PRODUCTION OF HYDROGEN BY NUCLEAR ENERGY: THE ENABLING TECHNOLOGY FOR THE HYDROGEN ECONOMY[†]

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SUMMARY

Hydrogen can replace fossil fuels in transportation, reducing vehicle emissions of CO_2, NOX and SOX and making possible fuel cell vehicles with double the mileage of conventional engines. A significant "Hydrogen Economy" is predicted that will end our dependence on petroleum and reduce pollution and greenhouse gas emissions.[1] The hydrogen can be produced from nuclear energy. Electricity from nuclear power can separate water into hydrogen and oxygen by electrolysis. The net efficiency is the product of the efficiency of the reactor in producing electricity, times the efficiency of the electrolysis cell, which, at the high pressure needed for distribution and utilization, is about 75%. For LWRs the net efficiency is about 24%. If an advanced high temperature reactor, is used, the net efficiency could be about 36%. Thermochemical water-splitting processes offer the promise of heat-to-hydrogen efficiencies of ~50%.

We carried out a detailed search for thermochemical water-splitting cycles well-suited for matching to nuclear energy.[2] We identified 115 different cycles and used detailed evaluation to select the Sulfur-Iodine cycle, the cycle with the highest reported efficiency, for development. We assessed the suitability of various nuclear reactors to the production of hydrogen using the Sulfur-Iodine cycle. A basic requirement is to deliver heat to the process at temperatures up to 900 °C. We chose the Modular Helium Reactor.

Design of an integrated chemical flowsheet for a S–I hydrogen production plant coupled to an MHR allowed us to estimate hydrogen production efficiency and capital cost. We predict an efficiency of about 50%, a capital cost of $328/kWt for the MHR, $43/kWt for the intermediate loop, and $315/kWt for the S-I process, leading to a total hydrogen production cost of $1.30/kg. With sale of the byproduct oxygen, nuclear production of hydrogen could compete in the current market. Nuclear production of hydrogen can be the "enabling technology" for the Hydrogen Economy.

I. BACKGROUND

Combustion of fossil fuels provides 86% of the world's energy.[3] Drawbacks to fossil fuel utilization include limited supply, pollution, and carbon dioxide emissions, thought to be responsible for global warming.[4] Hydrogen is an environmentally attractive fuel that has the potential to displace fossil fuels, but contemporary hydrogen production is primarily based on fossil fuels. This industry produces hydrogen for use in production for fertilizers, in oil refineries to lighten heavy crude oils and produce clearer-burning fuels, and for other industrial uses, primarily by steam reformation of methane. The fastest growing of these uses is for oil refining, shown on Fig. 1.[5] In the USA, this hydrogen industry produces 11 million tons of hydrogen a year with a thermal energy equivalent of 48 GWt. In so doing, it consumes 5% of the US natural gas usage and releases 74 million tons of CO_2.

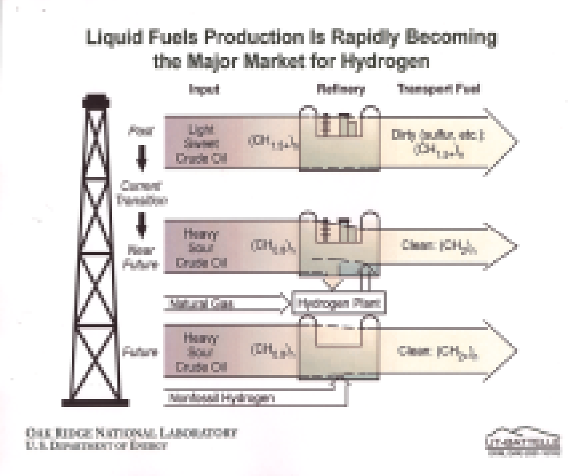

Figure 1. Use of hydrogen to lighten heavy crude oils is growing rapidly.[5]

We have recently completed a three-year project for the US DOE whose objective was to "define an economically feasible concept for production of hydrogen, by nuclear means, using an advanced high-temperature nuclear reactor as the energy source." Thermochemical water-splitting, a chemical process that accomplishes the decomposition of water into hydrogen and oxygen, could meet this objective. The goal of the first phase was to evaluate thermochemical processes which offer the potential for efficient, cost-effective, large-scale production of hydrogen and to select one for further detailed consideration. In the second phase, all the basic reactor types were reviewed for suitability to provide the high temperature heat needed by the selected thermochemical water splitting cycle for hydrogen production. In this paper we report estimates of the economic and environmental aspects of those studies.

