GA–A24428

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This is a preprint of a paper to be presented at the 2003 World Nuclear Association Symposium, September 3-5, 2003, in London, United Kingdom and to be published on *CD-ROM.*

**Work supported in part by
the U.S. Department of Energy
under Grant No. DE-FG03-99SF21888**

**GENERAL ATOMICS PROJECT 04962
SEPTEMBER 2003**

Use of the Modular Helium Reactor for Hydrogen Production

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Introduction

A significant "Hydrogen Economy" is predicted that will reduce our dependence on petroleum imports and reduce pollution and greenhouse gas emissions [1]. 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 of 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. In the USA, this hydrogen industry produces 11 million tons of hydrogen a year with a thermal energy equivalent of 48 GW(t). In so doing, it consumes 5% of the U.S. natural gas usage and releases 74 million tons of CO_2. Transition to a Hydrogen Economy will require significant expansion in the production and use of hydrogen. Use of hydrogen for all our transportation energy needs would require a factor of 18 more hydrogen than currently used. Use of hydrogen for all our non-electric energy needs would require a factor of 40 increase. Clearly, new sources of hydrogen will be needed. Hydrogen produced from water using nuclear energy can be one of the sources, and would avoid use of fossil fuels and greenhouse gas emissions.

Hydrogen could be produced from nuclear energy by several means [2]. 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%-80%. For LWRs with 32% electrical efficiency the net efficiency is about 24%-26%. If an advanced high temperature reactor with 48% electrical efficiency is used, the net efficiency could be about 36%-38%. Thermochemical water-splitting processes offer the promise of heat-to-hydrogen efficiencies of ~50%.

We have recently completed a three-year project for the U.S. Department of Energy (DOE) whose objective was to "define an economically feasible concept for production of hydrogen, using an advanced high-temperature nuclear reactor as the energy source"[3]. Thermochemical water-splitting, a chemical process that accomplishes the decomposition of water into hydrogen and oxygen, met this objective. The goal of the first phase of this study 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. We selected the Sulfur-Iodine cycle. In the second phase, we reviewed all the basic reactor types for suitability to provide the high temperature heat needed by the selected thermochemical water splitting cycle and chose the helium gas-cooled reactor. In the third phase we designed the chemical flowsheet for the thermochemical process and estimated the efficiency and cost of the process and the projected cost of producing hydrogen. These results are summarized in this paper.

Thermochemical Water-Splitting

Thermochemical water-splitting is the conversion of water into hydrogen and oxygen by a series of thermally driven chemical reactions. 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)	(Exothermic)	(1)
$H_2SO_4 \rightarrow SO_2 + H_2O + 1/2\ O_2$	(~850°C)	(Endothermic)	(2)
$2HI \rightarrow I_2 + H_2$	(~350°C)	(Endothermic)	(3)
$H_2O \rightarrow H_2 + 1/2\ O_2$			(4)

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, (2). Sulfuric acid and hydrogen iodide are formed in the exothermic reaction of H_2O, SO_2 and I_2 (1) and the hydrogen is generated in the decomposition of hydrogen iodide (3).

In phase one of the DOE-supported study described in Ref. [3], 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 Sulfur-Iodine (S-I) cycle, shown on Fig. 1, as the one best suited for high efficiency, practical application to a nuclear heat source.

The Sulfur-Iodine cycle was invented at General Atomics in the mid 1970s and first described in Ref. [4]. In this cycle, iodine and sulfur dioxide are added to water, forming hydrogen iodide and sulfuric acid in an exothermic reaction (1). 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 (2). The hydrogen iodide can be decomposed at about 350°C, releasing the hydrogen and recycling the iodine (3). The net reaction is the decomposition of water into hydrogen and oxygen (4). 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 involves 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 in the laboratory and in the Solar Power Tower at the Georgia Institute of Technology. A complete laboratory scale S-I cycle test loop was recently operated successfully in Japan [6]. A schematic for the process is shown on Fig. 2.

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.

Selection 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 [5]. 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 reactor outlet temperature of 950°C. This should give a peak process temperature of ~900°C and a process efficiency of 52%.

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. The reactor types were assessed against the five requirements and five important criteria given in Table 1.

Table 1. Reactor Selection Requirements and 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

Based on this assessment, and upon evaluation of the relative development requirements for candidate reactors, the following conclusions and recommendations were made:

- PWR, BWR, and organic-cooled reactors – not recommended: cannot achieve the high temperatures needed.
- Liquid-core and alkali metal-cooled reactors – significant development risk due to materials concerns at the high temperatures needed.
- Heavy metal and molten salt-cooled reactors – promising, but significant development needed.
- Gas cooled reactors – baseline choice, only modest development needed for helium gas cooled reactor.
- Gas-core reactors – not recommended, too speculative

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

The Modular Helium Reactor

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 [7]. This reactor consists of 600 MW(t) 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. 4.

This new generation of reactor has the potential to avoid the difficulties of earlier generation reactors that now have stalled nuclear power in the U.S. 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 system with a primary helium circulator, an intermediate heat exchanger, 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. 5.

