

NGNP Hydrogen Technology Down-Selection

Results of the Independent Review Team (IRT) Evaluation

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EXECUTIVE SUMMARY

An Independent Review Team (IRT) has been chartered by US Department of Energy Office of Nuclear Energy (DOE-NE) to investigate reducing or "down-selecting" the number of candidate hydrogen production technologies that are being considered for integration into the Next Generation Nuclear Plant (NGNP). Down-selecting from the three leading technologies is considered necessary to improve the potential that all NGNP technological, licensing, and programmatic goals can be met on schedule and within budget. One of the key NGNP goals is the demonstration of an advanced hydrogen production technology using heat, steam, or electric power generated by a high temperature gas cooled reactor (HTGR) plant. The NGNP, a prototype HTGR, is scheduled to be operational by 2021.

The IRT recommendations are expected to represent one input to the DOE-NE decision regarding the ultimate approach that will be taken for further development of hydrogen production at NGNP. The final decision will be made by DOE-NE in consultation with Idaho National Laboratory (INL), the lead DOE lab for nuclear energy, and could include an approach that: (1) focuses all resources on what is considered the single most viable hydrogen generation process in light of NGNP goals and operating conditions, or (2) focuses primarily on one technology with continuation of more-limited research and development (R&D) on one or both of the remaining two technologies as part of an overall risk-mitigation strategy.

While several hundred thermochemical, electrochemical, and hybrid methods for the generation of hydrogen using nuclear energy have been proposed over the last thirty years, the three that have garnered the most attention both in the US and internationally are the: (1) Sulfur Iodine (SI) process, (2) Hybrid Sulfur Electrolysis (HyS) process, and (3) High Temperature Steam Electrolysis (HTSE) process. While the IRT discussed other possible hydrogen generation processes, the down-selection evaluation focused on these three leading candidates because the other technologies were judged too immature for deployment with NGNP.

In this regard, the approach used by the IRT for the down-selection was as follows:

• The INL first identified "Technology Leads" for each of the three processes. These Technology Leads were individuals from corporations or laboratories that are considered at the forefront of research and development of the individual technologies. Each Technology Lead prepared a Technology Summary for their assigned hydrogen production

process. The Technology Summaries, together with a comprehensive set of other background references, were provided to the IRT for review.

- The IRT adopted a formal methodology for evaluating the SI, HyS, and HTSE hydrogen production technologies. This included establishing specific technical and economic goals and requirements for the hydrogen production process and the hydrogen product in terms of quantity, form, and purity.
- The IRT held a 5-day down-selection workshop in June 2009. One full day was devoted to each of the three technologies. Presentations were made by the Technology Leads and other researchers during these first three days. This was followed by two days of evaluations by the IRT. More than 80 specific technical issues were identified and considered in detail by the IRT over the course of the evaluations.
- The IRT thoroughly discussed each of the technologies, and then scored or graded each process using an evaluation methodology agreed to by the IRT with the concurrence of INL and DOE-NE. While some of the IRT conclusions were based on expert engineering judgment, scoping calculations of quantities such as hydrogen production rates, production costs, and process efficiencies were performed wherever possible.

The IRT finalized a recommendation for the down-selection based in large part on the results of the grading. The IRT also developed companion recommendations for potential short-term R&D tasks for each of the technologies. The IRT considers that completion of these R&D tasks would serve to improve the overall potential that hydrogen production processes can be developed for integration initially with NGNP, and then with HTGRs in the longer term. The continued pursuit of such short term R&D, particularly in areas not directly related to the leading hydrogen candidate(s), also reduces the overall NGNP program risk in the event that some technical barrier for the leading candidate cannot be overcome. The IRT defined short term R&D to be those limited scope activities that could be completed in 1 to 3 years. Short term R&D explicitly excluded any design activities for the processes.

The IRT notes that a key constraint in the evaluation of the technologies was the assumption that NGNP would operate with a 750 to 800°C reactor outlet temperature (ROT). It is expected that future HTGRs, for which NGNP will be a prototype, may operate with outlet temperatures as high as 950°C. Accordingly, much of the past development work on advanced hydrogen production has assumed availability of high temperature helium directly from the reactor or via an intermediate heat transfer loop at up to 900°C. The assumption that energy would be available only at temperatures less than 750 to 800°C (given thermal losses from the reactor to the hydrogen process) played a critical role in the IRT's down-selection process. However, the IRT acknowledges that the conservatism in the assumption on ROT must be appropriately balanced against the long term potential for operation at higher temperatures and hence the ultimate opportunities that exist for advanced hydrogen production by nuclear energy. In this

regard, the IRT considered the viability of each of the processes at ROTs of both 750 to 800°C and 950°C, with the lower temperature forming the principal basis for the down-selection.

The IRT makes the following recommendations to DOE-NE:

- 1. DOE-NE should focus on the continued development of HTSE as the leading candidate for integration with NGNP in 2021. This conclusion is based upon the IRT judgment that HTSE has the highest probability of meeting the down-selection criteria described in the report, including efficient production of hydrogen at NGNP conditions.
- 2. Both HyS and SI processes exhibit attractive attributes for hydrogen production that supports not abandoning either technology for future consideration. In fact, the IRT concluded it to be prudent to continue their development through the funding of well defined R&D projects of lesser scope than those dedicated to HTSE. The IRT identified between 10 to 15 short term R&D needs for each technology.
- 3. If faced with a choice between funding short term R&D for either HyS or SI, but not both, the DOE should consider as a minimum giving higher priority in any R&D to topics that benefit both of these technologies such as continued development and modeling of the sulfuric acid decomposer.
- 4. DOE-NE should prioritize short term R&D activities that were identified by the IRT through coordination with private corporations or international organizations.

The remainder of this report provides the support for the conclusions and recommendations of the IRT. Short term R&D needs are also described for each of the three technologies.

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NOMENCLATURE

This section lists and defines the abbreviations used in this report.				
ASR	Area Specific Resistance			
CEA	Commissariat à l'Énergie Atomique			
DE	Destructive Examination			
DOE-NE	Department of Energy Office of Nuclear Energy			
FMEA	Failure Modes and Effects Analysis			
F-T	Fischer-Tropsch			
GA	General Atomics			
Gen IV	Generation IV Nuclear Energy Systems initiative.			
H2A	Hydrogen Analysis			
HHV	Higher Heating Value			
HPS	Hydrogen Production System			
HTGR	High-Temperature Gas Cooled Reactor			
HTSE	High Temperature Steam Electrolysis			
HTTR	High Temperature Test Reactor			
HyS	Hybrid Sulfur Electrolysis			
IHX	Intermediate Heat Exchanger			
ILS	Integrated Laboratory-Scale			
INL	Idaho National Laboratory			
IRT	Independent Review Team			
JAEA	Japan Atomic Energy Agency			
LHV	Lower Heating Value			
LTE	Low Temperature Electrolysis			
LWR	Light Water Reactor			

MPa	Mega-Pascal
MW	Megawatt
N/A	Not Applicable
NASA	National Aeronautic and Space Administration
NDE	Non-Destructive Examination
NERI	Nuclear Energy Research Initiative
NGNP	Next Generation Nuclear Plant
NHI	Nuclear Hydrogen Initiative
NOAK	N th -of-a-kind
NR	Not Reported
PEM	Proton Exchange Membrane
РН	Process Heat
R&D	Research and Development
RAMI	Reliability, Availability, Maintainability, and Inspectability
RH	Resistance Heating
ROT	Reactor Outlet Temperature
SI	Sulfur-Iodine
SMR	Steam Methane Reforming
SNL	Sandia National Laboratory
SRNL	Savannah River National Laboratory
SOEC	Solid Oxide Electrolysis Cell
SOFC	Solid Oxide Fuel Cells
SRNL	Savannah River National Laboratory
TDRM	Technology Development Road Map
TPD	Tons Per Day
TRLs	Technology Readiness Levels

1 INTRODUCTION

1.1 Scope

This report documents the results of an independent evaluation of the hydrogen production technologies that are being considered for integration with the NGNP. The NGNP will be a prototype commercial-scale gas cooled reactor with a pebble bed or prismatic core. The thermal power rating of the plant is expected to be up to 600 megawatt (MW). The construction of the NGNP is slated to begin in about 2015 and the plant is expected to be fully operational by 2021.

The NGNP will be designed, licensed, and constructed through a public-private partnership or alliance between the DOE and private corporations that is expected to be comprised of nuclear supply vendors, electric power utilities, and companies that are investigating the use of gas cooled reactors as a source of electric power, process heat, and/or hydrogen.

One of the DOE goals for NGNP is to demonstrate the use of HTGRs for the production of hydrogen. More specifically, approximately 50 MW of power in the form of high temperature gas, steam, or electricity will be available to demonstrate at least one advanced hydrogen production process in parallel with the plant's principal role as a generator of electricity. The demonstration of hydrogen production by the NGNP is part of a larger DOE effort to develop a range of hydrogen production methods using a variety of feedstocks and power sources including fossil, renewable, and conventional light water reactor (LWR) nuclear power. The nuclear hydrogen projects are coordinated through the DOE-NE.