II. THERMOCHEMICAL WATER-SPLITTING

Thermochemical water-splitting is the conversion of water into hydrogen and oxygen by a series of thermally driven chemical reactions. The direct thermolysis of water requires temperatures in excess of 2500°C for significant hydrogen generation.

$$H_2O \rightarrow H_2 + 1/2\ O_2 \quad (2500°C\ \text{min.}) \tag{1}$$

A thermochemical water-splitting cycle accomplishes the same overall result using much lower temperatures. The Sulfur-Iodine cycle is a prime example of a thermochemical cycle. It consists of three chemical reactions, which sum to the dissociation of water.

$$I_2 + SO_2 + 2H_2O \rightarrow 2HI + H_2SO_4\ (120°C) \tag{2}$$

$$H_2SO_4 \rightarrow SO_2 + H_2O + 1/2\ O_2 \quad (850°C) \tag{3}$$

$$\underline{2HI \rightarrow I_2 + H_2 \quad (450°C)} \tag{4}$$

$$H_2O \rightarrow H_2 + 1/2\ O_2 \tag{1}$$

Energy, as heat, is input to a thermochemical cycle via one or more endothermic high-temperature chemical reactions. Heat is rejected via one or more exothermic low temperature reactions. All the reactants, other than water, are regenerated and recycled. In the S-I cycle most of the input heat goes into the dissociation of sulfuric acid. Sulfuric acid and hydrogen iodide are formed in the exothermic reaction of H_2O, SO_2 and I_2, and the hydrogen is generated in the mildly exothermic decomposition of hydrogen iodide.

In phase one of the DOE-supported study described in ref. 2, General Atomics, Sandia National Laboratories and Univ. of Kentucky carried out a search of the world literature on thermochemical water-splitting cycles. We located and catalogued 822 references and identified 115 separate thermochemical water-splitting cycles. We evaluated these against quantifiable screening criteria and selected the 25 most promising for detailed technical evaluation. We studied the chemical thermodynamics of these cycles and prepared preliminary engineering block flow diagrams to evaluate practicality. We focused our attention on pure thermochemical cycles and chose the University of Tokyo 3 (UT-3) Ca-Br-Fe cycle and the Sulfur-Iodine (S-I) cycle as the two best suited for high efficiency, practical application to a nuclear heat source. Of the two candidates, the S-I cycle has the highest reported efficiency (~50%) while the UT-3 cycle appears limited to about 40% by the 760°C melting point of $CaBr_2$. Further, the S-I cycle is an all-fluid cycle, while the UT-3 cycle utilizes solid-gas reactions with potential solid material handling and attrition concerns. We chose the S-I cycle, shown schematically on Fig. 2 for our work.

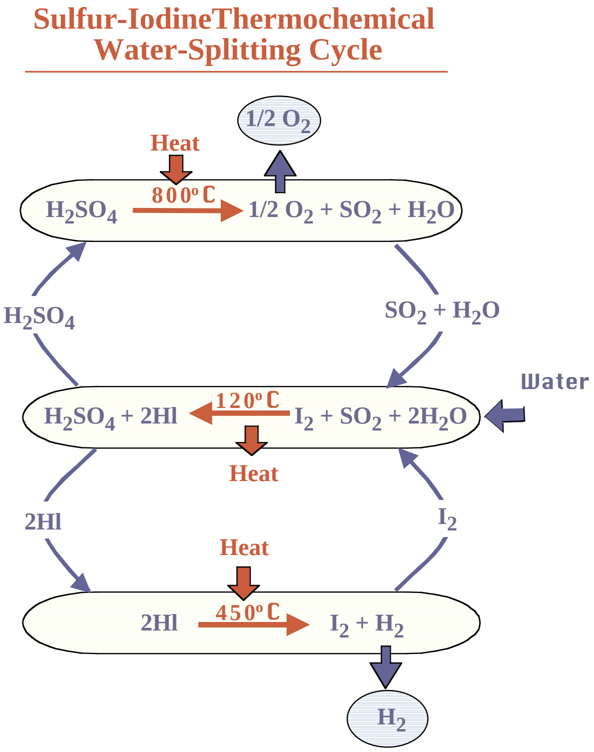

Figure 2. The S-I thermochemical water-splitting cycle is well suited for hydrogen production by nuclear energy.

