. Large ODS-steel forgings are currently impossible to produce, so $\gamma'$-strengthened austenitic super alloy rotors (Udimet 720) are currently under discussion. The problem is that superalloys are designed to resist high temperature deformations which in turn makes them difficult to be properly forged. Unsatisfactory inhomogeneous microstructures with partly inferior mechanical properties are the result as shown in conventional gas turbines. Advanced powder metallurgy techniques could possibly help to overcome these problems, which would also help to accommodate gas temperatures of more than 1000 °C [24].

### VHTR Fuel

Fuel for high temperature reactors consists of TRISO coated fuel particles which are either embedded in spheres (pebble bed reactor) or fuel rods (prismatic core design) [25]. The layered composition of fuel particles being part of a fuel rod is shown in Fig. 2.

The SiC-TRISO fuel design will be the starting point of the VHTR fuel development. It demonstrated excellent performance in former HTR reactor programs mainly in the United Sates and in Germany. Compared with past HTR designs VHTR fuel must comply with the following demands:

- Increasing the operational coolant temperatures from 850 °C up to 1000–1100 °C;
- Increasing the allowable temperature limits in case of accident (beyond 1600 °C);
- Increasing the maximum burn-up from 80 GWd/t up to 150–200 GWd/t;
- Increasing the power density above 6 MW/m$^3$, but still complying with a passive safety approach to remove the decay heat in case of a loss of primary coolant.

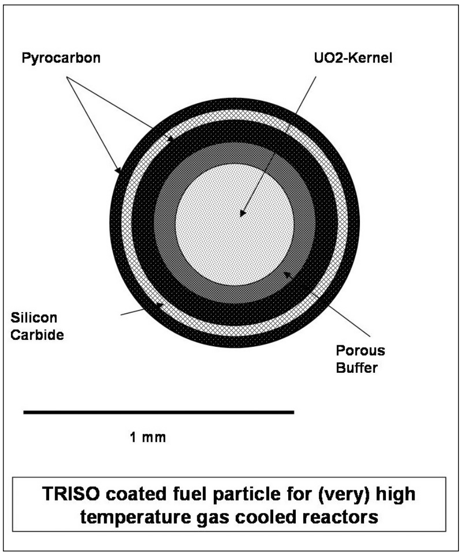

Fig. 2. TRISO coated fuel particles in a hexagonal fuel element

Use of ZrC instead of SiC coatings as in former HTRs fuels enables an increase in power density and an increase in power size under the same coolant outlet temperature and allows for greater resistance against chemical attack by the fission product palladium. The limited fabrication and performance data on ZrC indicates that although it is more difficult to fabricate, it could allow for substantially increased operating and safety envelopes (possibly approaching 1800 °C for emergency cases). Only laboratory-scale fabrication of ZrC-coated particle fuel has been performed to date. Research into more economical commercial-scale fabrication routes for ZrC-coated particle fuels, including process development at production scale, is required. Advanced coating techniques or advanced processing techniques (automation) should be considered. Process development on production-scale coating is required. Irradiation testing and high-temperature heating (safety) tests are needed to define operation and safety envelopes/limits for this fuel, with the goal of high burnup (>10% FIMA, *i.e.* fissions induced in metal atoms) and high-temperature (1300–1400 °C) operation. The facilities used for TRISO-coated particle testing can also be used for ZrC-coated fuel development. These activities would require 10 to 15 years to complete and could be performed at facilities adapted from those available around the world currently used for SiC-based coated particle fuel.

Disposal and reprocessing of VHTR fuel is another important research issue.

Like LWR spent fuel, VHTR spent fuel could be disposed of in a geologic repository after conditioning into the optimum waste form. The current HTGR particle fuel coatings already form an encapsulation for the spent fuel fission products that is extremely resistant to leaching in a final repository. Recycling of LWR and VHTR spent fuel in a symbiotic fuel cycle could achieve significant reductions in waste quantities and radiotoxicity because of the VHTR's ability to accommodate a wide variety of mixtures of fissile and fertile materials without significant modification of the core design. This flexibility was demonstrated in the AVR test reactor in Germany and is a result of the ability of gas reactors to decouple the optimization of the core cooling geometry from the neutronics. A reactor operation with a once-through fuel cycle is envisioned for the first VHTR reactors. For later versions of VHTR derivative reactors, it will be important to close the fuel cycle. For an actinide burning alternative, specific Pu-based driver fuel and transmutation fuel containing minor actinides would have to be developed. This fuel can benefit from the above-mentioned R&D on SiC and ZrC coating but will need more R&D than low enriched uranium (LEU) fuel.

## Hydrogen Production

Balance of plant elements like the intermediate heat exchanger (IHX) or the He-gas turbine are mainly limited by temperature and environmental constraints to the materials, which has already been elaborated on in the Materials Section. We would like therefore to restrict our considerations in this paper to the hydrogen production. The role of hydrogen as future energy carrier is currently heavily disputed. Concepts range from hydrocarbon enrichment of low grade natural fossil gas resources and fuel production from $CO_2$-sources all the way up to direct hydrogen burning *e.g.* in cars. Different possibilities are discussed also for the optimum hydrogen production and distribution concepts (decentralized *vs.* centralized). Mass production of hydrogen will need large amounts of energy and nuclear energy is a very attractive primary energy option. Three different options for use of nuclear energy to produce hydrogen can be considered:

- *Electrolysis*: Electrolysis needs in principle electric energy only. For reactor systems with high outlet temperature (700–900 °C) timely altering demands on electrical energy could be balanced with thermal energy. This would lead to improvements in efficiency and it could contribute to a reduction of production costs.
- *Steam Reforming*: Requirements on natural gas can be significantly reduced by using nuclear heat.
- *Thermo-chemical Processes*: Water splitting through thermochemical processes offers the potential for clean, efficient and cost-effective large-scale production of hydrogen. Among the different thermochemical processes, the sulphur-iodine (SI) process developed by General Atomics already in the 1970s is a prime candidate as it offers a highly efficient and economic hydrogen production path. The SI cycle consists of three main reactions leading in sum to water decomposition into $H_2$ and $O_2$. The first exothermic reaction (called Bunsen reaction) consists in the formation of sulphuric acid and hydrogen iodide from sulphur dioxide and iodine. The second endothermic reaction (operating at 200–300 °C and high pressure) produces hydrogen, while, in the third strongly endothermic reaction (operating at 800–900 °C) $H_2SO_4$ is split into $SO_2$ and water. Sulphuric acid and iodine are strongly oxidizing, while iodide is a strong complexing agent.

Such conditions create an extremely corrosive environment and impose severe demands on materials. Fig. 3 shows a schematic of the sulphur iodine process. Fig. 4 on the other hand shows that the hydrogen production efficiency strongly depends on the process temperature reaching 50–60% at 900–1000 °C.

## Conclusions and Outlook

The very high temperature reactor (VHTR) which is based on current helium cooled HTR concepts could become a very versatile element in future plants for combined cycle electricity and heat generation. It could be particularly useful for mass production of hydrogen which is expected to become an integral part for future sustainable concepts. Some R&D work must be undertaken to allow construction and operation of an advanced VHTR with a direct cycle gas turbine and sulphur-iodine

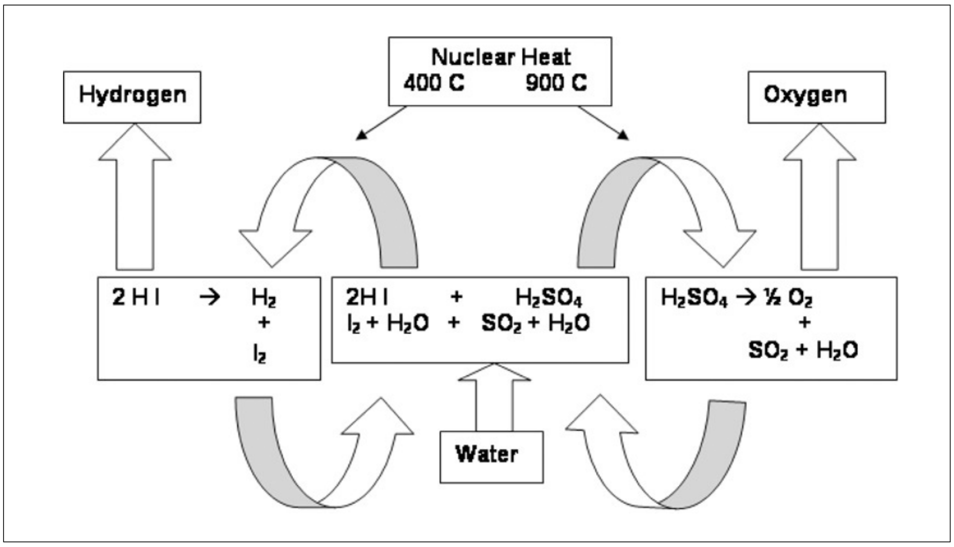

Fig. 3. Schematic drawing of the iodine sulphur process for thermochemical hydrogen production

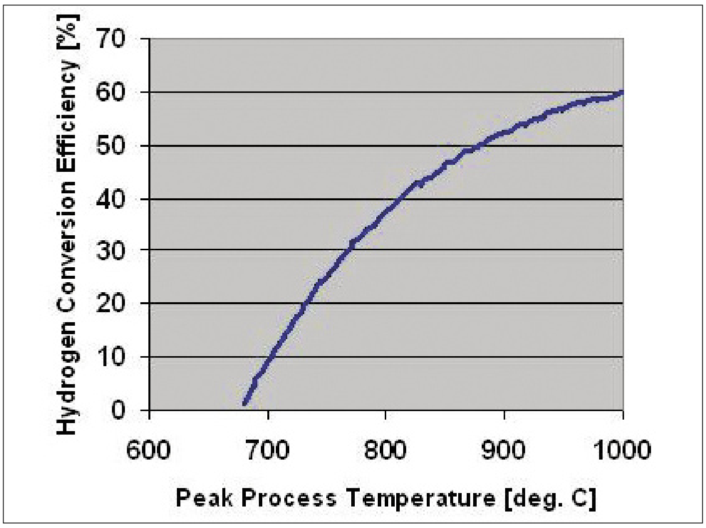

Fig. 4. Efficiency of hydrogen conversion in the thermochemical iodine sulphur process as a function of temperature

hydrogen production operating at gas outlet temperatures of 1000 °C and higher. System R&D work which is currently started within the International Generation IV Initiative could provide the basis for such plants. According to the risk of introducing new technologies it is very likely to assume that the near-term deployable VHTR concepts will operate below 1000 ºC with an indirect cycle. Deployments after 2020 might be direct cycle plants. As far as the fuel cycle is concerned a once-through cycle will be the next concept. Transmutation fuel and actinide burning has to be developed in the future.

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