The US Congress formally initiated the NGNP project as part of the Energy Policy Act of 2005 [1]. This act mandated that the NGNP project be initiated and include supporting R&D tasks, licensing activities, as well as the eventual design, construction, and operation of the NGNP prototype. The NGNP reactor will be based on concepts conceived by an international effort known as the Generation IV (Gen IV) Nuclear Energy Systems initiative. The DOE selected the INL as the laboratory to lead the development of NGNP including integration and coordination of R&D activities that are pre-requisites to the design of the plant.

Among these R&D efforts is the development of a hydrogen production process that is safe, efficient, reliable, and well-suited for integration with the NGNP. In this regard, a hydrogen production process that would be uniquely suited to the NGNP would take advantage of the high

temperatures of the reactor. More specifically, the hydrogen production technology for NGNP is expected to utilize either high temperature gas or high pressure/high temperature steam in combination with the highly efficient generation of electricity. HTGRs are capable of electric power generation efficiencies that are higher than those that can be achieved by LWRs or other conventional large fossil power plants.

The primary coolant of the NGNP is helium gas. During initial NGNP operations, helium coolant will exit the core at temperatures of about 750 to 800°C. This is well above the typical reactor coolant outlet temperatures of about 325°C achieved by LWRs. Future HTGRs may have outlet temperatures of 950°C or higher. The helium gas from NGNP may be used directly by the hydrogen production technology or used to generate steam or electricity which in turn supports the specific hydrogen production process.

1.2 Background

Today, about 96% of the world's industrial hydrogen demand of 50 million metric tons per year is met by using fossil energy and fossil feedstocks such as methane, coal, or oil. The most common production process is steam methane reforming (SMR). A large SMR plant can produce up to 200 tons of hydrogen per day. The plants are sometimes located near ammonia production plants, which in turn support fertilizer manufacturing, or near petrochemical plants where the hydrogen is required for improving the utilization of heavy feedstocks to permit the production of fossil transportation fuels such as gasoline and diesel. Since the SMR process generates a large amount of CO_2 , it represents a significant source of the world's anthropogenic greenhouse gas emissions.

Another conventional hydrogen production method particularly suited for producing high purity hydrogen for industrial processes and devices such as fuel cells, is conventional alkaline cell electrolysis. Operating at less than 100°C, the alkaline electrolysis process is called low-temperature electrolysis (LTE) and contributes to 4% of the world's hydrogen production. In this case, water is the feedstock and both hydrogen and oxygen are produced (hence the term *water splitting*).

The concept of using nuclear fission as a source of energy for hydrogen production has been studied for over 30 years. Hydrogen production has also been considered for integration into future fusion reactors. While gas-cooled HTGRs have been the focus of most of the R&D efforts in the nuclear hydrogen community, other designs such as molten salt reactors and liquid metal reactors have also been studied as sources of energy for hydrogen production.

Nuclear hydrogen production has not been demonstrated to date per se, with the exception that LTE systems draw electric power from the grid which, in turn, is partially supplied by electricity from LWRs.

The nuclear hydrogen technologies described herein represent "advanced" hydrogen production methods. Each of the technologies discussed in this report exhibit the following principal attributes:

- Potential for integration with Gen IV reactor technology such as HTGRs through use of high temperature gas and/or steam, and electricity produced at high efficiencies.
- No direct generation of greenhouse gases.
- Higher inherent hydrogen production efficiencies given the higher temperature at which hydrogen is generated (analogous to a Carnot efficiency).

The processes which are potentially suitable for integration with an HTGR could also be mated to other sources of energy which can generate high temperatures such as solar thermal energy heliostats. As such, the nuclear hydrogen processes are not nuclear fission (or fusion) dependent, but instead are high temperature hydrogen production processes. The attractiveness of using nuclear as the energy source is the large amount of power available from a nuclear plant such as an HTGR in a small footprint. This helps, in principal, justify the economics of building a large centralized advanced hydrogen production facility.

The three basic categories of high temperature hydrogen production processes are as follows:

- 1. Thermochemical processes that rely on providing energy for endothermic chemical reactions.
- 2. Electrochemical processes which are similar to LTE but operate at higher efficiency owing to their higher operating temperature.
- 3. Hybrid (also known as thermo-electrochemical) processes which produce hydrogen electrochemically, but also use chemicals other than water to optimize the characteristics of reactions in the electrolyzer.

In all three categories, water is the feedstock and hydrogen and oxygen are the products produced in an essentially stoichiometric ratio. All three processes are water-splitting processes since no other chemicals, even if used as intermediates, are consumed.

More than 300 high temperature hydrogen production processes have been identified in the literature since the 1950's [2]. Since the 1970s, a small subset of these processes has generally received the most attention in terms of laboratory scale R&D, process design and "flowsheeting" of centralized production facilities, methods for integration with an HTGR such as the NGNP, and economic assessments.

The three processes that have garnered the most attention as part of the Gen IV project in general and NGNP specifically are:

- The Sulfur-Iodine process (or SI process)¹
- High Temperature Steam Electrolysis process (HTSE)²
- The Hybrid Sulfur process (HyS)

These three processes depend upon several key processes and technologies. A brief description of each follows:

- 1. <u>SI Process</u>: A thermochemical process that incorporates: (1) endothermic decomposition of hydrogen iodide (HI) at about 450°C in the presence of a carbon catalyst to yield hydrogen and I₂, (2) recycle of the iodine to a concurrent or countercurrent column reactor where it reacts exothermically at about 120°C with sulfur dioxide (SO₂) via the Bunsen reaction to form hydriodic acid (HI) and sulfuric acid (H₂SO₄), (3) gravimetric separation of the HI and H₂SO₄, and (4) thermal/catalytic decomposition of sulfuric acid (H₂SO₄) to oxygen and SO₂ at high temperature, up to 900°C, with the SO₂ recycled back to the Bunsen reaction section. In the SI process, H₂SO₄ acts as oxygen carrier and HI as a hydrogen carrier.
- 2. <u>HTSE Process</u>: A ceramic electrolyte and electrode electrolysis process operating at up to 950°C. Water is the only reactant, and high temperatures allow for some of the energy required to split the steam (water) to be supplied as heat as opposed to electricity alone, as in LTE cells.
- 3. <u>HyS Process</u>: A hybrid process that produces hydrogen and oxygen in a polymer membrane based electrolysis cell operating at temperatures below 125°C. In the HyS process, SO₂ is used to depolarize the cell and allow it to operate at lower voltages and hence higher efficiencies and current densities as compared to higher temperature electrolysis cells. Sulfuric acid is produced along with hydrogen in the cells, with the SO₂ recycled from a sulfuric acid decomposer similar to that used in the SI process.

Figure 1-1 depicts one example of how an HTGR plant or plants could be used to support large central generation of hydrogen at 350 metric tons per day (TPD) [4]. Note that is this one study, the optimum plant configuration for each of the three hydrogen production technologies varied with respect to: (1) number of HTGRs, (2) the percentage of process heat (in the form of hot helium gas), steam, and electric power used from the reactor, and (3) the amount of supplemental electric power required form the grid.

¹ Also referred to as the IS process.

² Also known as "HTE"

Electric Pow

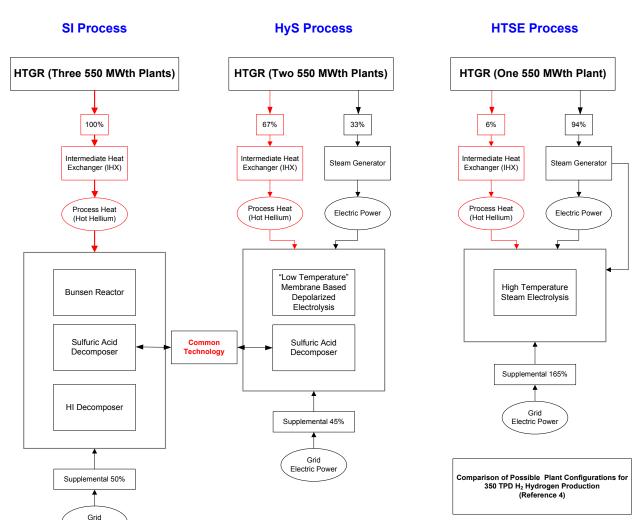


Figure 1-1. Potential Integration of Hydrogen Production Technology with HTGRs

Other processes have also shown promise for potential use in nuclear hydrogen production. These include the copper-chloride (Cu-Cl) process and another based on calcium-bromide (Ca-Br). While these two processes in particular show promise, they are less technically mature than the three current candidates being considered for NGNP.

Depending on the specific design of the process, a number of researchers have proposed that each of the three main high temperature hydrogen generation processes (SI, HyS, and HTSE) could be capable of generating hydrogen at efficiencies as high as 50% (in terms of the heating value of the product hydrogen divided by the energy required for the process)³. This compares to an efficiency of about 25 to 30% for LTE, the current principal non-greenhouse gas emitting

³ The methods by which efficiencies are calculated are discussed later in this report.

hydrogen production process. Clearly, a metric against which the attractiveness of nuclear generated hydrogen is judged is the efficiency of the process as compared with that of LTE.