The Sulfur-iodine cycle was invented at General Atomics in the mid 1970s and first described in Ref. 7. In this cycle, iodine and sulfur dioxide are added to water, forming hydrogen iodide and sulfuric acid in an exothermic reaction (2). Under proper conditions, these compounds are immiscible and can be readily separated. The sulfuric acid can be decomposed at about 850°C releasing the oxygen and recycling the sulfur-dioxide (3). The hydrogen iodide can be decomposed at about 400°C, releasing the hydrogen and recycling the iodine (4). The net reaction is the decomposition of water into hydrogen and oxygen (1). The whole process takes in only water and high temperature heat and releases only hydrogen, oxygen and low temperature heat. All reagents are recycled; there are literally no effluents. Each of the major chemical reactions of this process was demonstrated in the laboratory at GA. Work was done for application of this cycle to heat supplied by nuclear, solar and fusion energy sources. Decomposition of sulfuric acid and hydrogen iodide involve aggressive chemical environments. Materials candidates were chosen and corrosion tests performed to select preferred materials. The high temperature sulfuric acid decomposition reaction was demonstrated in the Solar Power Tower at the Georgia Institute of Technology.

The S-I cycle does require high temperatures, but offers the prospects for high efficiency conversion of heat energy to hydrogen energy as shown on Fig. 3. A schematic for the process is shown on Fig. 4.

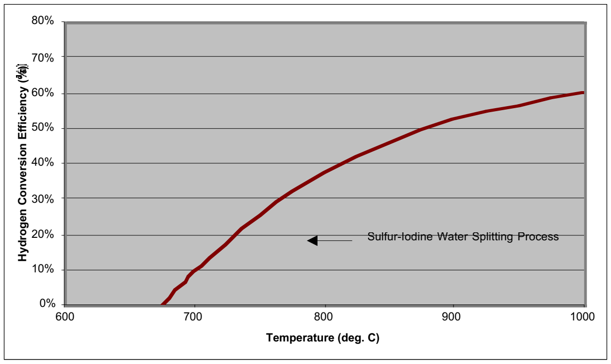

Figure 3. Estimated S-I process thermal-to-hydrogen efficiency.

