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Efficiency of hydrogen production systems using alternative nuclear energy technologies

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Abstract

Nuclear energy can be used as the primary energy source in centralized hydrogen production through high-temperature thermochemical processes, water electrolysis, or high-temperature steam electrolysis. Energy efficiency is important in providing hydrogen economically and in a climate friendly manner. High operating temperatures are needed for more efficient thermochemical and electrochemical hydrogen production using nuclear energy. Therefore, high-temperature reactors, such as the gas-cooled, molten-salt-cooled and liquid-metal-cooled reactor technologies, are the candidates for use in hydrogen production. Several candidate technologies that span the range from well developed to conceptual are compared in our analysis. Among these alternatives, high-temperature steam electrolysis (HTSE) coupled to an advanced gas reactor cooled by supercritical CO_2 (S-CO_2) and equipped with a supercritical CO_2 power conversion cycle has the potential to provide higher energy efficiency at a lower temperature range than the other alternatives.

Keywords: Hydrogen production; Nuclear energy; Thermochemical water splitting; High-temperature steam electrolysis; Hybrid process

1. Introduction

It has been suggested that hydrogen should replace petroleum products for fueling automotives to reduce carbon dioxide emissions, limit dependence on imported petroleum, and prepare for the time in which oil reserves would become depleted. Hydrogen is already a significant chemical product, chiefly used in making nitrogen fertilizers and, increasingly, to convert low-grade crude oils into transport fuels. World oil refineries and chemical plants' demand for hydrogen is correspondent to supplying them with about 200 GW of equivalent thermal energy at the time [1]. This energy is approximately equivalent to two-thirds of the US nuclear thermal power capacity. Significant increases in utilization of hydrogen can be expected. The total hydrogen demand in the US is expected to increase by 32% from 2002 to 2007 according to a recent forecast by BCC Research Inc. [2]. Most of the hydrogen is produced near its use site and there is little infrastructure for its distribution. Today, almost all hydrogen is made from natural gas, giving rise to large quantities of carbon dioxide emissions.

The interest in the use of hydrogen in the transportation industry is motivated by both the depletion of fossil fuel resources and the need of drastically reducing the carbon emissions that affect the climate. Both concerns can be addressed by developing energy—efficient and CO_2-free technologies, including either battery- or fuel-cell—operated vehicles. However, the H_2 fuel cells are currently preferred due to better projected energy density and overall efficiency and consequent vehicle performance. Therefore, use of fuel cell vehicles is expected to be the driving force for the increase in hydrogen consumption in the long term.

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Nomenclature

AHTR	Advanced high-temperature reactor
ALWR	Advanced light water reactor
E	Overpotential
ES	Electrolysis
G	Gibbs free energy
GT-MHR	Gas turbine modular helium reactor
H	Enthalpy
HHV	High heating value
HTGR	High-temperature gas reactor
HTSE	High-temperature steam electrolysis
LHV	Low heating value
LWR	Light water reactor
MHR	Modular helium reactor
Q	Heat
SOEC	Solid oxide electrolysis cell
SOFC	Solid oxide fuel cell
SCWR	Super critical water reactor
SFR	Sodium-cooled fast reactor
SI	Sulfur iodine
SMR	Steam methane reforming
UT-3	University of Tokyo-3 (Ca–Br–Fe thermochemical cycle)
VHTR	Very high temperature reactor
WSP	Westinghouse sulfur process

Greek letters

η	Energy efficiency
Δ	Change

Subscripts

el	Electrical
es	Electrolysis
H	Hydrogen
N	Nuclear
pol	Polarizations (ohmic, activation, and mass transfer)
th	Thermal

Many studies have appeared recently concerning future hydrogen demand; for example, one has been presented by Ogden et al. [3]. However, it is somewhat of a speculation to provide a certain time frame for when the H_2 vehicles would win a significant market share. In spite of the uncertainties, it is expected that ultimately the use of hydrogen as an energy carrier will be widely established in the transportation sector, which will give rise to a large-scale hydrogen demand.

Hence, in both the near term and long term, hydrogen demand is expected to increase significantly. There are several energy sources that can be used for hydrogen production, such as nuclear, renewables, and fossil fuels. One of these alternatives is the use of electricity and/or heat from a nuclear plant to break the chemical molecules with hydrogen as a product.

Among other alternatives, using nuclear energy as the primary energy source for hydrogen production is advantageous for two main reasons. The first advantage is that the nuclear reactors do not emit any greenhouse gases (GHG), the most important being CO_2. A large-scale hydrogen infrastructure can prove its environmental merits only if hydrogen is produced by non-GHG emitting methods, such as from nuclear or renewables. The second advantage is that nuclear energy can contribute to large-scale hydrogen production. Given the problem of fast growing energy demand in all sectors in the world, including the transportation sector, large scale, clean hydrogen production will be essential to address this issue, which cannot yet be sufficiently addressed by the renewable energy resources. The limited contribution of the renewables to total energy supply is due to their characteristics of being low-density and intermittent sources.

Efficient use of energy is a significant contributor to any plan for meeting the growing energy demand. Therefore, it is essential to evaluate the alternative technologies of nuclear hydrogen production in terms of their energy efficiency at the production stage. The overall efficiency performance of the alternative routes for using nuclear energy for this purpose depends on the operating temperature, conversion efficiency of the processes, and complexity of the plant systems. The overall choice will depend on the cost of the process, which means the cost of the equipment, as well as the efficiency. Therefore, efficiency is a factor that also determines the cost of the technology.

In this paper, we examine several alternatives for coupling nuclear and hydrogen production technologies, in terms of their thermal-to-hydrogen energy efficiency, and we do not present the cost analysis for each technology. A preliminary cost analysis of alternative routes for nuclear hydrogen production can be found in the report by Yildiz and Kazimi [4]. We emphasize that higher temperatures will enable more efficient thermochemical and electrochemical hydrogen producing reactions. Consequently, the nuclear technologies capable of producing reactor coolant temperatures on the order of 700 °C or higher would be suitable for hydrogen production. Nevertheless, high-temperature operation of both the nuclear plant and the hydrogen plant imposes stringent heat transfer-associated requirements that can differ among the technology alternatives. These necessitate materials development and intricate design requirements for integrated plant layouts, which consequently affect the cost of each technology.

The rest of the article is structured as follows. Section 2 explains the alternative nuclear and the coupling power conversion technologies that can be utilized for providing the primary energy source for hydrogen production. Section 3 analyzes alternative hydrogen production technologies that can be integrated to relevant nuclear technologies. Section 4 provides the concluding remarks and indicates the necessary future research areas.

2. Nuclear technologies for hydrogen production

Hydrogen can be produced by thermochemical and/or electrochemical processes using nuclear energy as the primary thermal energy source. The hydrogen production process properties determine the types of reactors that can appropriately be coupled to the relevant hydrogen production technology. The first important design requirement for both thermochemical and electrochemical hydrogen production is the high temperature needed for achieving high thermal-to-hydrogen energy efficiency. This is an important factor in the economics of the technologies. Another important design objective is attaining high thermal to electrical power conversion efficiency, which is essential for the efficiency and economics of specifically the electrochemical hydrogen production. Some other general requirements for a good match to the hydrogen plant are enabling effective heat transfer rate to the chemical plant with minimum temperature reduction of the reactor coolant, minimizing the pressure losses in the primary loop (or intermediate loop if one is used), ensuring safety by choosing chemically inert coolants and reducing the potential for power to flow mismatch in the reactor, and low capital costs. Since they can reach the required high temperatures, gas-cooled reactors, molten-salt-cooled reactors, and heavy-metal-cooled reactors, all with gas power cycles in the case for electrolysis, appear to be the most promising technologies to be coupled to the hydrogen plants for efficient production. The same conclusion concerning the promising choices of nuclear reactor technology is also affirmed in Refs. [6,8].

2.1. Candidate nuclear reactor technologies

The following is a brief description of the current candidate nuclear technologies for hydrogen production that we consider in this paper.