Economics Estimates

We have estimated the economics of hydrogen production from nuclear energy using the S-I thermochemical cycle. The Gas Turbine - Modular Helium Reactor has a predicted "overnight" capital cost of $975/kW(e) or $468/kW(e) [7]. We excluded the cost of the turbo-generator and included an intermediate heat exchanger, circulators and piping. We assumed a $45/kW$_t$ premium to provide 950°C for the S-I process instead of the 850°C

outlet temperature of the GT-MHR. We determined the capital cost of the hydrogen plant equipment and chemical inventory. We estimated the total capital and operating costs of the integrated hydrogen plant to calculate the cost of the hydrogen produced. The capital costs are for a 2,4000 MW(t) "nth of a kind" plant producing approximately 800 tons of hydrogen per day, enough to support a large oil refinery, or almost 1.5 million fuel cell cars. The costs include all direct and indirect costs, plus interest during construction. The Reactor operating costs include all fuel cycle costs (fuel, conversion, enrichment, fabrication, waste disposal and decommissioning) plus normal operation and maintenance costs. The Hydrogen Plant operating costs include normal operation and maintenance costs plus the cost of high purity water. All costs are in 2002 funds. Since both the reactor and the hydrogen plant are capital intensive, the hydrogen cost estimates using several different capital recovery factors (CRF) are shown.

Table 2. Economics of a 2,400 MW(e) H2-MHR at 850°C and 950°C.

	850°C, 42% Efficiency	950°C, 52% Efficiency
Reactor capital cost, M$	968.2	1,098.0
Hydrogen plant capital cost, M$	643.2	796.3
Reactor fuel + operating cost, M$/yr	93.9	97.1
Hydrogen plant operating cost, M$/yr	50.7	62.7
Hydrogen production rate, kg/yr	213×10^6	264×10^6
Cost of hydrogen, $/kg		
— Public utility – 10.5% CRF	1.53	1.42
— Regulated utility – 12.5% CRF	1.69	1.57
— Unregulated utility – 16.5% CRF	2.01	1.87

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 about $6/MBtu, steam reformation can produce hydrogen for about $1.40/kg. If carbon capture and sequestration is required, an estimated cost of $30/ton of CO_2 could add 20¢/kg of H_2 to the cost of hydrogen from methane. Nuclear production of hydrogen using the Modular Helium Reactor could thus be competitive at today's prices for natural gas. As the price of natural gas rises with increasing demand and decreasing reserves, nuclear production of hydrogen would become more and more cost effective.

Conclusions

Nuclear energy is an attractive potential source of hydrogen for the Hydrogen Economy. 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. 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. New sources of hydrogen will be needed. Our 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.

The 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. The economic attractiveness of hydrogen production from high temperature nuclear energy will 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 CO_2. Nuclear production of hydrogen could thus become the enabling technology for the Hydrogen Economy.

Acknowledgments
Work supported by General Atomics and U.S. Department of Energy under Grant No. DE-FG03-99SF21888.

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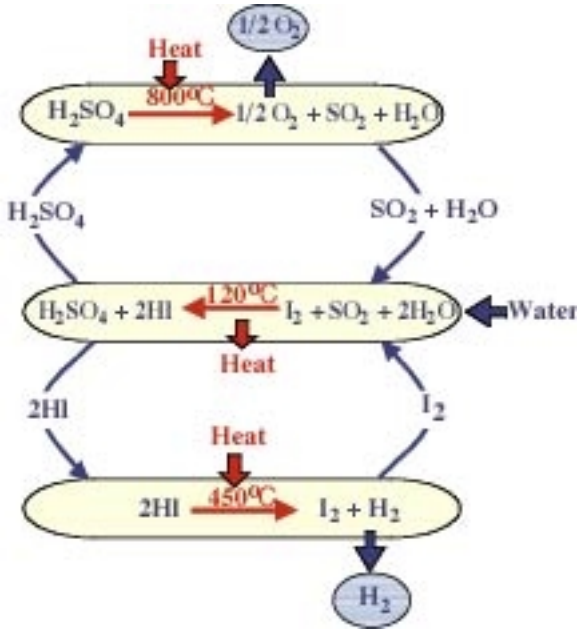

Figure 1. The S-I thermochemical water-splitting cycle is well suited for hydrogen production by nuclear energy.

