L.C. Brown et al.

High Efficiency Generation of Hydrogen Fuels Using Nuclear Power
Final Technical Report for the period August 1, 1999
through September 30, 2002

EXECUTIVE SUMMARY

Hydrogen is a promising energy carrier, which potentially could replace the fossil fuels used in the transportation sector of our economy. Fossil fuels are polluting and carbon dioxide emissions from their combustion are thought to be responsible for global warming. However, no large scale, cost-effective, environmentally attractive hydrogen production process is currently available for commercialization.

This report describes work accomplished by the team of General Atomics (GA), Sandia National Laboratories (SNL) and the University of Kentucky (UK) during a three-year project 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." The purpose of this work was to determine the potential for efficient, cost-effective, large-scale production of hydrogen utilizing high temperature heat from an advanced nuclear power station. The benefits of this technology include generation of a low-polluting transportable energy feedstock in a highly efficient method that has no greenhouse gas emissions, from an energy source whose availability and sources are domestically controlled. The work was divided into three phases. The work of Phases 1 [E–1] and 2 [E–2] have been reported earlier but are also summarized here for completeness.

The purpose of the first phase was to evaluate thermochemical processes which offer the potential for efficient, cost-effective, large-scale production of hydrogen from water, in which the primary energy input is high temperature heat from an advanced nuclear reactor and to select one for further detailed consideration. This was done in several steps: (1) a detailed literature search was performed to develop a database of all published thermochemical cycles, (2) a set of objective screening criteria was developed to rate each cycle and was used to reduce the initial list to 25 cycles [E–3], and (3) a detailed analysis was used to reduce the number of cycles under consideration to two and finally to one. The Phase 1 report is included as the Attachment. Appendix A presents an introduction to thermochemical water splitting.

Ten databases were searched (e.g., Chemical Abstracts, NTIS, etc.), and over 800 literature references, pertaining to thermochemical production of hydrogen from water, were organized in a computerized database. In the process, over 100 thermochemical water-splitting cycles were identified and organized into a separate, computer searchable database.

The first round of screening, using defined screening criteria and quantifiable metrics, yielded 25 cycles for more detailed study. The second round of screening, using refined criteria, reduced the 25 candidate cycles to 2 final options.

High Efficiency Generation of Hydrogen Fuels Using Nuclear Power   L.C. Brown et al.
Final Technical Report for the period August 1, 1999
through September 30, 2002

The two cycles selected for final consideration are the UT-3 cycle and the Sulfur-Iodine (S-I) cycle. The UT-3 cycle was invented at the University of Tokyo and much of the early development was done there. This cycle has been studied extensively by the Japan Atomic Energy Research Institute (JAERI). After considering several different flowsheets making use of the UT-3 cycle, JAERI selected the so-called Adiabatic UT-3 process for further development. The predicted efficiency of the Adiabatic UT-3 process varies between 35% and 40% depending upon the efficiency of membrane separators which are under development. A 10% overall efficiency increase is projected if electricity is co-generated along with the hydrogen.

The S-I cycle is the cycle with the highest reported efficiency based on an integrated flowsheet. The last full flowsheet of the process, developed in 1981–1984, had a predicted efficiency of 38% when coupled to a fusion reactor. Since that time, various researchers have pointed out improvements that should increase the already high efficiency of this cycle and, in addition, lower the capital cost. As the S-I cycle had both the highest predicted efficiency and the most potential for further improvement, it was selected as the basis for the ongoing effort. A schematic for the nuclear-matched S-I cycle is shown in Fig. Ex–1.
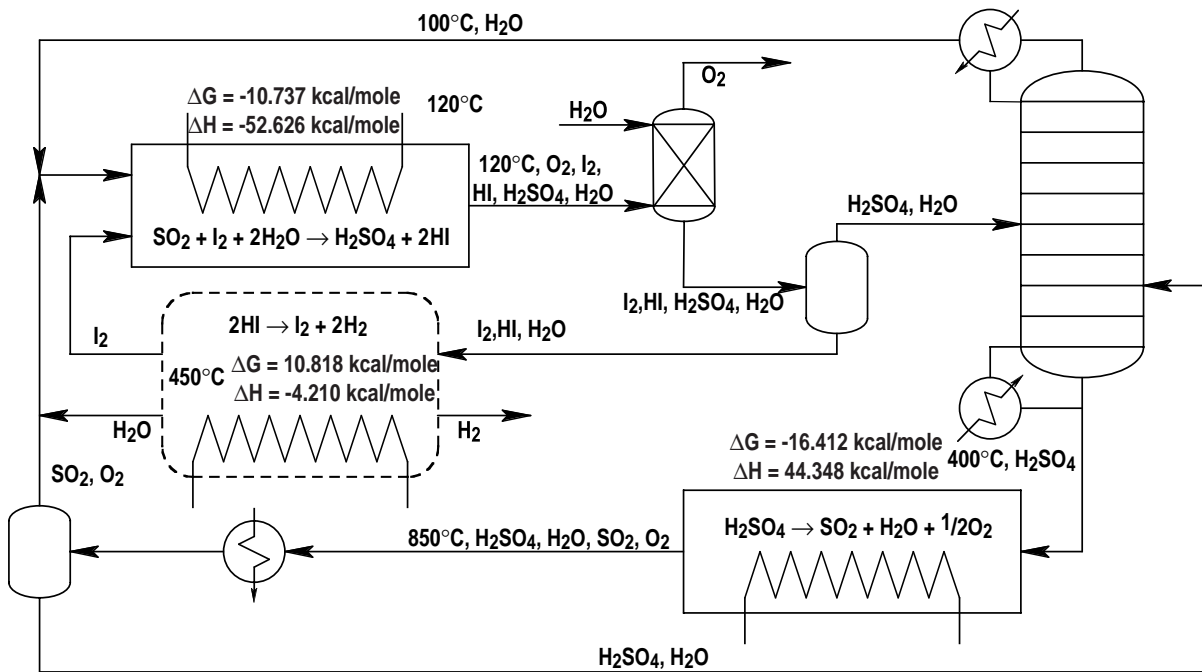