(a) *Modular helium reactor* (*MHR*)

The MHR is a thermal reactor that can be used both for hydrogen and electricity production in modules of 600 MWth. Its core consists of prismatic blocks of graphite that allow coolant flow and contains ceramic fuel such as the operating Japanese HTGR [5]. The temperature of the coolant, He gas, at the reactor exit is currently designed to achieve temperatures around 850 °C. The MHR has been based upon the demonstrated technologies of German AVR and the US Fort St. Vrain reactors. It is proposed to achieve 1000 °C in the future within a new design with the same reactor concepts called the very high temperature reactor (VHTR). The operating pressure of the MHR is 7 MPa. The core design can provide passive safety by accomplishing high temperatures during transients and by large thermal inertia. For hydrogen production, the system can supply heat that could be used efficiently by a thermochemical sulfur–iodine process or the Westinghouse sulfur process. The system can incorporate electricity generating equipment, i.e., He direct-cycle power conversion system, to meet cogeneration needs [6,7].

(b) *Advanced high-temperature reactor* (*AHTR*)

The AHTR is an advanced thermal reactor concept that is proposed to address the requirements specific to the H_2 production process [8], and has not been built yet. It can be integrated with a power conversion cycle also for electricity production. The AHTR uses the solid coated-particle fuel in a graphite-matrix like the MHR, but a molten-fluoride-salt as coolant. It combines the high-temperature fuel from the HTGR with a more dense coolant as of the molten salt reactor. The proposed design temperature of the coolant at the reactor exit is 1000 °C. The graphite blocks are compatible with fluoride salts as coolant. The reactor concept is designed for atmospheric pressure operation. This design uses Ni-based high-temperature alloys that have been similarly adopted for molten salts. The reactor is proposed to be built in large sizes (2000 MWth) with passive safety systems for decay heat removal.

(c) *Advanced gas reactor* (*AGR*)

The AGR is an already commercial thermal reactor that has been built in the UK for electricity production in 1550 MWth units, with 14 units still in operation [9]. The AGR core consists of uranium oxide fuel pellets in stainless-steel cladding within graphite blocks. The graphite acts as moderator and carbon dioxide is the coolant. The achievable temperature of the coolant at reactor exit at normal operation is around 650 °C. The carbon dioxide circulates through the core at 4.3 MPa. For future design and implementation, there is the potential to increase the operating pressure of the AGR in order to couple it to a direct cycle supercritical CO_2 power conversion system. The temperature of the reactor coolant for a future design can be driven up to 750 °C after a new design analysis. This combination can enable high efficiency, economical hydrogen production through steam electrolysis at medium temperatures.

(d) *Secure transportable autonomous reactor* (*STAR-H_2*)

The STAR-H_2 is a fast neutron spectrum, 400 MWth modular-sized reactor. The STAR itself can be used for both electricity and hydrogen production. STAR is based on Russian submarine reactor technology demonstrated at about 500 °C, but has not been built commercially. The reactor coolant is liquid lead (Pb), with reactor core outlet temperature at 800 °C for future design at atmospheric pressure. It is a low power density natural circulation cooled reactor with passive load following and passive safety response characteristics [10]. The 400 MWth sizing retains natural circulation capability in a rail shippable reactor vessel size as well as allowing for passive decay heat removal. The reactor is proposed to operate on a 15-year whole core cassette refueling interval using uranium/transuranic nitride fuel. The thermal energy from the STAR-H_2 can drive the UT-3 thermochemical process hydrogen production.

(e) *Sodium-cooled fast reactor* (*SFR*)

The sodium-cooled fast reactor (SFR) system features a fast-spectrum, sodium-cooled reactor and a closed fuel cycle for efficient management of actinides and conversion of fertile uranium [11]. SFR technology has a solid base of demonstration in France, Russia and the US, with reactor core outlet temperature at 500 °C. The concept future designs are developed for electricity production. There are two major future design options: One is an intermediate size (150–500 MWe) sodium-cooled reactor with uranium–plutonium–minor-actinide–zirconium metal alloy fuel, supported by a fuel cycle based on pyrometallurgical processing in facilities integrated with the reactor. The second is a medium to large (500–1500 MWe) sodium-cooled reactor with mixed uranium–plutonium oxide fuel, supported by a fuel cycle based upon advanced aqueous processing at a central location serving a number of reactors. The outlet temperature is approximately 550 °C for both at atmospheric pressure. This reactor is proposed to derive low-temperature thermochemical processes, such as Cu–Cl, for hydrogen production.

2.2. *Candidate power conversion systems*

For power conversion systems, the following two choices show potential due to their high efficiency at medium- to high-temperature operation.

(a) *Helium gas turbine power conversion system*

The high-temperature operation of the MHR enables a high efficiency power conversion system with helium as the working fluid. The MHR uses a direct cycle but an indirect cycle has also been proposed by other designers. The gross thermal efficiency (not the net efficiency) of this system is recorded as 48% with 900 °C of reactor exit temperature [12]. The South African electricity company ESKOM estimates the net efficiency of this design to be 42% [13, p. 93]. It can be coupled to the MHR to make the GT-MHR system [6,7] or to the AHTR, for electricity production.

(b) *Supercritical CO_2 (S-CO_2) gas turbine power conversion system*

The S-CO_2 recompression Brayton cycle has the potential to achieve high cycle efficiencies at lower temperature than that of the He cycle. Dostal et al. [13] records the net cycle efficiency as 45% for turbine inlet temperature of 550 °C at 20 MPa. It is a very compact and simple system, hence is promising economically, but has not yet been built. It can be directly coupled to an AGR to make the S-CO_2–AGR system, or indirectly to other nuclear reactor technologies.

3. Hydrogen production technologies using nuclear energy

Nuclear energy can be used in hydrogen production mainly in three ways:

- By using the electricity from the nuclear plant for conventional liquid water electrolysis.
- By using both the high-temperature heat and electricity from the nuclear plant for high-temperature steam electrolysis or the hybrid processes.
- By using the heat from the nuclear plant for thermochemical processes.

The technology options for large-scale production of hydrogen using nuclear energy are presented in this section. Fig. 1 illustrates the map for coupling the possible hydrogen and nuclear technologies that can be considered. Fig. 1 shows the range of all advanced nuclear technology alternatives that can drive the thermochemical, electrochemical and hybrid processes of hydrogen production. The nuclear alternatives in this figure cover more than those presented in Section 2. The reason for this is that Fig. 1 presents also the water electrolysis option for hydrogen production that can be driven by low-temperature reactors, too. Water electrolysis, even though a commercially proven technology, may not present an energy-efficient centralized hydrogen production method for the future if supported by only the light water-cooled reactors. On the other hand, high-temperature steam electrolysis does have good energy efficiency. As a result, we do not include some of the low-temperature advanced reactor concepts that can be used only for water electrolysis in the list of promising nuclear technologies for hydrogen production in Section 2.

The following sections provide technical features and the energy efficiency assessment of the stated technologies in Fig. 1. Table 1 presents an overview of nuclear hydrogen production technologies. As seen from Table 1, the water electrolysis coupled to an advanced reactor is likely to be the least energy efficient, and consequently the most energy expensive alternative. Hence, this paper does not include any analysis of the water electrolysis path and concentrates on the more promising long-term future options of nuclear hydrogen production.