Each of the hydrogen production technologies discussed in this report has evolved to varying states of maturity. HTSE has been demonstrated at laboratory scale producing over 100 liters of hydrogen per hour, albeit at lower efficiencies than those envisioned for commercial deployment. The major sections of the SI process have been demonstrated at laboratory scale through an international effort sponsored by the International Nuclear Energy Research Initiative (NERI). The HyS process has been demonstrated through bench scale testing to produce hydrogen continuously for hundreds of hours at rates that are consistent with expectations.

While it would be desirable to continue development of all three technologies in parallel over the next few years, practical considerations and budgetary limitations have led to a motivation to "down-select" the development pathway to one leading candidate with the option for continued R&D on one or both of the other two technologies at a more limited level. The limitations on budget are understandable given that it is likely to require several hundred million dollars (in 2009 USD) to complete the R&D, testing, demonstration and eventual design, build and integration of a single 50 MW hydrogen production facility into NGNP. Pursuing all three technologies is not practicable given these costs. Furthermore, some of the practical considerations that must be considered include the fact that operation of NGNP by 2021 will require preliminary design and licensing efforts for the entire plant to be completed by about 2013 or 2014. The hydrogen production technology is likely to have a significant effect on portions of the plant design and as such, the schedule for NGNP. In addition, codes and standards for the components required to build a hydrogen production facility or the NGNP interface may not exist today. Since each of the three leading candidates for hydrogen production may differ with respect to component requirements, it may not be feasible to codify all three technologies in parallel.

1.3 Motivation for Down-Selection

As discussed above, the DOE has elected to narrow the number of hydrogen production technologies that will be the focus of near term R&D. The SI, HyS and HTSE processes are the three technologies that have been chosen for consideration in the down-selection process. The overall objective is to identify the process which, based on a balance of technical, licensing, programmatic, and economic criteria, is most likely to support the NGNP in meeting a key goal - the demonstration of nuclear generated hydrogen via an advanced process.

1.4 Role of Independent Review Team

An IRT was selected by the INL and then approved by DOE-NE to evaluate the three candidate technologies and make down-selection recommendations. The team consisted of five members with backgrounds in academia and industry, and complementary technical expertise in hydrogen production, nuclear plant operations, hydrogen infrastructure, and technology development in general. The following specific tasks and activities were completed in the down-selection process:

- 1. INL prepared a Hydrogen Technology Down-Selection Methodology, Criteria and Weighting Plan. This Plan was subsequently approved by DOE-NE [3] and reviewed by the IRT.
- 2. INL identified Technology Leads independent from the IRT to compile and summarize information on each of the three candidate technologies. These Leads were identified based on their past experience in the development of the technologies both in the US and internationally. The Technology Leads coordinated the preparation of written Technology Summaries for review by the IRT members prior to the formal presentations to the IRT.
- 3. INL provided the IRT with background documents and references on NGNP, HTGR, and each of the three main hydrogen production technologies as well as some background on alternative technologies that have or are still being considered for use with HTGRs or LWRs. This included references pertaining to alternative processes such at the Cu-Cl and Ca-Br cycles.
- 4. INL sponsored a 5-day workshop from June 21 to June 26 in Denver, Colorado during which the following activities were completed.
 - A review of the Down-Selection Plan was conducted and a discussion of methodologies, criteria, and weightings that would be used in the subsequent evaluations was completed on the first day.
 - Presentations by each of the Technology Leads with support from other experts, as required. Approximately 2 to 3 hours was allowed for follow up questions and answers. In some cases, the Technology Leads were requested to provide additional supporting information after the meeting.
 - At the end of each presentation day, the IRT developed initial scores for the technology described that day. The scores were based on the criteria and weightings for the three project goals described in the Down-Selection Plan: (1) performance, (2) cost, and (3) risk.
 - Two days of evaluations that included summarizing technical issues associated with each technology. The IRT considered the following specific items during the evaluation:
 - Significant technical issues for each technology. Typically, the IRT identified from between 20 to 40 issues for each technology. The IRT then graded these issues in terms of a high, medium and low (risk) criteria.

- Past technical uncertainties, which based on the information provided to the IRT, were judged to have been largely overcome and were now deemed not significant in terms of risk to the continued development of that process
- Major advantages of the technology as compared to the others
- Recent accomplishments that have the potential to advance the cause for that technology
- "Short-term" R&D needs, that if completed successfully, would reduce the uncertainty in the risk of continued development of each of the three candidates, and
- Opportunity costs of abandoning any of the technologies in favor of another.
- In a separate evaluation, the impact of ranking each of the three technologies in terms of the suitability for integration with NGNP with a 750 to 800°C outlet temperature was considered. This was discussed in light of the potential that future HTGRs may operate at temperatures up to 950°C.
- Also considered was the progress on the development of each technology to date in terms of funding it has received. More specifically, attempts were made to levelize the maturity of the process in light of the funding received, and the specific allocation of funds to R&D, fundamental science supporting the technology, and capital costs on equipment. While no evaluation of this kind is perfect, the IRT did not want to penalize a technology for not having achieved an R&D or programmatic milestone as a result of significantly less funding or less development time.
- After reviews of the detailed assessments described above, a more quantitative scoring or grading of the three technologies was performed based on the INL QuickCompare (Microsoft Excel) spreadsheet program. Some refinement to the weightings and criteria that had been agreed to on the first day of the meeting were incorporated into the final scoring sheets. The technologies were then scored which led to a ranking of the three candidates in terms of technical risk, cost, and performance.

1.5 Organization of this Report

This report is organized as follows:

- INTRODUCTION (Section 1)
 This section defines the scope of the present evaluation, provides the necessary background, and defines the objective of the evaluation and the role of the Independent Review Team.
- 2. SUMMARY, CONCLUSIONS (Section 2) This section presents a summary of the IRT activities, the key conclusions of this evaluation, and the IRT's recommendations.

- 3. SUMMARY OF THE METHODOLOGY USED TO EVALUATE AND RECOMMEND A HYDROGEN PRODUCTION TECHNOLOGY (Section 3) This section discusses the activities completed by the IRT to prepare for and perform the hydrogen production process evaluations.
- 4. BRIEF TECHNOLOGY SUMMARIES (Section 4) This section provides a concise description of the three hydrogen production technologies that were considered by the IRT.
- 5. RESULTS OF THE HYDROGEN TECHNOLOGY DOWN-SELECTION EVALUATION (Section 5) This section summarizes the results of the evaluation of the hydrogen production technologies, including recommendations for short term R&D that should be considered for each technology.
- 6. REFERENCES (Section 6) This chapter provides a list of the references cited in this report.

2 SUMMARY, CONCLUSIONS, AND RECOMMENDATIONS

2.1 Summary

The IRT completed an assessment of the candidate advanced hydrogen production technologies considered for integration into NGNP. The assessment consisted of an evaluation of background information provided to the IRT followed by a 5-day workshop in June 2009. The workshop included presentations by technical experts for each of the three leading candidate technologies followed by question and answer sessions. The IRT generated an extensive compilation of technical issues identified through the discussions. An evaluation of these technical issues using procedures provided by the Down-Selection Plan was completed, which in turn led to each of the technologies receiving a numerical score or ranking. These rankings served as the principal basis for the IRT recommendations contained herein.

2.2 Conclusions

The following are the principal conclusions of the IRT:

- 1. Given that each technology has been the focus of many years of R&D in the US and abroad, all three technologies were concluded to be worthy of consideration for integration in the NGNP. All three technologies have routinely been selected as the leading candidates for nuclear hydrogen production out of the hundreds of technologies or cycles that have been proposed.
- 2. The IRT ranking indicates that HTSE is the hydrogen production technology that presents the greatest potential for successful deployment and demonstration at NGNP. This conclusion is primarily based on the following points:
 - a. Limited or no use of intermediate chemicals (such as I_2 or H_2SO_4).
 - b. Less complex design.
 - c. Overall progress to date.
 - d. Inherent efficiencies which are less sensitive to operating temperature, a feature that is particularly relevant with NGNP operation beginning at a relatively low HTGR temperature of 750 to 800°C.
 - e. The ability to build upon billions of dollars in SOFC research that has been completed or is pending.
 - f. The intrinsic knowledge of many of the fundamental chemical and physical processes involved in the technology.

- g. Several novel design features that have been incorporated into the latest laboratoryscale demonstration tests including integral recuperators (which may or may not be used in the final HTSE design for NGNP).
- h. The ability to accommodate load-following, as well as timely startup and shutdown time requirements.
- i. The potential for mass production of HTSE components which could be advantageous with respect to the long term life-cycle costs as compared to the other two leading candidates.
- 3. Both HyS and SI processes exhibit attractive attributes for hydrogen production that supports not abandoning either technology for future consideration. Specifically, it would be prudent to continue their development through the funding of well defined R&D projects of lesser scope than those dedicated to HTSE. The IRT identified between 10 to 15 short term R&D needs for each technology.
- 4. If faced with a choice between funding short term R&D for either HyS or SI, but not both, the DOE should consider as a minimum giving higher priority in any R&D to topics that benefit both of these technologies such as continued development and modeling of the sulfuric acid decomposer.
- 5. DOE-NE should discuss the manner in which short term R&D needs that were identified by the IRT be pursued through coordination with private entities or international organizations.