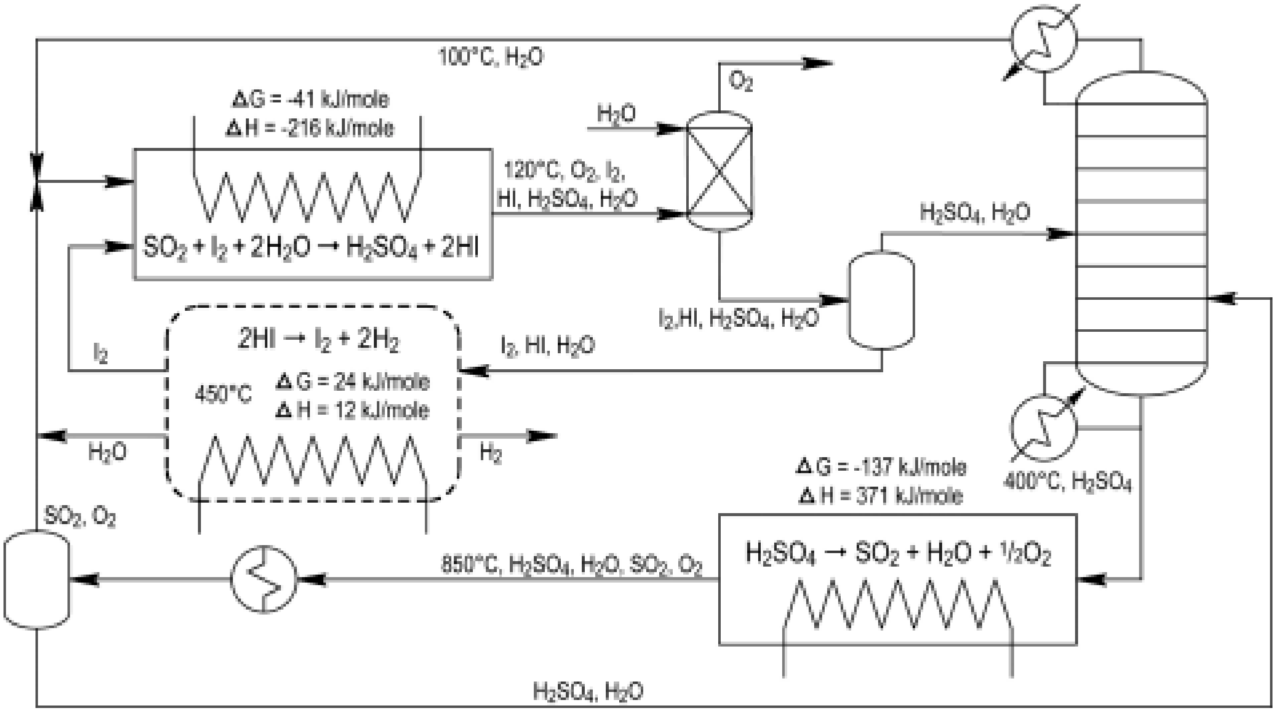

Figure 4. Sulfur-Iodine thermochemical water-splitting process schematic.

III. CHOICE OF NUCLEAR REACTOR

Sandia National Laboratories evaluated various nuclear reactors for their ability to provide the high temperature heat needed by the S-I process, and to be interfaced safely and economically to the hydrogen production process. The recommended reactor technology should require minimal technology development to meet the high temperature requirement and should not present any significant design, safety, operational, or economic issues.

We will use an intermediate helium loop between the reactor coolant loop and the hydrogen production system. This assures that any leakage from the reactor coolant loop will not contaminate the hydrogen production system or expose hydrogen plant personnel to radiation from the primary loop coolant. It also assures that the corrosive process chemicals cannot enter the core of the nuclear reactor. The heat exchanger interface sets the boundary conditions for selection of the reactor system. The principal requirement is the temperature requirement for the Sulfur-Iodine cycle, which must account for the temperature drop between the core outlet and the point of application in the hydrogen production system. We assumed a required reactor outlet temperature of 900°C.

The reactor coolant becomes a primary consideration for determining which concepts are most appropriate. The reactor/coolant types considered include pressurized water-cooled reactors, boiling water-cooled reactors, alkali liquid metal-cooled reactors, heavy liquid metal-cooled reactors, gas-cooled reactors, organic-cooled reactors, molten salt-cooled reactors, liquid-core reactors, and gas-core reactors. Four assessment stages were used in this study:

Stage 1. The level of development of the basic reactor types was reviewed. Speculative concepts with extreme developmental requirements could be eliminated at this stage.

Stage 2. Coolant properties were examined to identify merits, issues, and limitations. Fundamental limitations of coolant choices could result in the elimination. A baseline coolant option was selected for each reactor type; e.g., Li was be selected from Na, Li, NaK, and K for alkali metal-cooled reactors.

Stage 3. The reactor types were assessed against the five requirements and five important criteria given in Table I. A subjective grade is given for each reactor type (A through F) for each assessment criterion.

Stage 4. Developmental requirements were reviewed for the top three of the remaining candidates. Based on this analysis a baseline concept was recommended.

Table I. Requirements and important criteria

Basic Requirements
1. Chemical compatibility of coolant with primary loop materials and fuel.
2. Coolant molecular stability at operating temperatures in a radiation environment.
3. Pressure requirements for primary loop.
4. Nuclear requirements: parasitic neutron capture, neutron activation, fission product effects, gas buildup, etc.
5. Basic feasibility, general development requirements, and development risk

Important Criteria
1. Safety
2. Operational issues
3. Capital costs
4. Intermediate loop compatibility
5. Other merits and issues

A. Status and Characteristics of Reactor Types

Gas-core reactors were considered too speculative to be seriously considered for hydrogen production and were eliminated. Reactor coolants and heat transport fluids should have low melting points, good heat transport properties, and low potential for chemical attack on vessels and piping. Reasonable operating pressures and compositional stability at operating temperature are also important characteristics. Other desirable properties include low toxicity and low fire and explosion hazard. Reactor coolants must also possess desirable nuclear properties, such as radiation stability and low neutron activation. Low parasitic capture cross sections are required.