3.1. *Thermochemical processes*

Thermochemical processes for hydrogen production involve thermally assisted chemical reactions that release the hydrogen e.g. from hydrocarbons or water. The most widespread thermochemical process for hydrogen production is the steam methane reforming (SMR) process. While this technology is the most economic today, it yields considerable carbon dioxide emissions. Therefore we do not favor SMR for a long-term hydrogen economy. Alternative thermochemical processes are those which do not have

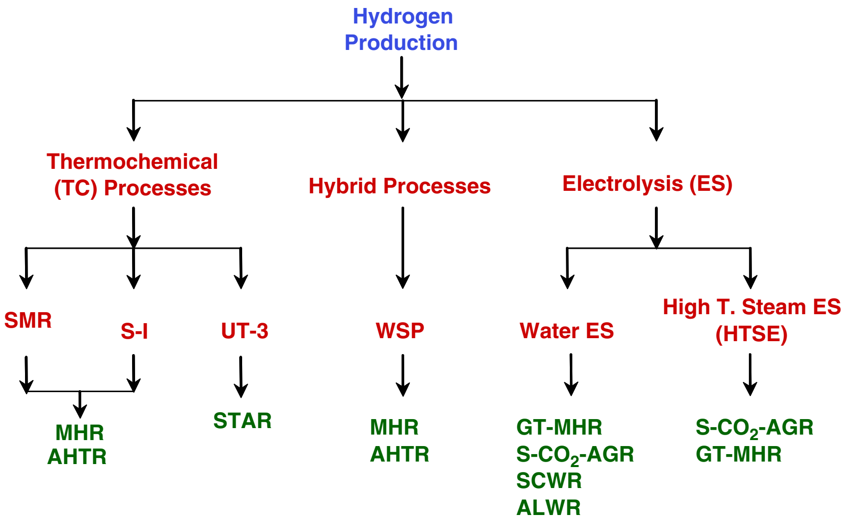

Fig. 1. Technology options for nuclear hydrogen production.

Table 1
Overview of nuclear hydrogen production technologies

Approach	Electrochemical		Thermochemical	
Feature	Water electrolysis	High-temperature steam electrolysis	Steam-methane reforming	Thermochemical water splitting
Required temperature, (°C)	< 100, at P_{atm}	> 500, at P_{atm}	> 700	> 800 for S-I and WSP > 700 for UT-3 > 600 for Cu–Cl
Efficiency of the process (%)	85–90	90–95 (at $T > 800\,^{\circ}\mathrm{C}$)	> 60, depending on temperature	> 40, depending on TC cycle and temperature
Energy efficiency coupled to LWR, or ALWR%	~ 27	~ 30	Not feasible	Not feasible
Energy efficiency coupled to MHR, ALWR, ATHR, or S-AGR (%)	> 35	> 45, depending on power cycle and temperature	> 60, depending on temperature	> 40, depending on TC cycle and temperature
Advantage	+ Proven technology	+ High efficiency + Can be coupled to reactors operating at intermediate temperatures + Eliminates CO_2 emission	+ Proven technology + ReducesCO_2 emission	+ Eliminates CO_2 emission
Disadvantage	– Low energy efficiency	– Requires development of durable, large-scale HTSE units	– CO_2 emissions – Dependent on methane prices	– Aggressive chemistry – Requires very high temperature reactors – Requires development at large scale

hydrocarbon feedstock but which split water into hydrogen and oxygen through a series of thermally driven chemical reactions. This approach is called the thermochemical water splitting process. The purpose is to generate hydrogen at lower temperatures than that for pyrolysis of water, which takes place at temperatures greater than 4000 °C. In the long term, the low cost of water itself and avoiding CO_2 emissions totally are the main benefits of the water splitting approach. A recent screening study [14] has identified two thermochemical water splitting cycles which have the highest commercialization potential, and practical applicability to nuclear heat sources. These are the sulfur–iodine (SI) and calcium–bromine–iron (UT-3) cycles. The SI cycle is being investigated by General Atomics and Japanese Atomic Energy Research Institute. The UT-3 cycle, which is called 'UT-3' in recognition of its origin at the University of Tokyo, is being investigated by JAERI. Argonne National Laboratory (ANL) is currently working on achieving a thermochemical water splitting process at lower temperatures than the SI and UT-3 cycles. ANL has identified the copper–chlorine (Cu–Cl) thermochemical cycle for this purpose [15]. The currently commercial steam methane reforming technology, which is not a CO_2-free option, can also be coupled to a nuclear source for near-term application. This technology poses a higher near-term implementation potential due to the proven operation of the method.

The thermochemical processes can involve stringent heat transfer requirements. The first one is that effective heat transfer be accomplished at high temperature, since the driving factor for the process efficiency is mainly the operating temperature. The second is maintaining the operating peak temperature of the hydrogen plant sufficiently close to the reactor exit temperature while trying to achieve a bulk temperature in the thermochemical decomposer as high as possible. This can help to attain a nearly uniform temperature distribution at high values in the decomposer for more homogeneous and faster reaction rates and to decrease the losses. The accomplishment of these requirements differs among the various options.

In the following, we briefly discuss the alternative thermochemical processes that can use nuclear energy as the primary heat source.

(a) *Steam methane reforming* (*SMR*)

Steam methane reforming (SMR) is currently the most common commercial technology for hydrogen production. The SMR process requires high process temperature, and the most common practice for providing the needed heat for the process is via burning natural gas. The process is as follows:

$$\text{Reforming: } CH_4 + H_2O \rightarrow CO + 3H_2, \quad \text{endothermic } (750\text{–}800\,^\circ\text{C}),$$

$$\text{Shift: } CO + H_2O \rightarrow CO_2 + H_2, \quad \text{exothermic } (350\,^\circ\text{C}).$$

High-temperature reactors can substitute for the natural gas burning furnaces as a heat source. This approach reduces the CO_2 emissions to the atmosphere in large quantities.

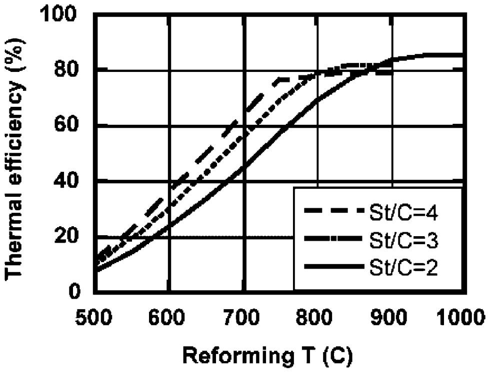

Fig. 2. Energy efficiency of the steam methane reforming process for different steam/carbon ratios [17].

Nevertheless, due to the nature of the chemical reforming and shifting processes, there is still a need for natural gas feedstock and consequently CO_2 would still be emitted.

The efficiency of the SMR process, $\eta_{H,SMR}$, can be described as the fraction of energy supplied that can be recovered by burning the product, i.e. hydrogen [16]:

$$\eta_{H,SMR} = \frac{n_H HHV_H}{HHV_{CH_4} + Q_{in,SMR}}, \tag{1}$$

where n_H is the number of moles of hydrogen produced from one mole of fuel, methane (CH_4), and HHV_k[1] is the high heating value per mole of species k. This definition takes into account the heating value of the fuel and the external heat added to the process as the primary energy supplied for producing hydrogen. The steam-to-carbon ratio (St/C) is an important factor affecting the total thermal energy supplied for SMR, $Q_{in,SMR}$. The ideal value of St/C is 2. However, most reformers run at larger values of St/C in order to prevent coking and enhance the reaction progress. Consequently, the increased St/C enhances the process efficiency at medium temperatures and decreases the efficiency somewhat at higher temperatures compared to the efficiency with $St/C = 2$. The theoretical efficiency of this process as a function of temperature and St/C as calculated in [16,17] is shown in Fig. 2.

In the near term, the SMR process can potentially be coupled to the high-temperature helium-cooled reactor, the MHR, thus making an MHR–SMR system. The MHR can function as the high-temperature heat source operating at about 850 °C, to replace the natural gas burning furnace. The high operating temperature can enable the process to

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[1] HHV of hydrogen is used in the energy efficiency calculations for all the processes considered in our analysis. Hence, it allows a consistent comparison of the resulting efficiency values between the different technologies. Converting the resulting efficiency values to those based on the LHV of hydrogen would involve scaling the results from HHV to LHV.