2.3 Additional IRT Recommendations

In addition to focusing on continued overall development of HTSE for advanced hydrogen production with NGNP, the IRT recommends that the following "short-term" (1 to 3 year) R&D efforts be considered by the DOE independently or in coordination with private entities or international organizations:

- HTSE R&D Areas:
 - Refine the understanding of cell/stack degradation modes and mechanisms.
 - Demonstrate pressurized cell/stack operation at a laboratory scale.
 - Continue to evaluate National Aeronautic and Space Administration (NASA) and St.
 Gobain cells as alternatives to the current preferred cell supplier.
 - Perform more thorough destructive examinations (DE) of integrated laboratory scale (ILS) stacks and recuperator components used to date in the ILS experiments.
 - Resolve the design/operating issue associated with the use of high temperature gas (up to 950°C) for future HTGR integration – develop a plan for periodic operations at NGNP with "topping cycle" to evaluate higher temperature operations.
 - Complete preliminary designs for an ILS experiment that uses pressurized manifolded stacks.
 - Identify degradation modes under pressurized operation.
 - Complete extended operations of existing stacks (>2000 hours).

- Develop concepts for pressure boundary penetrations.
- Perform fundamental modeling of physical and chemical processes in cells/stacks.
- Demonstrate operation of cells at 700°C (NGNP conditions) characterize efficiencies of the devices at these temperatures.
- Resolve need for sweep gas including hazards analyses associated with pure oxygen byproduct streams (document issues related to efficiency, safety and feasibility).
- Continue development of co-electrolysis as a companion to hydrogen generation (although not specific to NGNP, development in parallel is likely to benefit NGNP objectives).
- Demonstrate load following and cyclic operation of cells and stacks at ILS scale.
- HyS R&D Areas:
 - Investigate "control" of SO₂ gas dynamics and distribution in manifolded stacks.
 - Model "solid state" processes in electrolyzer.
 - Complete pressurized experiments of most promising cell designs for electrolyzer at temperatures up to 120°C or higher.
 - Continue testing/analyses of membrane and catalysts.
 - Finalize decision on use of gaseous or liquid phase at anode (SO₂).
 - Continue to verify/demonstrate high current operation of cells.
 - Confirm sulfur diffusion and accumulation issue has been resolved and that proposed approach to resolving the sulfur issue is achievable in larger scale cells and stacks.
 - Pursue failure mode and effects analysis (FMEA) and reliability, availability, maintainability, and inspectability (RAMI) analyses for issues such as membrane breach.
 - Develop design concepts for multiple bayonet decomposers.
 - Refine concepts for sulfuric acid decomposer bayonet NDE, catalyst recharge or regeneration, and bayonet replacement.
 - Refine modeling of bayonet heat and mass transfer (extend work already completed).
 - Complete small-scale heat transfer experiments required to support modeling of decomposer.
 - Develop plans for the scope and approach for an ILS demonstration.
- SI R&D Areas:
 - Continue fundamental chemical reaction engineering and chemical kinetics studies of the chemical processes occurring in each of the three process sections.
 - Follow-up on the potential for membrane separation processes (HI).
 - Further explore / complete work on thermodynamics of processes in Bunsen reactor.
 - Test at up to ILS scale the co-current operation of Bunsen reactor.
 - Complete further experimental studies in sections of the ILS.
 - Refine selection and investigation of catalysts for both the sulfuric acid decomposer and the HI reactor.
 - Perform laboratory-scale materials and corrosion testing including electrochemical studies.

- Develop an optimized 750°C flowsheet with high HI section recycle.
- Refine concepts for bayonet non-destructive examination (NDE), catalyst recharge or regeneration, and bayonet replacement.
- Refine modeling of bayonet heat and mass transfer (extend work already completed, conduct any and all small scale heat transfer experiments required to support modeling).

3 SUMMARY OF THE METHODOLOGY USED TO EVALUATE AND RECOMMEND A HYDROGEN PRODUCTION TECHNOLOGY

This section of the report summarizes the methodology used to evaluate and recommend a hydrogen production technology for continued development and deployment with the NGNP.

3.1 Overview

The key activities that were completed in the evaluation, in chronological order were:

- May 2008: The NGNP Project issued a report indicating that up to \$140M could be saved by focusing R&D on the most viable hydrogen production technology in addition to funding a backup technology for the near-term.
- January 2009: DOE-NE directed the INL to evaluate and recommend a nuclear hydrogen production technology for deployment with NGNP.
- February 2009: The INL issued a plan outlining the evaluation methodology for down-selection of a hydrogen generation technology [1].
- March and April 2009: INL with concurrence from DOE-NE selected an IRT.
- June 2009: DOE-NE approved the proposed down-selection criteria and weighting [2].
- June 2009: A 5-day workshop was held in Denver where Technology Leads presented information to the IRT and the technologies were scored.

The down-selection process activities are shown graphically in Figure 3-1.

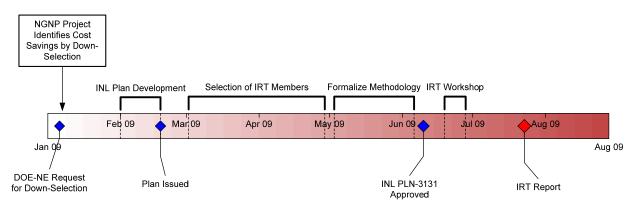


Figure 3-1. Summary of Hydrogen Technology Down-Selection by the IRT

The following provides additional detail on the methodology that was adopted by the IRT. To begin, a set of NGNP baseline characteristics was established to allow consistent evaluation. The baseline characteristics included:

- Up to 600 MWth per reactor unit
- 750 to 800°C Initial ROT
- 7 MPa Reactor Outlet Pressure
- Helium Primary Coolant
- Graphite Moderated
- Pebble-Bed or Prismatic Reactor Core
- 60-year Design Life
- Scheduled Startup 2021.

Although future reactor outlet temperatures could be as high as 950°C, the IRT evaluation was based on an initial assumed ROT in the range of 750 to 800°C and served as the primary evaluation criteria for near-term deployment of a hydrogen production process at NGNP.

The hydrogen production technology will interface with the NGNP via an intermediate heat exchanger (IHX) or steam generator. The interface parameters are as follows:

- Utilize up to 50 MWth total reactor power with any electric conversion at 40%.
- He IHX outlet to the hydrogen process at 700°C and 7 mega-paschal (MPa) pressure.
- Outlet temperature from steam generator up to 550°C at up to 15 MPa pressure.
- No contaminants from NGNP would be introduced at or upstream of this interface to affect the safety or operations of the hydrogen process.

Three technologies were the focus of this down-selection evaluation. They consisted of: (1) the HyS process, which is being developed jointly by Savannah River National Laboratory (SRNL) and Sandia National Laboratory (SNL), (2) the SI process, which is being developed jointly by General Atomics (GA), SNL, and Commissariat à l'Énergie Atomique (CEA), and (3) the HTSE, which is being developed by INL.

DOE has funded research in nuclear hydrogen production since 2003 resulting in the development of these three technologies at or near integrated laboratory scale and their consideration as final contenders in the down-selection process. A number of studies (including a NERI project by University of Kentucky, GA, and SNL) investigated the most promising technologies to guide DOE's nuclear hydrogen program [1]. Other technologies have been pursued (such as copper-chloride and calcium-bromine cycles), but these have not advanced as

far as the three technologies evaluated in this report. Furthermore, other potential technologies may exist, either in the U.S. or internationally, but do not appear to be as mature as these three for near-term deployment with NGNP.

After defining the baseline NGNP configuration, the INL developed a set of candidate evaluation criteria. A detailed, systematic review was made of primary source documents to identify potential evaluation criteria and weightings. In addition to identifying potential evaluation criteria, the review also identified constraints and general assumptions which apply to all three technologies. A significant effort was made to identify discrete criteria that could discriminate important characteristics. The criteria were developed and evaluated prior to assigning their relative weighting to ensure that the criteria were independent, complete, understandable, and discriminating. These candidate criteria were improved by review and comment from the technology leads and others. The criteria, which will be discussed in detail in Section 3.4.4, were later modified by the IRT to improve their applicability and clarity.