Fig. Ex–1. S-I cycle process flow diagram.

The goal of Phase 2 and 3 efforts was to determine the optimal configuration of a high temperature nuclear reactor coupled to a thermochemical hydrogen plant so as to deliver hydrogen at the minimum total cost. The reactor selection and integration task was headed by

L.C. Brown et al.   High Efficiency Generation of Hydrogen Fuels Using Nuclear Power
Final Technical Report for the period August 1, 1999
through September 30, 2002

SNL while the hydrogen production flowsheet and costing effort was split between GA and UK.

The main elements comprising Phases 2 and 3 are:

- Evaluate and select the nuclear reactor best suited for production of hydrogen by the S-I thermochemical cycle process.

    — Develop the concept for the interface which matches the reactor to the process.

    — Develop the flowsheets for the chemical process.

    — Conceptually design each major piece of process equipment to the degree necessary for estimating the capital equipment cost and then determine that cost.

    — Using standard chemical engineering cost estimating techniques, estimate the total chemical plant capital cost based on the equipment costs.

    — Combine the chemical plant capital costs with estimates of the reactor capital cost and operating costs to predict the cost of hydrogen from the project.

During Phase 2, a detailed evaluation of alternative nuclear reactor concepts was performed to select the reactor best suited to thermochemical hydrogen production using the selected S-I process. SNL evaluated nine categories of nuclear reactor, identified by their coolants. These ranged from reactors that have been successfully commercialized [the Light Water Reactors (LWRs)], to those that have been demonstrated but not successfully commercialized (helium and liquid metal-cooled reactors), to reactors that have been conceptualized but never built. SNL evaluated these reactors against a set of five design requirements and five performance criteria. Today's LWRs simply cannot achieve adequate temperature to be used for thermochemical water-splitting. Several nonwater-cooled reactors do appear to have high enough temperature potential to drive the S-I cycle. Of these, the helium-cooled reactor, the heavy metal (Pb-Bi) cooled reactor and the molten salt-cooled reactor rated best. The SNL study, Appendix B, indicated that although heavy metal and molten salt-cooled reactor concepts could potentially be developed to the point that they could support hydrogen production, helium gas-cooled reactors had reached the point in development where nuclear hydrogen production would be possible with essentially no further development. We selected the helium gas-cooled reactor for our study.

The starting point for process development and improvement is the S-I cycle flowsheet developed in 1979–1984 [E–4]. This flowsheet was produced at the time when computer simulation of chemical processes was coming into vogue using general-purpose chemical flowsheet simulators. Several attempts were made at that time to use various simulation programs then extant but the thermodynamic models available at that time could not deal with the nonidealities of even the simplest part of the process and hand calculations were used for the entire flowsheet.