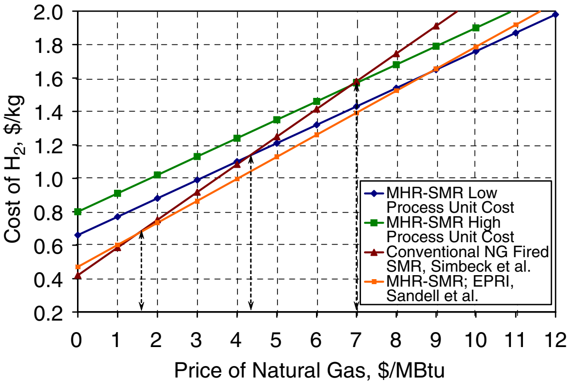

Fig. 3. Cost of hydrogen production by using the MHR–SMR and conventional SMR technologies [4].

take place at about 80% efficiency. This alternative has been estimated to be potentially cost competitive in the near future with the conventional SMR process [4,18]. Fig. 3 shows the boundary of our cost estimate for hydrogen production via the MHR–SMR as a function of natural gas price [4]. We note that the cost estimates in Fig. 3 do not include any penalty or taxation concerning the CO_2 emissions due to the nature of the technology. This figure indicates that, for natural gas prices greater than $4.5–7/MMBtu, the MHR–SMR technology would be economical and more competitive than the conventional SMR technology. In the last year, the long-term price of natural gas supplies has risen considerably, which suggests that $4.5–7/MMBtu is a reasonable near-term range for the cost of natural gas that can be used in the SMR process. Earlier, an EPRI estimate of the hydrogen cost via the MHR–SMR technology indicated that the price of natural gas which could enable the MHR–SMR technology to be economically competitive is around $1.5/MMBtu. Therefore, according to the EPRI analysis, MHR–SMR is already more attractive than the conventional SMR technology for economical hydrogen production. Our analysis is based on a different set of estimates for the rate of return of invested capital and takes into consideration a range of potential process efficiencies.

(b) *Sulfur–iodine* (*SI*) *cycle*

The sulfur–iodine (SI) cycle was proposed by General Atomics in the mid-1970s. It consists of the following three chemical reactions which yield the dissociation of water [7]:

$$I_2 + SO_2 + 2H_2O \rightarrow 2HI + H_2SO_4 \quad (120\,°C),$$

$$H_2SO_4 \rightarrow SO_2 + H_2O + 1/2O_2 \quad (830\text{–}900\,°C),$$

$$2HI \rightarrow I_2 + H_2 \quad (300\text{–}450\,°C).$$

The whole process takes in water and high-temperature heat, and releases hydrogen and oxygen. All reactions are in fluid interactions. All reagents are to be recycled; there are no effluents. Each of the chemical reactions in this process was demonstrated in the laboratory [11]. Japan Atomic Energy Research Institute has also worked on the research, development and demonstration of the SI cycle [19,20]. Decomposition of sulfuric acid and hydrogen iodide involve aggressive chemical environments. Hence, the material candidates for the SI cycle hydrogen plant should be chosen carefully to accommodate corrosion problems. A schematic of the SI flowsheet is shown in Fig. 4.

As seen from this figure, heat transfer is necessary at each step of the cycle, along with internal recuperation. If heat losses can further be eliminated by using highly effective and compact heat exchangers, the energy efficiency of the process will be enhanced, contributing to better economics. Fig. 5 represents the energy efficiency of this process with the current design conditions. Energy efficiency, $\eta_{H,SI}$, in this figure is defined as the ratio of the energy (in terms of HHV of hydrogen, HHV_H) that is carried by a unit amount of product, $Q_{H,out}$, to the total thermal energy demand by the process, $Q_{in,SI}$, to produce a unit amount of product hydrogen [21]:

$$\eta_{H,SI} = \frac{Q_{H,out}}{Q_{in,SI}} = \frac{HHV_H}{Q_{in,SI}}. \qquad (2)$$

The SI cycle can be coupled to the modular high-temperature reactor (MHR) [6]. MHR–SI can have a range of operating temperatures. With the current plant design concept, in the near term the SI cycle operation peak temperature could correspond to approximately 827 °C [12]. If a more effective heat recuperation configuration and better heat exchanger materials are achievable, the peak and bulk operating temperatures of the process can be higher, enhancing the energy efficiency of the process.

A new reactor technology that can also be coupled to the SI cycle is the advanced high-temperature reactor (AHTR). This advanced reactor concept is designed specifically for H_2 production processes, as presented in Section 3, particularly for the SI cycle. It has the capability to provide heat at high temperature over a small temperature difference between the inlet and exit of the heat exchange area. This is due to good heat transfer capabilities of molten salt coolant of AHTR.

(c) *Ca–Br–Fe* (*UT*-3) *cycle*

The UT-3 cycle was first developed by the University of Tokyo. It involves solid–gas interactions which may facilitate the reagent–product separations, as opposed to the all-fluid interactions in the SI cycle. It is formed of the following reactions [22,23]:

$$CaBr_2 + H_2O \rightarrow CaO + 2HBr \quad (730\,°C),$$

$$CaO + Br_2 \rightarrow CaBr_2 + 1/2O_2 \quad (550\,°C),$$

$$Fe_3O_4 + 8HBr \rightarrow 3FeBr_2 + 4H_2O + Br_2 \quad (220\,°C),$$

$$3FeBr_2 + 4H_2O \rightarrow Fe_3O_4 + 6HBr + H_2 \quad (650\,°C).$$

The thermodynamics of these reactions have been found favorable. However, the hydrogen production efficiency of the process is limited to about 40% due to the melting point

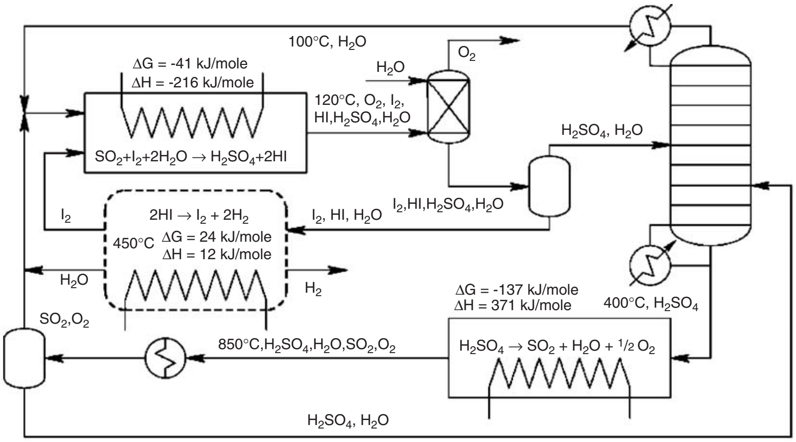

Fig. 4. Overview schematic of SI flowsheet [7].

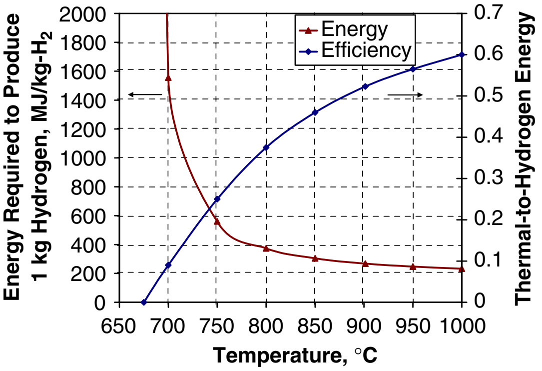

Fig. 5. Thermal-to-hydrogen efficiency [7], and the thermal energy required to produce 1 kg of hydrogen using the MHR–SI.

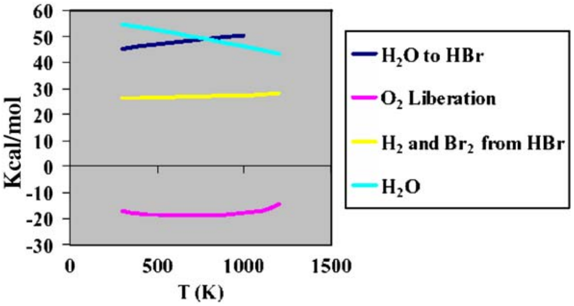

Fig. 6. Gibbs free energies for the calcium–bromine cycle [23].

of $CaBr_2$ at 760 °C [11]. Fig. 6 shows the free energies involved in this thermochemical cycle as studied at ANL, which show the viability of the processes.