Pressurized water and boiling water reactors could not reasonably expect to achieve the temperatures needed for the S-I cycle. Organic coolants were similarly found to be not well-suited. For the alkali metal-cooled reactors, lithium was selected as the preferred coolant due to its low vapor pressure at high temperature. For the heavy metal-cooled reactors, the PbBi eutectic was selected due to its lower melting point and lower radiotoxicity than Pb or Bi alone. For the gas-cooled reactors, helium was selected as preferred due to its chemical inertness at high temperature.

Using the requirements and criteria presented in Table I, a subjective grade was assessed for each of the remaining candidate reactor options. A summary of the assessment grades for each requirement and criteria is provided in Table II.

From the preceding analysis, the gas-cooled reactors (GCR), molten salt-cooled reactors (MSCR), and heavy metal-cooled reactors (HMR) appear to be the most promising. An estimate of the relative development cost of the three concepts was used to select a baseline concept. The expected development cost trends for MSCR and HMR systems were compared relative to GCR development costs. The following simple indictors were used:

0	Approximately the same development cost as for gas-cooled reactors
-1, 2	Lower development cost than for gas-cooled reactors
+1, 2	Higher development cost than for gas-cooled reactors

The following needed development activities were identified and evaluated: Materials development, Fuel development, Component development, System design, and Fabrication facility development.

Development cost trends were assessed relative to GCR maximum and minimum development costs. The results of this assessment are presented in Table III, which shows that the GCR appears to result in the lowest development cost and risk.

B. Conclusions and Reactor Selection

Based on the forgoing discussion, the following conclusions and recommendations are made:

- PWR, BWR, organic-cooled, and gas-core reactors – not recommended.
- Liquid-core and alkali metal-cooled reactors – significant development risk.
- Heavy metal and molten salt-cooled reactors – promising.
- Gas cooled reactors – baseline choice.

Helium gas-cooled reactors are recommended as the baseline choice for a reactor heat source for a Sulfur-Iodine thermochemical cycle for hydrogen production.

Table II. Assessment of reactor concepts for Sulfur-Iodine thermochemical cycle

Coolant	Gas	Salt	Heavy Metal	Alkali Metal	Molten Core	PWR	BWR	Organic	Gas Core
1. Materials compatibility	A	B	B	C	B	–	F	–	–
2. Coolant stability	A	A	A	A	B	–	–	F	–
3. Operating pressure	A	A	A	A	A	F	–	–	–
4. Nuclear issues	A	A	A	B	B	–	–	–	–
5. Feasibility-development	A	B	B	C	C	–	–	–	F
1. Safety	B	B	B	B	B	–	–	–	–
2. Operations	A	B	B	B	C	–	–	–	
3. Capital costs	B	B	B	C	C				
4. Intermed. loop compatibility	A	B	B	B	B	–	–	–	–
5. Other merits and issues	B	B	B	B	B	–	–	–	–
Unweighted mean score (A=4.0)	3.67	3.30	3.33	2.87	2.80	N/A	N/A	N/A	N/A

Table III. Development cost trends relative to GCRs

	Materials	Fuel	Component	System	Fab.-Facility	Total
Molten salt	+1	+1	+1	+2	0	+6
Heavy metal	+2	+2	+1	+1	+1	+7

IV. THE H2-MHR

Selection of the helium gas-cooled reactor for coupling to the S-I hydrogen production process allows us to propose a design concept and do preliminary cost estimates for a system for nuclear production of hydrogen. The latest design for the helium gas cooled reactor is the Gas Turbine-Modular Helium Reactor.[8] This reactor consists of 600 MWt modules that are located in underground silos. The direct-cycle gas turbine power conversion system is located in an adjacent silo, as shown in Fig. 5.

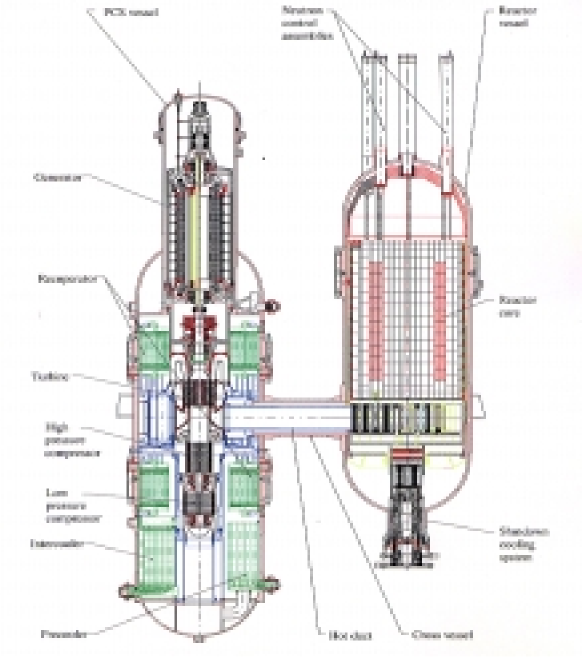

Figure 5. The GT-MHR.