3.2 Selection of Independent Review Team (IRT)

The IRT was selected based on the following predefined criteria:

- 1. Up to five members total
- 2. Two or three members from a hydrogen production related industry
- 3. Two or three members from a hydrogen production related university program
- 4. Members recognized in the hydrogen field as credible
- 5. Members have hydrogen related experience and education
- 6. Members are available from April through June 2009 including a trip the week of June 22nd
- 7. No conflicts of interest exist with any of the candidate technologies

Each of the technology leads were asked to recommend hydrogen experts. DOE-NE and NGNP also provided candidate names. In addition, internationally recognized experts were included from organizations such as the National Hydrogen Association. Sixteen candidates were identified and the list was vetted to eliminate any participants that might not fully meet the membership or qualification criteria. None were eliminated.

Candidate biographies were collected along with their education and professional experience. The candidates were prioritized to allow the first calls to be to those candidates that had the best overall qualifications. The technical leads and the sponsors provided their recommendations. In one case, two of the technology leads recommended the same person. This candidate was therefore placed high on the list. In another case, a candidate worked for the Federal government

and was placed lower on the list. In one case, a candidate worked in industry but was also an adjunct professor so was placed high on the list.

INL procurement contacted the candidates to determine their interest, availability, and the lack of conflict of interest. In one case a candidate declined due to a conflict of interest regarding one of the technologies. In one case, a substitute from the same organization was contacted because this person had more experience with hydrogen production. Not all of the candidates were contacted due to five members having been found to be qualified, interested, available, and non-biased prior to reaching the end of the sixteen-candidate list. All of the contract paperwork and negotiation was conducted by INL procurement personnel.

The contracted members of the IRT are:

- Robert D. Varrin, Jr. (Team Lead), Dominion Engineering, Inc.
- Kenneth Reifsnider, University of South Carolina
- David Sandborn Scott, University of British Columbia
- Patricia Irving, InnovaTek
- Gregory A. Rolfson, Entergy, Inc.

3.3 Review Process

This team reviewed general information on hydrogen production in addition to technologyspecific information provided by the technology leads prior to the workshop held during the week of June 22 in Denver, Colorado.

In addition to the IRT, the Denver workshop included several independent observers including:

- Carl Sink (DOE-HQ)
- Melissa Bates (DOE-ID)
- Paul Pickard (SNL)
- Ed Wenzinger (MPR)
- Mike Patterson (INL)
- Charles Park (INL)

The review process started on Monday, June 22, 2009, with a discussion of the review process and criteria and a presentation by Dan Allen (Technology Insights) related to a prior independent economic evaluation of the technologies [7].

The HTSE team then presented their material. The HTSE presenters included:

- Steve Herring (INL)
- Mike McKellar (INL)
- Carl Stoots (INL)
- Joe Hartvigsen (Ceramatec, Inc.)
- S. Elangovan (Ceramatec, Inc.)

On Tuesday (June 23), the SI team provided their material with the following presenters:

- Ben Russ (GA)
- Bob Buckingham (GA)
- Lloyd Brown (GA)
- Bob Moore (SNL)
- Dan Ginosar (INL)

On Wednesday the HyS team composed of the following members presented their material:

- Bill Summers (SRNL)
- Hector Colon-Mercado (SRNL)
- David Herman (SRNL)
- Max Gorensek (SRNL)
- Bob Moore (SNL)
- Dan Ginosar (INL)

On Thursday and Friday, the IRT evaluated the information that was presented and reviewed, modified the criteria to more appropriately represent the meaningful characteristics, revised the proposed weighting, and reconciled the scores for consistency.

Following the workshop, the team drafted this report summarizing their work and recommendations.

3.4 Down-Selection Plan

A key input to the down-selection evaluation performed by the IRT was the guidance provided by INL Plan PLN-3131 "Hydrogen Technology Down-Selection Methodology, Criteria" [2]. This document describes a proposed process for evaluating and recommending a hydrogen production technology to deploy with NGNP. It includes a set of down-selection criteria and weightings and describes how these were developed. INL developed these potential criteria to help the IRT evaluate the viability of candidate technologies. INL incorporated suggestions from the Technology Leads and the IRT into PLN-3131 prior to the final publication.

The initial criteria and weightings set forth in this plan were approved by DOE-NE, and were intended to provide only a starting point for discussions and not to limit dialogue within the IRT. Later modifications to the criteria and weighting that were made by the IRT were approved by DOE-NE.

The overall evaluation methodology followed an established systems engineering selection process, as shown in Figure 3-2. Each of the steps illustrated in this figure is briefly summarized below.

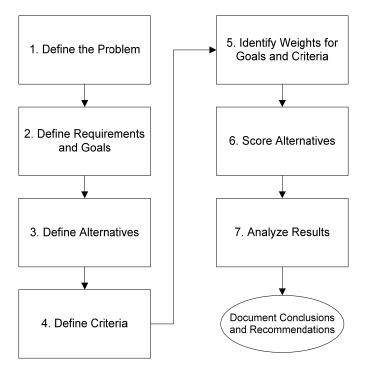


Figure 3-2. Systematic Selection Methodology

3.4.1 Problem Statement

It is considered desirable by DOE to focus limited research and development (R&D) funds on the most viable hydrogen production technologies for deployment with NGNP. The candidate problem statement was therefore defined as:

Aggressive NGNP R&D schedule and limited R&D funding require near-term selection of the most viable hydrogen production technologies.

3.4.2 Goals and Requirements

The overall goals for hydrogen production technologies, as defined by INL and DOE-NE, are summarized in Figure 3-3.

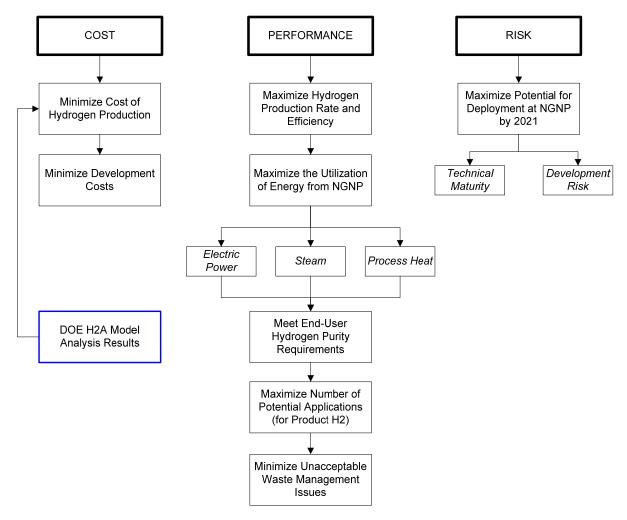


Figure 3-3. Summary of Goals and Requirements

3.4.3 Define Alternatives

As explained earlier, the down-selection process focused on the three leading technologies: (1) HyS, which is being developed jointly by SRNL and SNL, (2) SI, which is being developed jointly by GA, SNL, and CEA, and (3) HTSE, which is being developed by INL.

3.4.4 Develop Criteria

The *Hydrogen Technology Down-Selection Plan* prepared by INL in early 2009 [3] identified key references and sources of information and data that the INL had used to initially formulate selection criteria that were then incorporated into the *Down Selection Methodology, Criteria and Weighting Document* [5]. These key sources were:

- NGNP Technology Development Road Maps (TDRMs) [6]
- NGNP Hydrogen Plant Alternatives Study [4]
- Maintaining a Technology-Neutral Approach to Hydrogen Production Process Development Through Conceptual Design of the Next Generation Nuclear Plant [7]

INL also used other sources such as the 2007 National Research Council review of DOE-NE [7] and key primary NGNP documents to refine the criteria.

The Technology Neutral Report summarized overall goals for the down-selection including the consideration of both technical and economic issues. Important factors such as licensing considerations which will support NGNP and the hydrogen production process, at the INL were discussed.

In accordance with the DOE Project Management Practices (page 6-44 of Draft issued in October 2000), INL also focused on identifying discrete evaluation criteria that could:

- Differentiate between alternatives.
- Relate to project goals, objectives, and values.
- Be reasonably measurable or estimable.
- Be independent of each other.
- Be well understood by decision makers.

As a general rule, the criteria were developed, evaluated, revised prior to assigning the relative weighting to ensure that the criteria were independent, complete, understandable, and discriminating.

3.4.5 Assign Weighting

The INL assigned potential weightings to each criterion to accentuate those characteristics determined to be the most important to the selection. INL documented the weightings in the Hydrogen Technology Down-Selection Methodology, Criteria and Weighting document [5]. The potential weightings had been distributed for review by DOE-NE, the Nuclear Hydrogen

Initiative (NHI) Technical Directors, and the IRT. Comments were resolved and incorporated as appropriate, and communicated to each commenter.

As discussed later, the IRT further modified the weightings over the course of their evaluations.

3.4.6 Evaluating and Scoring the Technologies

The IRT evaluated and assigned a score to each criteria for all three technologies evaluated during a workshop in June 2009. The scoring used a scale of 1 to 5, with 5 being the most favorable. The IRT then applied final weightings to those attributes that were judged to be most important to achieve NGNP hydrogen production goals. The IRT analyzed the resulting weighted scores for consistency and relevance. Table 3-1 summarizes the criteria proposed in the down selection plan developed by the INL. This table also shows one case where the criteria were modified by the IRT during the June 2009 workshop. For instance, development cost data was subject to enough uncertainty that the IRT considered relative costs (high to low).