There is ongoing work at Argonne National Laboratory to couple the UT-3 cycle to the secure transportable autonomous reactor (STAR). The operation of the STAR, which is a liquid-metal reactor, above 600 °C requires especially extensive materials development.

(d) *Cu–Cl cycle*

Argonne National Laboratory's (ANL) Chemical Engineering Division is studying the copper–chlorine thermochemical cycle. This cycle is expected to operate at 500 °C to produce hydrogen (and oxygen)—a temperature compatible with current power plant technologies [24], such as the sodium-cooled fast reactor (SFR).

If proven, it will have the advantages that corrosion issues are more tractable at 500 °C than at higher temperatures as those for the SI and UT-3 cycles. The energy efficiency of the process is projected to be 40–45%. This work is currently investigated only by ANL, at a counter top level. Preliminary studies of this cycle indicate that the reactions should be as follows [15]:

$$2Cu + 2HCl \rightarrow 2CuCl + H_2 \quad (450\,°C),$$

$$4CuCl + 4Cl^- \rightarrow 4CuCl_2^- \quad (30\,°C),$$

$$4CuCl_2^- \rightarrow 2CuCl_2 + 2Cu + 4Cl^- \quad (30\,°C),$$

$$2CuCl_2(aq) \rightarrow 2CuCl_2(s) \quad (100\,°C),$$

$$2CuCl_2 + H_2O \rightarrow CuO + CuCl_2 + 2HCl \quad (400\,°C),$$

$$CuO + CuCl_2 \rightarrow 2CuCl + 1/2O_2 \quad (500\,°C).$$

The free energies are plotted versus temperature in Fig. 7. It is concluded that all of these reactions are thermodynamically viable based on the values of the free energies.

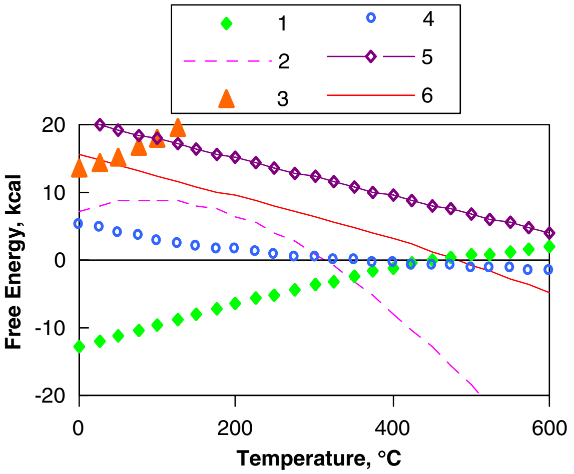

Fig. 7. Gibbs free energy changes associated with the Cu–Cl process reactions [15].

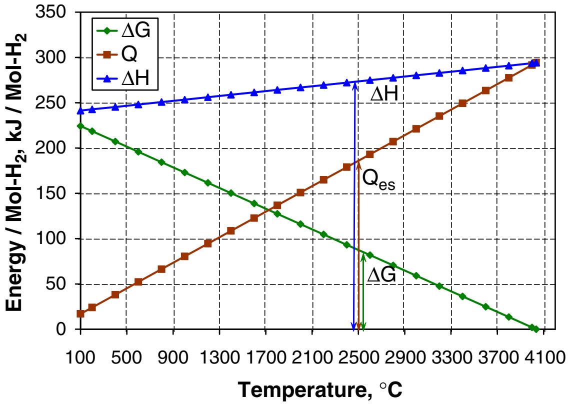

Fig. 8. Energy required for steam electrolysis.

3.2. Electrochemical processes

Electrolysis of water or steam at different temperatures can lead to the decomposition of water. It is a proven and currently commercial technology for hydrogen production. Nevertheless, it involves high electrical energy consumption that renders the process expensive. Therefore, for future production schemes, we consider high-temperature steam electrolysis to be the more energy efficient and clean way of producing hydrogen.

(a) *High-temperature steam electrolysis* (*HTSE*)

High-temperature steam electrolysis (HTSE) is the electrolysis of steam at high temperatures. The total energy required, ΔH, which is composed of the required thermal energy, Q, and the Gibbs free energy (electrical energy demand) ΔG, for the steam electrolysis is shown in Fig. 8. The total energy increases slightly with temperature. The electrical energy demand, ΔG, decreases with increasing temperature leading to increased direct heat requirement, as presented in this figure. The decrease in electrical energy demand drives the thermal-to-hydrogen energy conversion efficiency to higher values. This is one of the primary advantages of HTSE. The higher temperature also favors electrode activity and helps lower the cathodic and anodic over-voltages. Therefore, it is possible to increase the electric current density at higher temperatures and consequently lower the polarization losses, which yields an increase in the process efficiency. Thus, the HTSE is advantageous from both thermodynamic and kinetic standpoints.

The materials of the HTSE cell can be made of ceramics, which avoid corrosion problems. High-temperature steam electrolysis using ceramic electrolysis cells is representative of the new advanced technologies. The reaction scheme in the HTSE process is the reverse of that in a solid oxide fuel cell (SOFC), which is being developed vigorously for application in the power industry. Water vapor molecules are dissociated at the porous cathode, producing an enriched H_2O/H_2 mixture, while the oxygen ions are transported through the nonporous, ion-conducting solid electrolyte to the porous anode where they recombine. Thus the product gases hydrogen and oxygen are automatically separated by the solid electrolyte membrane. Only the gases H_2O, O_2, and H_2 have to be circulated in the electrolysis plant and no other chemicals are involved that could give rise to safety or environmental problems. Fig. 9 illustrates a representative electrolysis cell for high-temperature steam electrolysis that is being tested at the Idaho National Engineering and Environmental Laboratory (INEEL) [25]. Such a system can be named a solid oxide electrolysis cell (SOEC). The specific materials for the electrodes and electrolyte and the geometry of the unit cell can change depending on the operating temperature in order to provide optimized performance. The HTSE process is particularly advantageous when coupled to high efficiency power cycles and can consequently yield very high overall thermal-to-hydrogen efficiency. The steam electrolysis concept can be coupled to a range of nuclear technologies, such as supercritical water-cooled reactors, gas-cooled reactors, lead–bismuth-cooled reactors, and molten-salt-cooled reactors, all of which can deliver relatively high temperatures and high net power cycle efficiencies. This feature makes the HTSE option very advantageous. Here, we assume that the HTSE cells that can efficiently operate at intermediate temperatures (650–800 °C) can become available, as well as those that can operate at higher temperatures (> 800 °C) in the near term. This assumption is based on the ongoing positive research for developing well performing SOFC materials for intermediate temperature range.

The net thermal-to-hydrogen energy efficiency of the nuclear-assisted HTSE is governed critically by the operating temperature and the power cycle efficiency. In this assessment, we adopt a design where the heat from the nuclear reactor is divided into two flow paths such that one path feeds heat into the power conversion system for

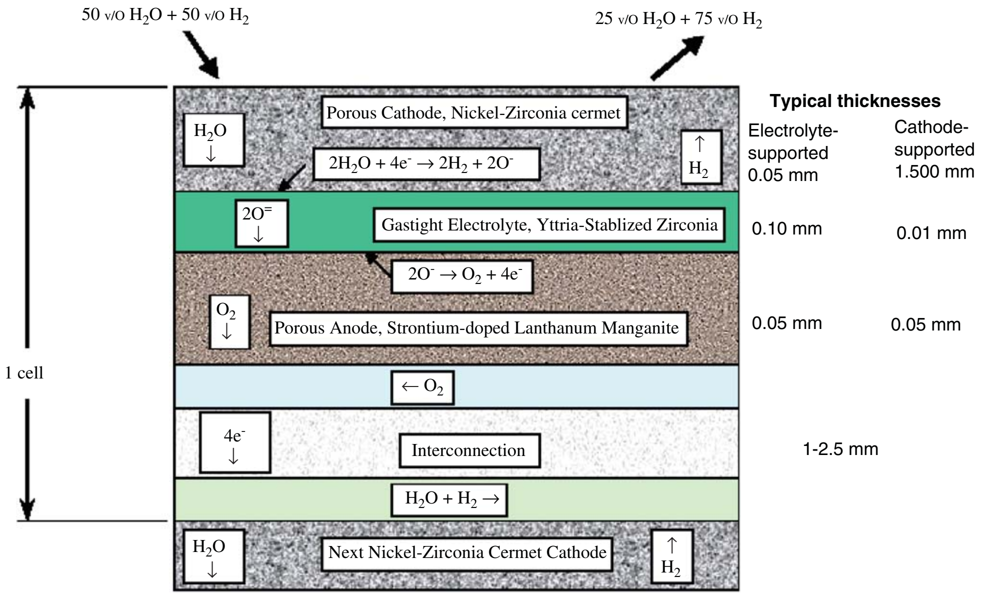

Fig. 9. Schematics for representative solid oxide electrolysis cell [25].