This new generation of reactor has the potential to avoid the difficulties of earlier generation reactors that now have stalled nuclear power in the United States. The GT-MHR has high temperature ceramic fuel and a core design that provide passive safety. A catastrophic accident is not possible. Under all conceivable accident conditions the reactor fuel stays well below failure conditions with no actions required by the plant operators or equipment. By avoiding the need for massive active safety back-up systems, the capital cost of the GT-MHR is reduced. The high temperature fuel also allows high efficiency power conversion. The gas turbine cycle is projected to give 48% efficiency.

The high helium outlet temperature also makes possible the use of the MHR for production of hydrogen using the S-I cycle. By replacing the gas turbine with a primary helium circulator, an intermediate heat exchange, an intermediate helium loop circulator and the intermediate loop piping to connect to the hydrogen production plant, the GT-MHR can be changed into the H2-MHR, as shown in Fig. 6.

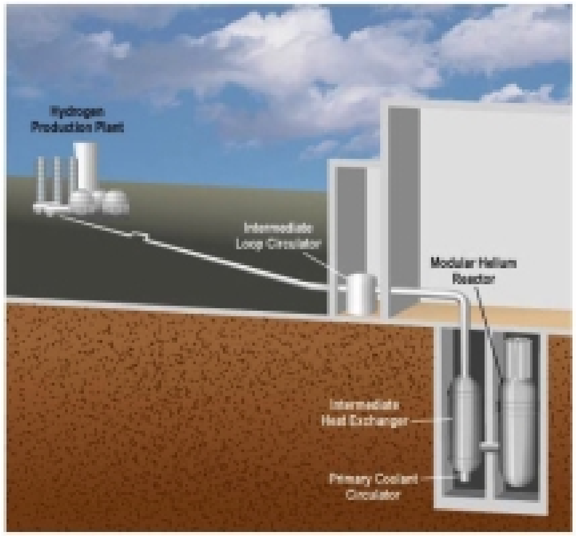

Figure 6. The H2-MHR.

We have made preliminary projections about the economics of hydrogen production from nuclear energy. The Gas Turbine - Modular Helium Reactor has a predicted capital cost of \$975/kWe or \$468/kWt. The predicted capital cost of the reactor portion of the GT-MHR (excluding the cost of the turbo-generator and including an intermediate heat exchanger, circulators nd piping) is \$371/kWt. We estimate that cost of the S-I cycle hydrogen plant will be around \$315/kWt, for a total of \$686/kWt and with an estimated heat-to-hydrogen efficiency of 50%, would give a total capital cost of \$686 / 0.50 = \$1,372/kWh ("\$ per kilowatt hydrogen"). The details of these costs are shown on Table IV.

The operating cost of the GT-MHR is estimated to be 3.0 \$/MW$_e$h for O&M cost plus \$7.4/MW$_e$h for fuel cycle costs, for a total of \$10.3/MW$_e$h or \$4.9/MW$_t$h for all operating costs (fuel, O&M, waste disposal, decommissioning)[8]. We assume these scale with capital cost for the process heat MHR to \$3.9/MW$_t$h. The S-I cycle O&M cost is predicted to be ~7% of initial capital cost/year or \$2.8/MW$_t$h. The total H2-MHR plant operating cost is thus \$6.7/MW$_t$h. These costs assume 90% capacity factor.