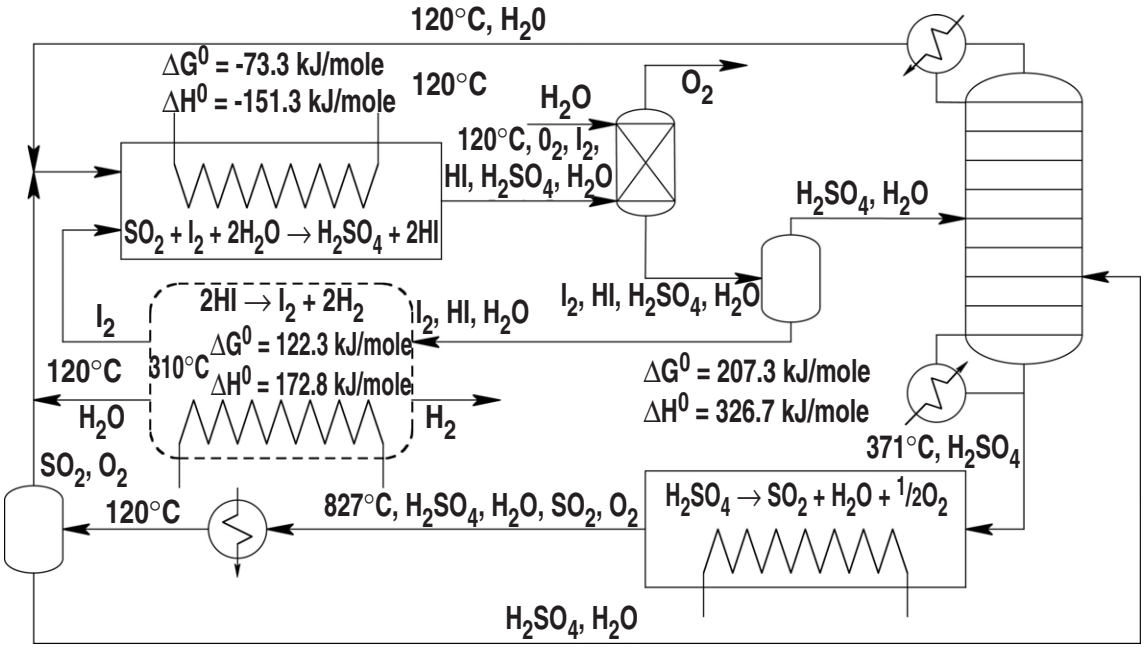

Figure 2. Sulfur-iodine thermochemical water-splitting process schematic.

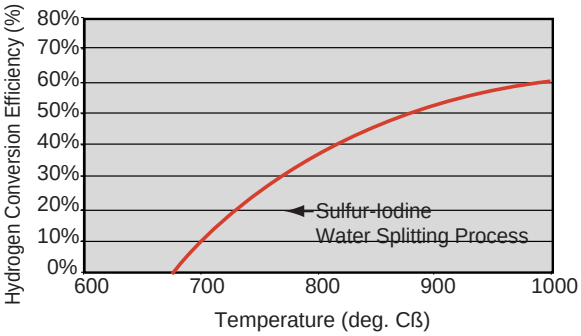

Figure 3. Estimated S-I process thermal-to-hydrogen efficiency vs. peak process temperature. Hydrogen production efficiency can exceed 50% if temperatures of ~900°C are available.

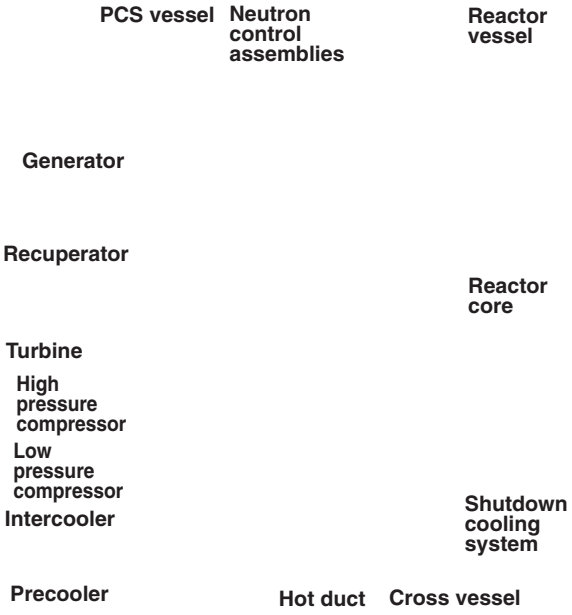

Figure 4. The high temperature capability of the GT-MHR allows efficient production of hydrogen from nuclear energy.

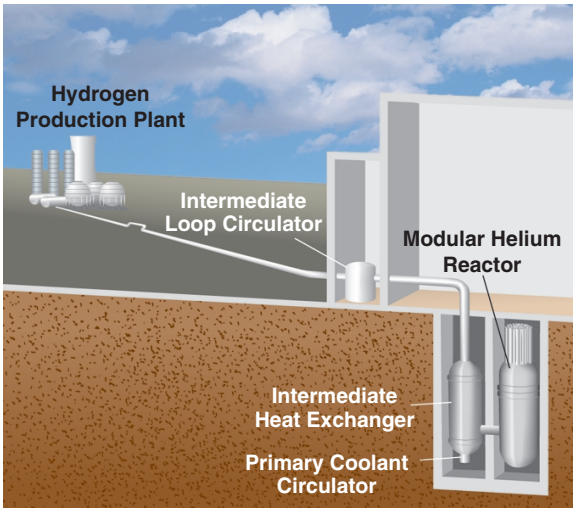

Figure 5. The H2-MHR. Below grade installation enhances inherent safety of MHR for hydrogen producton.