the electricity generation and the other path feeds heat into the HTSE process. Due to the small-size restriction of the HTSE cells in attaining high efficiency, we assume that the large-scale production of hydrogen is accomplished by a series of HTSE cell modules in one hydrogen plant. It is assumed that thermal energy (in addition to the electrical energy) necessary for splitting steam at a given temperature and the heat generated due to polarization-related irreversibilities are equivalent and no net heat generation exists in the cell. Consequently, the change of operation temperature from inlet to the outlet of the HTSE modules is assumed negligible. Therefore, in this analysis we assume that the coolant outlet, the power turbine inlet and the HTSE operation temperatures are equal.

The total energy demand for steam electrolysis, ΔH, for producing a unit amount of hydrogen within the electrolysis process is represented as the sum of the electrical energy demand (Gibbs free energy change) ΔG, and the thermal energy demand, Q_{es}. This relation is also represented in Fig. 8.

$$\Delta H = \Delta G + Q_{\mathrm{es}}. \tag{3}$$

The electrical energy that is used in the electrolysis is obtained from the nuclear power plant itself. Therefore, ΔG can be expressed as

$$\Delta G = \eta_{\mathrm{el}} \times Q_{\mathrm{N,el}}, \tag{4}$$

where η_{el} is the net electric power cycle efficiency and $Q_{\mathrm{N,el}}$ is the thermal energy from the nuclear reactor used for electricity production in the power cycle.

The thermal power of the nuclear reactor is consumed both for electricity production and for heating the steam to the desired electrolysis temperature. Therefore, the total thermal energy extracted from the nuclear reactor is the sum of the heat necessary for the two processes. Assuming loss-free use of reactor thermal energy in the heating process, we get

$$Q_{\mathrm{N}} = Q_{\mathrm{N,el}} + Q_{\mathrm{es}} = Q_{\mathrm{N,el}} + Q_{\mathrm{N,es}}. \tag{5}$$

The ideal value of the overall thermal-to-hydrogen production efficiency from HTSE, $\eta_{\mathrm{H,HTSE,ideal}}$, is the ratio of the amount of energy that is carried by unit amount of hydrogen produced, $Q_{\mathrm{H,out}}$ (in terms of HHV of hydrogen, $\mathrm{HHV_H}$) to the total thermal energy required in the steam electrolysis process to produce the unit amount of hydrogen.

$$\eta_{\mathrm{H,HTSE,ideal}} = \frac{Q_{\mathrm{H,out}}}{Q_{\mathrm{N,el}} + Q_{\mathrm{N,es}}} = \frac{\mathrm{HHV_H}}{\Delta G/\eta_{\mathrm{el}} + Q_{\mathrm{N,es}}}. \tag{6}$$

Practically, there will be energy losses that we denote as Q_{loss} which account for the polarization and ohmic losses. Therefore, the actual amount of energy required for producing a unit amount of hydrogen via HTSE should take into account the relevant loss which should be supplied as heat initially. Finally, the actual thermal-to-hydrogen energy production efficiency of the HTSE process, $\eta_{\mathrm{H,HTSE}}$, that we use in our evaluation takes the form

$$\begin{aligned}\eta_{\mathrm{H,HTSE}} &= \frac{Q_{\mathrm{H,out}}}{\Delta G/\eta_{\mathrm{el}} + Q_{\mathrm{N,es}} + Q_{\mathrm{loss}}} \\ &= \frac{\mathrm{HHV_H}}{\Delta G/\eta_{\mathrm{el}} + Q_{\mathrm{N,es}} + Q_{\mathrm{loss}}}, \\ &\quad \text{where } Q_{\mathrm{loss}} = \frac{\sum E_{\mathrm{pol}}}{\eta_{\mathrm{el}}}.\end{aligned} \tag{7}$$

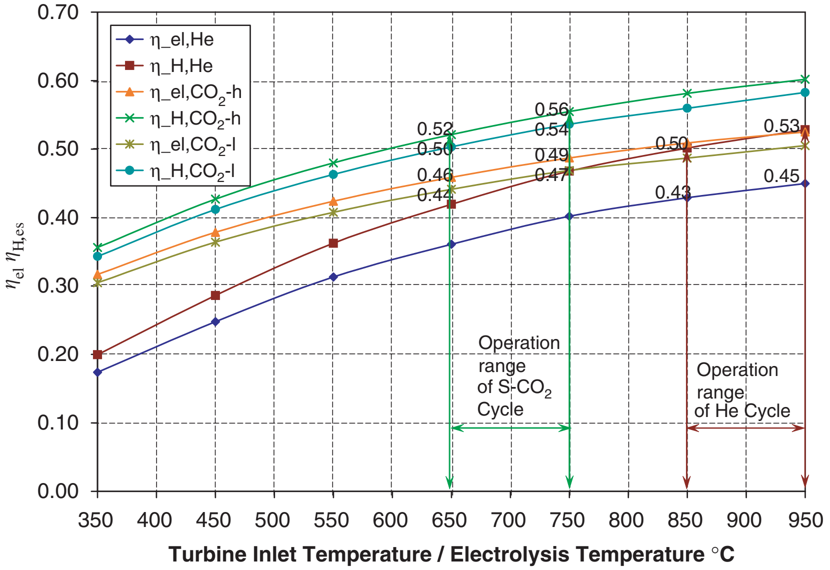

Fig. 10. Power cycle net efficiency and thermal-to-hydrogen energy efficiency for the GT-MHR–HTSE and AGR–S-CO$_2$–HTSE technologies [4].

As the electrolysis temperature increases, ΔG and Q_{loss} decrease sufficiently such that the overall thermal energy required for the electrolysis that is represented in the denominator of Eq. (7) decreases. Therefore, the overall thermal-to-hydrogen energy efficiency increases with increasing operating temperatures. This feature makes HTSE a technically attractive approach in hydrogen production using nuclear heat.

Eq. (7) represents the actual thermal-to-hydrogen energy efficiency of a hydrogen production cycle where the heat from the H_2 and O_2 off-gas stream is not recuperated. We can also recuperate the heat from the off-gas stream to help preheat the H_2O to be fed into the high-temperature steam electrolysis unit. Consequently, the use of excess heat in the H_2 and O_2 gas streams helps reduce the amount of nuclear energy required to heat the water vapor to the required electrolysis temperature. The reduced energy to be supplied by the nuclear reactor for producing the same amount of hydrogen means that the overall thermal-to-hydrogen efficiency of the cycle is increased. However, in this analysis, only the direct cycle without off-gas heat recuperation has been considered.

Two alternatives are considered for both the nuclear reactor and the electrical power cycle in this work at the preliminary evaluation stage. The first is the MHR with He power conversion cycle, that makes the GT-MHR–HTSE system. This approach has also started to be examined at INEEL [26,27]. The second alternative is the AGR with the supercritical CO_2 power conversion cycle that makes the S-CO_2–AGR–HTSE system. The second is a proposal by us to couple an advanced version of the AGR to a direct supercritical CO_2 (S-CO_2) power conversion cycle. The S-CO_2 power conversion cycle has recently been proposed also for fast gas reactors [13]. This proposed technology necessitates upgrading the currently operating AGR design from 4.3 MPa to the operating pressure at the inlet of the S-CO_2 turbine, which is 20 MPa.