Table IV. Modular Helium Reactor Capital Costs
Estimated "Nth of a kind" costs for 4x600MWt plant

Acct	Direct Costs	GT-MHR[8] Electric Plant (4x286 MWe) Yr 2002 M$	PH-MHR Process Heat Plant (4x600 MWt) Yr 2002 M$	Intermediate Loops (2400 MWt) Yr 2002 M$	S-I H2 Plant Hydrogen Plant (2400 MWt) Yr 2002 M$
20	Land And Land Rights	0	0		
21	Structures And Improvements	132	132		
22	Reactor Plant Equipment	443	343		
23	Turbine Plant Equipment	91	0		
24	Electric Plant Equipment	62	50		
25	Miscellaneous Plant Equipment	28	28		
26	Heat Rejection Or S-I System	33	0		534
	Interm. Loop Circ. & Piping			73	
2	Total Direct Cost	789	553	73	534
9	Total Indirect Costs	274	192	25	191
	Base Construction Cost	1063	745	98	720
	Contingency	53	37	5	36
	Total Cost	1116	783	103	756
	$/kWe / $/kWt	975 / 468	"684" / 328	- / 43	- / 315

Both the MHR and the S-I process are capital intensive. Thus the cost of hydrogen production depends on interest rate used in the economic calculations, as shown on Fig. 7, assuming a 40 year lifetime with zero recovery value.

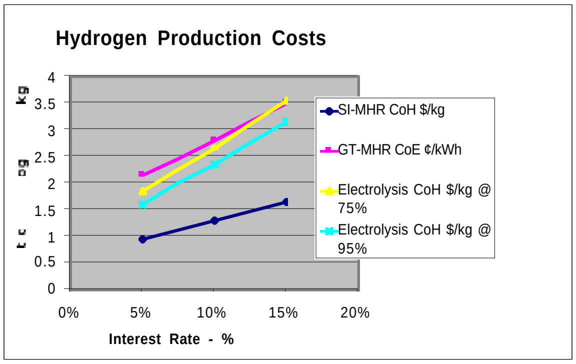

Figure 7. Estimated cost of hydrogen.

Figure 6 shows that for a nominal interest rate of 10%, the H2-MHR could produce hydrogen for about $1.30/kg. Shown for comparison are the cost of electricity from the GT-MHR in ¢/kW$_e$h and the cost of producing hydrogen by electrolysis using that electricity. The benefit of the higher efficiency and lower total capital cost of thermochemical water-splitting is evident.

The cost of producing hydrogen from natural gas by steam reformation of methane depends strongly on the cost of the natural gas, which is used for both the feedstock and the energy source. At the current natural gas cost of $3.50/MBtu, steam reformation can produce hydrogen for about $1.00/kg. However, if carbon capture and sequestration is required, the estimated cost of $100/ton of CO_2 would add about 20¢/kg of H_2 to the cost of hydrogen from methane. If the H2-MHR were able to also sell the oxygen produced at the current price of about 5.3¢/kg, it would reduce the cost of nuclear hydrogen production by about 40¢/kg of H_2. This would mean that nuclear production of hydrogen using the Modular Helium reactor coupled to the sulfur-iodine thermochemical water-splitting cycle would be competitive with hydrogen produced from fossil fuels even at today's low prices for natural gas. As the price of natural gas rises with increasing demand and decreasing reserves, nuclear production of hydrogen would become still more cost effective. This could result in a large demand for nuclear power plants to produce the hydrogen.

V. CONCLUSIONS

Production of hydrogen is a very attractive application of nuclear energy. A large hydrogen market already exists and it is growing rapidly to provide increasing amounts of hydrogen to oil refineries for upgrading heavy crude oils and producing clean-burning products. If all of this hydrogen were to be provided by nuclear plants operating at 50% heat-to-hydrogen efficiency, it would take 100 GWt of nuclear power to do so. And this market is expected to continue growing at ~10%/yr, doubling by 2010 and doubling again by 2020. To transition to a "Hydrogen Economy" would take still more hydrogen. Serving all the US transportation energy needs with hydrogen would multiply current hydrogen demand by a factor of 18. Serving all our non-electric energy needs would require a factor of 40 over current hydrogen production.

The recent DOE-supported study of nuclear production of hydrogen identified the Sulfur-Iodine thermochemical water-splitting cycle coupled to the Modular Helium Reactor (the H2-MHR) as an attractive candidate system for hydrogen production.

Estimated costs presented in this paper show that hydrogen production by the H2-MHR could be competitive with current techniques of hydrogen production from fossil fuels if CO_2 capture and sequestration is required and if the by-product oxygen can be sold. This favorable situation is expected to further improve as the cost of natural gas rises.

Nuclear production of hydrogen would allow large scale production of hydrogen at economic prices while avoiding the release of CO2. Nuclear production of hydrogen could thus become the enabling technology for the Hydrogen Economy.

ACKNOWLEDGMENTS

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