High Efficiency Generation of Hydrogen Fuels Using Nuclear Power   L.C. Brown et al.
Final Technical Report for the period August 1, 1999
through September 30, 2002

When beginning this effort, we assumed that current computer simulation programs could adequately represent the complex behaviors characteristic of the S-I cycle. This is, to a large extent, true but transforming the available data into computer usable form proved to be a considerable undertaking. Aspen Plus, the process simulation program selected for this effort, has, perhaps, the best implementation of electrolytic solution thermodynamics available. But, even for the simple system H_2SO_4/H_2O, there was no valid model covering the range of temperature, pressure and composition needed to describe the thermochemistry of the S-I cycle. The services of Aspen Technology, licenser of Aspen Plus, were utilized to regress sulfuric acid thermodynamics data to generate the model used for our analysis. The resulting electrolytic nonrandom two-liquid sulfuric acid model is a good representation of the H_2SO_4/H_2O system and was used to model Section 2 of the flowsheet.

We were unable to develop Aspen Plus models for $HI/I_2/H_2O$ system (Section 3) or the $H_2SO_4/HI/I_2/H_2O$ system (Section 1). The current state of the equilibrium data for the $HI/I_2/H_2O$ system appears to be inadequate to be able to regress a model that will successfully converge. The flowsheets for Sections 1 and 3 were based on earlier analyses, calculated by hand without a chemical simulation computer program. For Section 3, HI decomposition, we use the reactive distillation process calculated by Roth and Knocke [E–5] at the University of Aachen. For Section 1, we started with the 1982 flow sheet and calculated the compositions after accounting for the large recycle flows from Section 3. While these analyses are adequate to calculate self-consistent flowsheets, they did not allow us to optimize the overall S-I system to the extent desired. We believe that recuperation of heat from Section 2 into Section 3 would allow still further increases in the efficiencies we calculated for the current flowsheet. This can be done when the $HI/I_2/H_2O$ system equilibrium data are measured, a full chemical system model is regressed from these data, and a complete Aspen Plus model for the entire S-I flowsheet is developed. Measurement of the equilibrium data is a high priority R&D need identified by this study.

In Phase 2, we investigated several alternatives to the 1982 flowsheet, which was based on the use of phosphoric acid, H_3PO_4, to pull the water out of the $HI/I_2/H_2O$ mixture in Section 3 prior to HI decomposition. While this process is effective, the energy needed and capital cost of the equipment used to then extract the water to regenerate the H_3PO_4 are high and they reduce the efficiency and increase the cost of the 1982 S-I system. At that time, GA suggested reactive distillation as an alternative but did not pursue it. Roth and Knocke at the University of Aachen analyzed this option and predicted that a higher efficiency and up to 40% lower capital cost might be obtained. We chose the reactive distillation option for Section 3 of our flowsheet design. This process only decomposes one-sixth of the HI and recycles the remaining five-sixths to Section 1. This results in increased costs and we had hoped to further optimize the process to increase the fractional decomposition. The computational difficulties described above prevented us from completing this step and we adopted the original Roth and Knocke reactive distillation process for our design.

L.C. Brown et al.   High Efficiency Generation of Hydrogen Fuels Using Nuclear Power
Final Technical Report for the period August 1, 1999
through September 30, 2002

In Phase 3 of the study, we completed a flowsheet for each Section of the S-I process. These are presented in detail in this report. These flowsheets were developed with an assumed peak process temperature of 827°C. This matched the peak temperature of the 1978 flowsheet and we expect that this temperature could be attained using the current 850°C Modular Helium Reactor outlet temperature and a high effectiveness compact heat exchanger. The complete design at this temperature only achieved 42% thermal efficiency. We estimated the increase in efficiency that could be achieved at higher peak process temperature and believe that at 900°C peak process temperature, we will be able to achieve 52% efficiency. This would require about 950°C reactor outlet temperature. We have used this system for our economics estimates.

We performed preliminary equipment sizing calculations to determine the capital cost of the equipment and then estimated the total capital and operating costs of the integrated hydrogen plant. Finally, we incorporated the reactor capital cost and operating costs for an estimated 950°C Modular Helium Reactor to estimate the cost of the hydrogen produced by a high temperature nuclear reactor coupled to the S-I process. Selected results of this economics analysis are shown below. The capital costs are for an "nth of a kind" plant and include all direct and indirect costs, plus interest during construction. The Reactor operating costs include all fuel cycle costs (fuel, conversion, enrichment, fabrication, and waste disposal) 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 funding. Since both the reactor and the hydrogen plant are capital intensive, the hydrogen cost using several different capital recovery factors (CRF) are shown in Table Ex–1.

Table Ex–1
Cost of 2400 MWt 4-module Modular Helium Reactor Hydrogen Production Plant

	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

On the basis of these results, we recommend that work be carried out to demonstrate and develop the S-I cycle for production of hydrogen from nuclear energy.