The coupling of GT-MHR and HTSE, GT-MHR–HTSE, can provide high-efficiency hydrogen production due to the high operating temperature of the GT-MHR. Fig. 10 shows the net electrical efficiency of the He power cycle as estimated from the relevant thermal efficiency, η_{th}, results of Dostal et al. In order to obtain an estimate of the net electrical power cycle efficiency, η_{el}, we consider 13% fractional reduction over the theoretical thermal efficiency to remain consistent with the η_{el} estimated by Wang et al. [28], in taking into account the losses due to friction, leakage and component cooling.

The integrated system of AGR–S-CO_2 and HTSE, AGR–S-CO_2–HTSE, can provide high-efficiency hydrogen production due to both the relatively high operating temperature (although lower than the He cycle reference temperature), and the higher thermal power cycle efficiency. Fig. 10 shows the net electrical efficiency of the S-CO_2 power cycle as estimated from the relevant η_{th} results of Dostal et al. [13]. In order to obtain an estimate of the net electrical power cycle efficiency, η_{el}, we consider 6–9% fractional reduction over the thermal efficiency in taking into account the losses due to friction, leakage and component cooling. The main reasons for the difference in the fractional reduction between the He and S-CO_2 cycle efficiencies are the atomic configuration and molecular size of the coolant,

coolant volumetric flow rate and the system configuration, which all favor the S-CO_2 cycle operation. The tri-atomic configuration and higher molecular weight of CO_2 incurs less leakage than that of mono-atomic He. High pressure and high fluid density in the case of S-CO_2 causes the volumetric flow rate to be much smaller than that of the He and consequently yields much less viscosity-related losses. Finally, the simplicity of the system and relatively lower operating temperature also causes less heat losses from plant components than that in the He cycle. A range of fractional reduction for the S-CO_2 cycle efficiency estimate, instead of a fixed fractional reduction as that of the He cycle, is due to the fact that the He cycle has been studied more extensively and the data concerning the He cycle design is more certain compared to that for the S-CO_2. However, the technical advantages of the S-CO_2 are still preserved in our assumptions.

As a candidate HTSE hydrogen plant, we consider the reference design called high operating temperature electrolysis (HOTELLY) by Dornier GmbH, Lurgi GmbH and Robert Bosch Gmbh [29]. By using the characteristics of HOTELLY, we evaluate the overall thermal-to-hydrogen energy conversion efficiency, η_H, as a function of nuclear plant exit temperature, which is the HTSE operating temperature, for both of the technology options, GT-MHR–HTSE and AGR–S-CO_2–HTSE. Fig. 10 shows the results relevant to the electrical and hydrogen production efficiencies, η_{el} and η_H, using these technologies.

The GT-MHR–HTSE and AGR–S-CO_2–HTSE provide large-scale centralized production of hydrogen. These technologies can take place in the future at different stages. It is possible to expect achievement of the operating temperature of 850 °C for the near-term implementation and the operating temperature of 950 °C for the long-term implementation of the GT-MHR–HTSE. Similarly, for the AGR–S-CO_2–HTSE, we propose 650 and 750 °C, respectively, which may require protective coating of the graphite in the updated design of AGR. The electrical net cycle efficiencies, η_{el}, of the GT-MHR and AGR–S-CO_2 corresponding to the respective ranges of reactor exit temperature of the working fluid, He and CO_2, are given in Fig. 10.

It is clear from Fig. 10 that attaining high power cycle efficiency can have a more important effect on the hydrogen production efficiency than solely attaining a high operating temperature. Even though the AGR–S-CO_2 is proposed to attain a lower range of operating temperature than that of the GT-MHR, thermodynamically, both η_{el} and η_H are in favor of the AGR–S-CO_2–HTSE.

In addition to its efficiency advantage over the He cycle, the S-CO_2 power cycle has a more compact and less complex structure than the He power cycle of the GT-MHR. Fig. 11 presents a schematic comparison between the physical sizes of the steam, He and S-CO_2 cycle turbines. Hence, the cost of the S-CO_2 power cycle is expected to be lower than that of the He power cycle. However, we note that the AGR has a demonstrated record of operating experience and applicable cost estimates, while the cost estimates for the GT-MHR [11] could be somewhat optimistic predictions, particularly for the first few units.

Both the GT-MHR–HTSE and AGR–S-CO_2–HTSE, as well as other possible nuclear reactors coupled to HTSE, are capital intensive technologies. Future cost reductions can be achieved by reduction in the capital cost of both the nuclear plant and in the hydrogen plant. The development of economical HTSE unit materials, which can be similar to those of the solid oxide fuel cell units, can contribute to the cost reduction. In our preliminary analysis, we have adopted the reference HTSE unit HOTELLY. However, the development of improved HTSE units with low electrode overpotential and high efficiency at lower temperatures can enable its use with lower temperature–lower cost nuclear plants. In addition, attaining high power cycle efficiency at the nuclear plant with relatively low temperatures can contribute to the cost reduction of the nuclear side, and consequently to the reduction of hydrogen production cost. Finally, economical corrosion- and irradiation-resistant materials development for both the nuclear and HTSE units would be important contributors to enhanced reliability and thus to the cost reduction.

3.3. Thermochemical hybrid cycles

A thermochemical hybrid process is a combined cycle process with both thermochemical and electrolytic reactions of water splitting. The hybrid process offers the possibility to run low-temperature reactions using electricity. One cycle developed by Westinghouse in 1975 is a promising hybrid cycle, called *sulfuric acid hybrid cycle* or the *Westinghouse Sulfur Process* (WSP) [30]. This cycle is given by the reaction equations:

$$H_2SO_4 \rightarrow SO_2 + H_2O + 1/2O_2 \qquad (800\,^\circ C),$$

$$2H_2O + SO_2 \rightarrow H_2SO_4 + H_2 - \text{electrolytic} \qquad (80\,^\circ C).$$

Fig. 12 represents a schematic overview of the WSP technology. As seen from this figure, the hydrogen generation stage involves the electrolysis but not the heat transfer at high temperature from the nuclear reactor. Therefore, this stage of the process can be located away from the nuclear plant at a distance that may be required in the future for safety concerns. At the same time, the stage of the process that require the high-temperature heat from the nuclear reactor is not involved with hydrogen generation, and hence can be kept close to the reactor. This configuration of the hydrogen plant can help reduce the heat losses that may incur if the heat transfer were to take place over long distances to the hydrogen plant. Consequently, this feature can contribute to a gain in the final net energy efficiency.

The energy efficiency of the WSP is shown in Fig. 13 as a function of process temperature. The efficiency estimate is based on the thermal and electrical energy use from

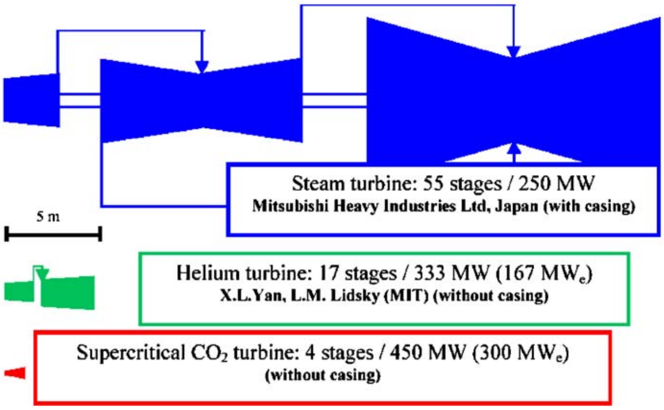

Fig. 11. Comparison of turbine sizes for steam, He, and S-CO_2 cycles [13].