For the second and third criteria, Purity of Hydrogen and Flexibility to Serve Applications, the end-users and their needs were considered using information contained in the down-selection plan. These are summarized in Figure 3-4. For fuel cell operation, these criteria only apply for low temperature proton exchange membranes (PEM).

3.5 Weighting of the Evaluation Criteria

The evaluation and recommendation of hydrogen production technologies included a mathematical combination of scores and weights. As described earlier, the scores were assigned to each technology for each criterion by the IRT in accordance with criteria and score definitions. The proposed weights for each criterion as described in the Down-Selection Methodology, Criteria and Weighting Plan [3] were evaluated by the IRT and modifications approved by DOE-NE prior to use. The weights represented the relative importance of each criterion. More weight was also given to criteria for which greater confidence was demonstrated by the maturity of the data. Table 3-2 summarizes the set of weighted criteria adopted by the IRT. The weighted scores were combined to yield an overall ranking for each technology.

Table 3-1.	Summary of Evaluation Criteria
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Number	PLN-3131 Designation	Criterion	Description	Comment		5	4	3	2	1	
1	3.1	Quantity of H2 Produced		Quantitative metric in kg/day. Efficiency of production based upon NOAK deployment also to be considered.	Metric Proposed by INL	>20,000	15,000 to 19,999	12,000 to 14,999	10,000 to 11,999	<10,000	
					Metric Adopted by IRT	Same	Same	Same	Same	Same	
2	3.2	Purity of Hydrogen	Ability of process to supply hydrogen that satisfies purity	Evaluate impacts of contaminants end-users. Focus on large quantity applications such as refining (hydrotreating and hydrocracking) and	Metric Proposed by INL	No impact	Minor impact	Eliminate some applications	Eliminate most applications	Severely limits applications	
			requirements of end users.	ammonia synthesis.	Metric Adopted by IRT	Same	Same	Same	Same	Same	
3	3.3	Flexibility to Serve Applications	Nominal temperature and pressure of exit stream hydrogen as compared to	Focus on potential limits or impacts on end- users due to supply conditions (temperature or pressure). Consider impact of heating or	Metric Proposed by INL	No impact	Minor impact	Eliminate some applications	Eliminate most applications	Severely limits applications	
			end-user needs.	pressurizing product hydrogen in economic assessment.	Metric Adopted by IRT	Same	Same	Same	Same	Same	
4	3.4	Waste Management	Relative difficulty of process waste management and	Criterion considers industrial and hazardous wastes, not radioactive or mixed waste.	Metric Proposed by INL	Little or no waste	Modest amounts	Common industrial waste	Significant hazardous waste	Extreme waste issues	
	disposal.		Metric Adopted by IRT	Same	Same	Same	Same	Same			
5	3.5	Overall Cost of Hydrogen Production	Cost of hydrogen in USD(\$)/kg.	Production costs based on H2A model. Costs may take into account providing hydrogen in a suitable form (purity, T, P).	Metric Proposed by INL	<\$3/kg	\$3/kg to \$4.9/kg	\$5/kg to \$6.9/kg	\$7 /kg to \$8.9/kg	>\$9/kg	
					Metric Adopted by IRT	Same	Same	Same	Same	Same	
6	3.6	Validity of H2A Cost Assumptions	in assumptions.		Metric Proposed by INL	Very Conservative	Conservative	Reasonable	Optimistic	Very Optimistic	
		, local plicito			Metric Adopted by IRT	Same	Same	Same	Same	Same	
7	3.7	Development Cost	Cost to deploy but not operate a 50 MWt plant as part of NGNP.	Costs should not include steam generator(s) or IHX. Costs should include infrastructure and BOP costs.	Metric Proposed by INL	<\$600 Million	\$600 to \$799 Million	\$800 to \$999 Million	\$1,000 to \$1,999 Million	>\$1,200 Million	
					Metric Adopted by IRT (Relative Cost)	Low	Low-Medium	Medium	Medium-High	High	
8	3.8	Maturity- readiness of the performed independently by the IRT as		Metric Proposed by INL	>4.5	4.1 to 4.5	3.5 to 4.0	2.5 to 3.4	<2.5		
					Metric Adopted by IRT	Same	Same	Same	Same	Same	
9	3.9	Development Risk	the technology to technical, economic or	Based on TDRM Report and assessments performed independently by the IRT as required.	Metric Proposed by INL	Low Risk	Moderate Risk	Medium risk with work-arounds	High risk with possible work- arounds	Insurmountable risks and no work-arounds	
	programmatic hurdles.		Metric Adopted by IRT	Same	Same	Same	Same	Same			

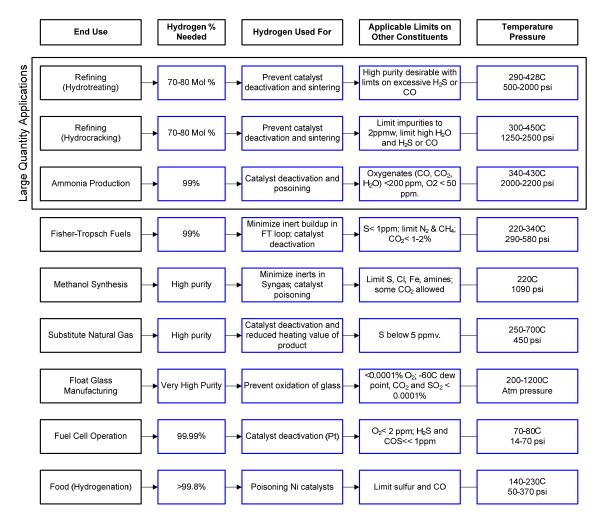


Figure 3-4. Summary of End User Requirements

Table 3-2.Criteria Weighting

	Goal Weight	Criteria Weight
Performance	35	
Quantity of Hydrogen Produced		10
Purity of Hydrogen Produced		5
Flexibility of Application		15
Waste Management		5
Cost	30	
Cost of Hydrogen		10
Validity of H2A Assumptions		10
Development Cost		10
Risk	35	
Technical Maturity		15
Development Risk		20
Total	100	100

3.6 Additional Considerations by the IRT

In addition to the methodology set forth by the INL in the Down Selection Plan, the IRT considered a number of other factors in its evaluation of the three candidate hydrogen production processes.

- <u>Stakeholder Inputs</u>: A general consideration of the degree to which each technology satisfies the needs and wants of various stakeholders including DOE, INL, national laboratories, universities, end-users and the public.
- <u>Process Efficiencies</u>: An independent evaluation of efficiencies of each process including calculation of efficiencies at both 950°C and 750 to 800°C (efficiencies for all technologies at either 750 or 800°C NGNP conditions were not available). Process efficiencies were calculated using both the lower heating value (LHV) and the higher heating value (HHV), the difference being approximately the effect of the heat of vaporization of water.
- <u>Incorporation of a Topping Cycle into the 50 MWt Facility at NGNP</u>: Assessment of the need for and cost of supplementing the energy provided by NGNP at about 710 to 760°C with a "topping cycle" that could provide hot helium or steam at temperatures up to 900°C or higher for those technologies that require higher temperatures for efficient or practical operation. An example would be the sulfuric acid decomposer for either the HyS or SI processes. In this regard, the topping cycle would be a feature of the NGNP but long term deployment of the hydrogen production process would be with HTGRs operating with ROTs up to 950°C.
- <u>Longer Term Deployment/Integration with HTGRs</u>: Consideration of the most viable hydrogen technologies beyond 2021 taking into account higher ROTs.

In addition to considering the above, the IRT reviewed but did not weigh heavily the following:

- <u>Alternative Applications for the Underlying Technology:</u> In a number of cases, the inherent hydrogen production technology being considered for NGNP has the potential to support non-hydrogen generation applications. This includes use of HTSE cell technology for co-electrolysis of CO2 in support of greenhouse gas reduction and petrochemical processes such as production of syngas or Fisher-Tropsch (F-T) liquid feedstocks and fuels.
- <u>Crosscutting Technologies:</u> While the IRT was certainly aware of significant opportunities for reducing the development costs and technical risks by weighing the value of crosscutting with other technologies, it was not a formal evaluation criterion. A good example was the potential benefits to the development of HTSE owing to work in SOFCs and the benefits to HyS from PEM fuel cells.
- <u>Alternate Energy Sources:</u> For all of the technologies, the potential for using non-nuclear energy sources exists. An example would be concentrated solar heliostats for the SI process. The IRT discussed these opportunities but the potential for development of the process using alternate energy sources was not a formal evaluation criterion.

Finally, as part of the preparations for the IRT workshop, each team member was provided an edited copy of the "Shaw Study" report [4]. This report was considered an important background reference given that it was the most complete evaluation of the comparative costs of

the hydrogen production technologies to date. It includes detailed consideration of: (1) capital costs, (2) operating costs, (3) technical risk, (4) safety, and (5) operability of the process. A single "flowsheet" or plant configuration was developed for each of the three hydrogen production technologies. Only limited credit was taken for potential technology breakthroughs that could occur between 2009 and the final design of the process somewhere in the 2013 to 2015 timeframe.