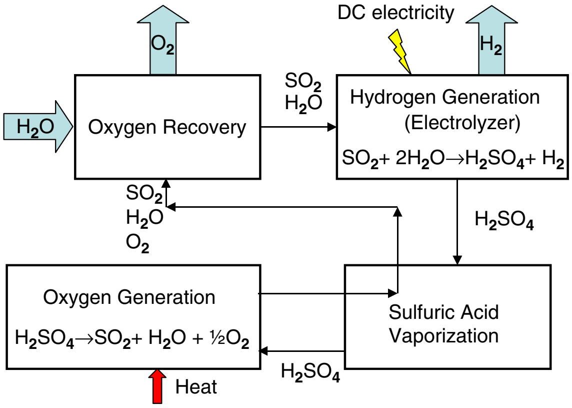

Fig. 12. Overview schematic of WSP flowsheet [31].

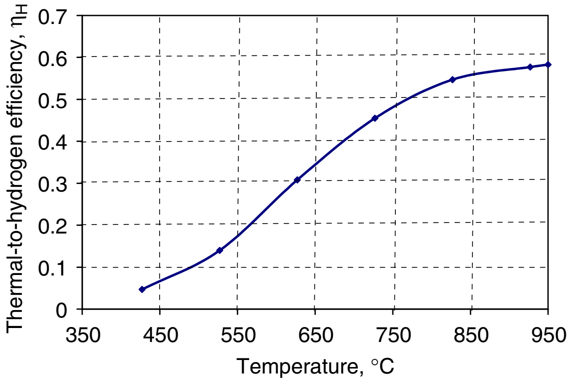

Fig. 13. Thermal-to-hydrogen energy efficiency for WSP [31] coupled to a GT-MHR.

a GT-MHR. The electrical energy use is assumed to be through a He gas turbine power conversion system of net efficiency 42%, as represented in [31].

3.4. Comparison of leading technologies

Finally, the energy efficiency of the leading candidate thermochemical cycle, SI, the HTSE process coupled to the GT-MHR and the S-CO_2–AGR nuclear technologies, and the WSP coupled to MHR technology are shown in Fig. 14. In terms of energy efficiency, the HTSE technology coupled to the direct-cycle S-CO_2 cooled AGR promises higher values at lower operating temperatures than the SI and HTSE coupled to the GT-MHR technology. The possibility of achieving high efficiency at lower temperatures is an advantage from the materials needed and reliability which ultimately translates into favorable economics.

The heat transfer requirements differ between the thermochemical water splitting processes and the HTSE process. The heat transfer to steam for the HTSE process is inherently over a small temperature range between the inlet and the exit. The reason for this is the size limitation on the performance of HTSE cells. Thermochemical process units increase in size as the capacity increases, leading to large volumes of fluids. The bulk temperature of the fluids in large volumes is more challenging to be maintained at high temperatures. The temperature decrease of steam over the HTSE cell can be taken to be negligible keeping the bulk process temperature nearly as high as the inlet temperature. Therefore, the performance of the HTSE units can be independent of the temperature difference between the inlet and exit of heating steam, as long as the steam temperature is driven up to the required temperature before it enters the HTSE cells. This feature can ease the heat transfer challenges in a nuclear-driven HTSE system.

4. Concluding remarks and recommendations for future R&D

Nuclear energy can be used as the primary thermal energy source to produce hydrogen to address the globally grow-

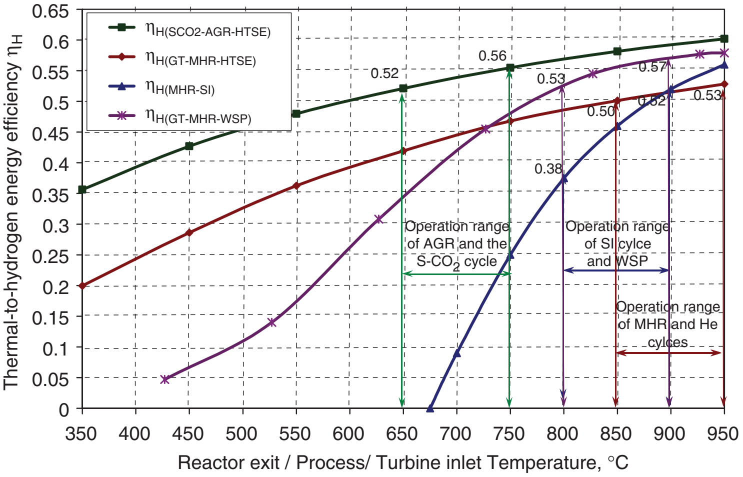

Fig. 14. Comparison of the thermal-to-hydrogen efficiency of the HTSE, SI and WSP related technologies as a function of temperature.

ing demand for hydrogen without carbon emissions. Conventional water electrolysis coupled to a currently operating nuclear plant, i.e. an LWR, is the only nuclear technology option of today, even though it represents a costly alternative in terms of energy efficiency and economics [4]. The nuclear-heated steam methane reforming, MHR–SMR, can be realized economically in the near future due to the maturity of the SMR hydrogen production technology and the partially demonstrated MHR nuclear technology. However, the MHR–SMR still involves CO_2 emissions due to the nature of SMR and can be subject to environmental taxes or penalties which add to the uncertainties in future cost estimates. High-temperature steam electrolysis and high-temperature thermochemical or hybrid water splitting processes are the primary technologies that a nuclear reactor can drive for the long term. With the right high-temperature nuclear technology integrated to the relevant hydrogen production process, both the thermochemical and electrolytic production of hydrogen have a promising future. In this work, we find the coupling of a direct cycle AGR and supercritical CO_2 power conversion system to the HTSE process potentially more energy efficient, specifically at the medium temperature range of 550–750 °C. All of the candidate technologies discussed in this paper are subject to further research and development.

The research in the nuclear field should address both the nuclear reactor-related issues and the electric power cycle-related developments. A high priority should be given to the development of high-temperature reactors that can provide coolants at temperatures higher than 800 °C. Today, this seems most readily achievable using the helium-cooled gas reactor technology of the HTGRs. The ability of the reactor fuel and structural materials to operate for a long time under irradiation and at high temperatures up to 1100 °C needs to be established through further research.

The thermal design in all nuclear hydrogen technologies is subject to demanding requirements. These requirements have different nature for thermochemical and electrochemical alternatives. The AHTR is expected to deliver heat at very high temperature over a small temperature change between the inlet and the exit. This feature of AHTR makes it specifically advantageous for providing heat for the SI cycle or the WSP at a nearly constant high temperature. This can decrease some of the inefficiencies in the sulfuric acid decomposition process by maintaining more reactant flow close to the peak temperatures where the decomposition is faster. The heat transfer requirements are less demanding for steam electrolysis, in less harsh environment, than those for the high-temperature thermochemical processes involving acidic solutions. Therefore, although the AGR is a gas-cooled reactor that operates at intermediate temperature over a large temperature difference between its inlet and exit, well-performing HTSE units can be coupled to this technology in an effective manner. Nevertheless, effective heat transfer to achieve as high a temperature as required for the process is necessary for all technology candidates.

Development of a supercritical CO_2 cycle should be given high priority. It can be directly used with an AGR that has already been commercial in the UK, or indirectly used with an HTGR or a VHTR in the long term. It can be the bottoming power cycle for a high-temperature reactor, which uses its higher temperature to permit high-performance steam electrolysis. A demonstration of the thermal conversion efficiency for a moderate size turbomachine (in the MWe range) is needed to validate the cycle thermodynamics.

Emphasis should be given to the development of the high-temperature steam electrolysis units, both for ambient and high-pressure operation. The research in this area should concentrate on achieving high performance through enhanced electrode reactions, good electronic and ionic conductivity of the electrodes and electrolyte at relatively lower temperatures (600–800 °C), stability and durability of the materials under prolonged operation, and thermal properties in operation. All these areas can further improve the overall efficiency of hydrogen production using HTSE. The already ongoing research with the same purposes for solid oxide fuel cells development can make the basis of the work for HTSE units. Another useful investigation can be evaluating the effectiveness of the WSP at industrial scale units. The past work on this process was promising but performed at small scale only.

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