Because there had been some technical discrepancy between the plant configurations and assumptions used in the Shaw Study report (including overall process efficiencies, availability of raw materials, etc.), and the corresponding information developed more recently by the Technology Leads, a decision was made to provide only selected sections of the final Shaw Study report to the IRT prior to the workshop. In general, the copies provided to the IRT were edited to remove conclusions that would have potentially influenced an independent evaluation.

At the end of the workshop, full copies of the study report were provided to the IRT members, but only after scoring of the hydrogen production technologies had been completed.

4 BRIEF TECHNOLOGY SUMMARIES

This section provides brief summaries of the three candidate hydrogen production technologies for NGNP listed below:

- 1. <u>SI Process:</u> A thermochemical process that incorporates: (1) endothermic decomposition of hydrogen iodide (HI) at about 450°C in the presence of a carbon catalyst to yield hydrogen and I₂, (2) recycling of the iodine to a concurrent or countercurrent column reactor where it reacts exothermically at about 120°C with sulfur dioxide (SO₂) via the Bunsen reaction to form hydriodic acid (HI) and sulfuric acid (H₂SO₄), (3) gravimetric separation of the HI and H₂SO₄, and (4) thermal/catalytic decomposition of sulfuric acid (H₂SO₄) to oxygen and SO₂ at high temperature, up to 900°C, with the SO₂ recycled back to the Bunsen reaction section. In the SI process, H₂SO₄ acts as an oxygen carrier and HI as a hydrogen carrier.
- 2. <u>HTSE Process:</u> A ceramic electrolyte and electrode electrolysis process operating at up to 950°C. Water is the only reactant, and high temperatures allow for some of the energy required to split the steam (water) to be supplied as heat as opposed to electricity alone as in LTE cells.
- 3. <u>HyS Process:</u> A hybrid process that produces hydrogen and oxygen in a polymer membrane based electrolysis cell operating at temperatures below 125°C. In the HyS process, SO₂ is used to depolarize the cell and allow it to operate at lower voltages and hence higher efficiencies and current densities as compared to higher temperature electrolysis cells. Sulfuric acid is produced along with hydrogen in the cells, with the SO₂ recycled from a sulfuric acid decomposer similar to that used in the SI process.

Further descriptions of these processes and a discussion of technical uncertainties identified by the Technology Leads or the IRT follows.

4.1 SI Process

This section summarizes the SI thermochemical water splitting process that is considered a hydrogen production technology to deploy with the NGNP. The description below is a synopsis of the Technology Summary prepared for the IRT by the SI Technology Lead.

4.1.1 Process Description

As discussed in Section 2, the SI cycle has often been characterized as one of the most promising cycles for coupling to a HTGR because of its high efficiency at high temperature, and its potential for large central generation of hydrogen at scales comparable to or larger than current SMR based processes.

For example, the Japan Atomic Energy Agency (JAEA) (formally JAERI) selected the SI process for development and has successfully completed bench-scale demonstrations of the SI process at atmospheric pressure [1]. JAEA also plans to proceed with pilot-scale demonstrations of the SI process and eventually plans to couple an SI demonstration plant to its High Temperature Test Reactor (HTTR) [2, 3].

As part of an international NERI project, GA, SNL, and CEA have performed integrated ILS demonstrations of the SI process at prototypical temperatures and pressures. This demonstration was performed at the GA facilities in San Diego, CA and concluded in April 2009. This ILS is briefly discussed later in this section.

As shown in Figure 4-1, the SI process consists of three primary chemical reaction sections that result in the dissociation of water into hydrogen and oxygen;

HI Decomposition: Net Reaction	$2HI \rightarrow I_2 + H_2$ $H_2O \rightarrow H_2 + \frac{1}{2}O_2$	$(T > 350^{\circ}C)$
H ₂ SO ₄ Decomposition:	$H_2SO_4 \rightarrow SO_2 + H_2O + \frac{1}{2}O_2$	(T > 800°C)
Bunsen Reaction:	$I_2 + SO_2 + 2H_2O \rightarrow 2HI + H_2SO_4$	(T ~ 120°C)

All three reactions are operated under conditions of chemical equilibrium.

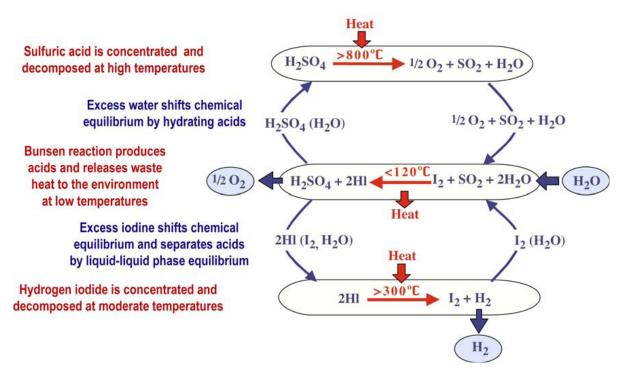


Figure 4-1. SI Thermochemical Water Splitting Process

The process involves decomposition of sulfuric acid and hydrogen iodide, and regeneration of these reagents using the Bunsen reaction. Process heat is supplied at temperatures equal to or greater than 700°C to concentrate and decompose sulfuric acid. The exothermic Bunsen reaction is performed at temperatures near 120°C and releases waste heat to the environment. Hydrogen is generated during the decomposition of hydrogen iodide, using process heat at temperatures greater than 300°C. Energy inputs to the process are heat to the endothermic H₂SO₄ and HI decomposition reactions and electrical energy required for pumping process fluids and heat pumps. Figure 4-2 shows a simplified process flow diagram of the SI cycle. The product hydrogen gas is produced at a pressure of 4.0 MPa (580 psi).

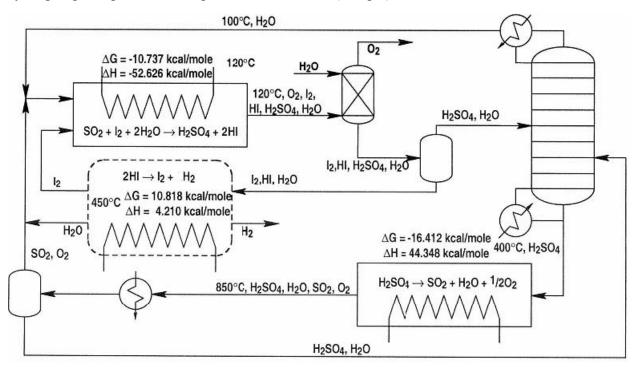


Figure 4-2. Simplified SI Process Flow Schematic

The Hydrogen Production System (HPS) design illustrated in Figure 4-2 is based on the design developed in 2003 [4]. More recent designs reflect optimization of the flowsheets to increase the hydrogen production rate and to improve efficiency.

The SI HPS design is organized into subsystems according to the three chemical reactions discussed above. These subsystems are referred to as Sections 1, 2, and 3.

Section 1 (Bunsen reaction) includes all the process equipment associated with production of the aqueous sulfuric acid phase and the hydrogen iodide ($HI/I_2/H_2O$) phase. Section 1 also includes equipment to purify the oxygen before release.

Section 2 concentrates the aqueous sulfuric acid phase and then decomposes the concentrated acid. The decomposition products and the water removed from concentrating the acid are returned to Section 1.

Section 3 concentrates and decomposes hydrogen iodide. Section 3 also includes equipment to purify the product hydrogen gas. Approximately two-thirds of the fresh water required for hydrogen production is supplied to Section 3 and the remainder is supplied to Section 1.

All of the heat from the HTGR (or NGNP) reactor is transferred via a hot helium stream to Section 2 via an IHX. The sulfuric acid vaporizers and decomposers are operated at a pressure slightly lower than that of the secondary helium coolant in order to prevent chemical contamination of the heat transfer loop while minimizing pressure differentials across these components.

The sulfuric acid vapors are decomposed in two steps;

 $\begin{array}{ll} H_2 SO_4 \leftrightarrow SO_3 + H_2 O & (T > 450^{\circ}C) \\ SO_3 \rightarrow SO_2 + \frac{1}{2}O_2 & (T > 800^{\circ}C) \end{array}$

The first reaction is very fast and equilibrium is maintained, shifting to more complete decomposition as the temperature is raised or as SO_3 is removed by the second reaction. The first reaction begins to occur in an initial decomposer or vaporizer, and continues in a second, final decomposer or vaporizer. The second reaction also requires a catalyst. Conceptually the decomposers are counter-current heat exchangers with a catalyst on the heat transfer surfaces.

With regard to Section 3, two different processes have been investigated for HI decomposition. One process, referred to as extractive distillation, uses phosphoric acid to strip HI from the HI/H₂O/I₂ (HIx) mixture and to break the HI/H₂O azeotrope. The other process is referred to as reactive distillation and involves reacting the HI/H₂O/I₂ mixture in a reactive bed to affect the separation process and produce hydrogen. Extractive distillation is a proven process, but requires significant amounts of energy and many components to perform the extraction, distillation, concentration, reaction, and separation steps of the process. The kinetics of reactive distillation are still relatively unknown, but the process can be performed in a single component without requiring concentration of the acid. For the NOAK SI-Based H₂ plant design, the HI decomposition flowsheet is based on the reactive distillation process. One disadvantage of reactive distillation is that there is significant recycling of HI back to Section 1, which increases the scale of equipment needed for Section 1.

The SI process cycle was one of the baseline nuclear hydrogen production methods being investigated under the DOE NHI. The objective of DOE NHI was to conduct an ILS experiment in order to provide a technical basis for the assessment of the viability of the SI cycle for nuclear hydrogen production.

The overall ILS project objectives were as follows:

- Complete the open-loop component reaction experiments that demonstrate the key chemical processes in stand-alone mode in engineering materials of construction.
- Perform closed-loop lab scale hydrogen production experiments under prototypic conditions to provide the technical basis for evaluating the SI cycle for nuclear hydrogen production.
- Develop a process flowsheet for pilot scale plants that can be used to project process efficiency and preliminary costs.

As discussed earlier, the development of the SI cycle ILS loop has been performed as an international effort involving US National Laboratories, industry, and CEA of France. The SNL developed the high temperature sulfuric acid decomposition section, GA developed the HI decomposition (hydrogen generation) section, and the CEA developed the primary Bunsen reaction section. These three sections along with a fourth interface section have been integrated together to demonstrate closed loop operation of the SI cycle. GA was chosen by DOE to host the ILS loop.

Each section of the ILS loop is constructed on an individual skid (pallet) and all the associated process equipment is enclosed within. The walls and the tops of the skids are constructed from Lexan or similar material. The bases are made from steel and are shaped to contain any chemical spill. The four sections are linked together with pipes that transport the process chemical streams. A schematic of the ILS loop layout is shown in Figure 4-3. The three process sections are connected to each other via the interface skid, which holds the storage and feed tanks for the circulating reaction fluids. With this design, each individual section can operate on its own without the need for concurrent operation of another section to either supply or receive the process chemicals. This helps ensure the safe operation of the individual sections. A photograph of the overall integrated lab scale equipment is shown in Figure 4-4.

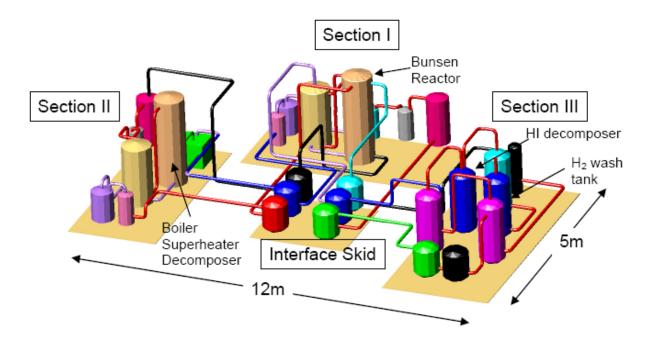


Figure 4-3. 3D Representation of SI ILS Experiment



Figure 4-4. Photograph of SI ILS Experiment

One of the key technical accomplishments of the ILS program was the design of the Bayonet Acid Decomposers.

The bayonet is shown schematically and as disassembled components in Figure 4-5. The operation of the bayonet has been described in previous reports. The basic operating parameters for the unit are the acid flow rate, the inlet acid temperature, and the maximum temperature. The bayonet is heated in the ILS using electrical heaters controlled to a maximum temperature at the outside top portion of the bayonet. The products of acid decomposition are sulfur dioxide, oxygen and water vapor. These are separated from un-decomposed acid in the lower section of the inner SiC tube.

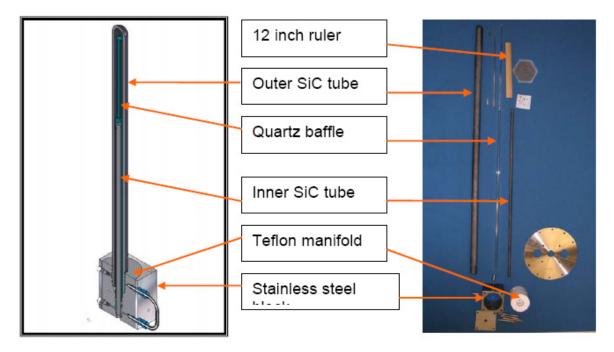


Figure 4-5. Bayonet Sulfuric Acid Decomposer

The initial results from the ILS tests in April 2008 provided some results for the integrated system, but these results also indicated major equipment modifications were required for the CEA hydrogen production process and the GA hydrogen iodide decomposition process. Although hydrogen iodide was produced in the CEA process, it was determined that additional modifications were required and a modified Bunsen reactor was installed. Once the operational parameters for the modified Bunsen reactor were identified, integrated tests with all three process sections were continued. In addition to replacement of the Bunsen reactor, a new iodine pumping system, using pressure to transfer iodine from the chemical storage skid to the Bunsen reactor, has been installed and successfully operated. The hydrogen decomposition reactor in the General Atomics hydrogen decomposition process has been replaced with a smaller unit and the

process has been successfully operated multiple times to produce hydrogen at a rate of 75 L/min (almost 5000 L/hr). The SNL acid decomposition process section underwent several modifications to improve process efficiency and integrated operation. The most significant improvement was the installation of a backpressure regulator and buffer tank between the SNL section and the CEA section to prevent pressure fluctuations in the SNL acid decomposition reactor that can result in equipment failure. The SNL sulfuric acid decomposition process using a bayonet decomposer has been successfully operated multiple times with sulfur dioxide produced at a rate of 100 to 300 L/hr.

4.1.2 Key Features and Technical Uncertainties

As a large chemical process, the attractiveness of SI is its potential to support large scale centralized production of hydrogen with good economies of scale. At high temperatures (>900°C), it offers hydrogen production efficiencies approaching 50% net not taking into account loads needed for pumping, compressing, etc. As a large chemical plant, it is not as tolerant of operating in a load following manner as other processes such as HTSE. In addition, operation of NGNP with ROTs of 750 to 800°C penalizes the SI process efficiency unless designed with very high iodine recycle flow rates.

The following is a summary of technical uncertainties associated with the SI process:

- Chemistry: While the overall chemistry of the SI process is well understood, there are a few competitive side reactions. The side reactions, which could be problematic, include the formation of sulfur or hydrogen sulfide in Section 1. Sulfur dioxide can be disproportionate to form either elemental sulfur or hydrogen sulfide along with sulfuric acid. These reactions are in competition with the much more rapid Bunsen reaction but are energetically more favorable. Any hydrogen sulfide that is formed will eventually be recycled back to the oxygen scrubbers in Section 1 where conditions are favorable to the formation of elemental sulfur from hydrogen sulfide and sulfur dioxide. Thus the ultimate questions concerning side reactions are how much sulfur, if any, will be formed, where will it go, and what will happen to it? Elemental sulfur is molten at the exit temperatures of Section 1. Any sulfur accompanying the sulfuric acid to Section 2 would accumulate in the H₂SO₄ vaporizer where it would react with the SO₃ forming SO₂. More likely, any elemental sulfur formed will accumulate in the iodine recycle stream which would require the demonstration of sulfur removal capabilities. Sulfur and iodine should be easily separable by distillation as their boiling points differ by over 250°C. The sulfur would then be fed to the outlet of the sulfuric acid decomposer where the sulfur would be oxidized to SO_2 by O_2 . Alternatively, the sulfur might be removed from the iodine by electrochemical oxidation. In either case, the maximum permissible concentration of sulfur in the iodine must be determined.
- <u>Solution Thermodynamics:</u> The reactive distillation process for generating hydrogen appears to be viable, but the current flowsheet is uncertain regarding the extent to which

the vapor-liquid equilibria of the system $HI/I_2/H_2O$ are based on models and not experimentally determined. There is also some uncertainty in the enthalpy of solution of this system. The missing data can either be determined by specific experiments or in integral form from operation of prototypical process steps. The liquid-liquid equilibria for $HI/I_2/H_2O/H_2SO_4$ are also very limited. Additional data would allow extrapolation to operating conditions outside the current operating envelope. Alternatively, one could extend the operating range through operation of prototypical process equipment.

- <u>Chemical Kinetics</u>: The chemical kinetics of HI decomposition under the conditions of the reactive distillation column are unknown. If the kinetics are too slow for economic operation, a noble metal catalyst may be needed. The kinetics need to be examined in the absence of catalysts and in the presence of supported catalysts in both gas and liquid phases.
- <u>Material Science:</u> Significant progress has been made in determining applicable materials of construction for the sulfur-iodine process. Materials applicable to Sections 1 and 2 have been, for the most part, adequately researched. The one area of Section 2 that presents some concerns is that of boiling heat transfer. The only materials resistant to concentrated sulfuric acid at the gas-liquid interface tend to be brittle materials such as siliconized cast iron and silicon carbide. Small heat exchangers have been made from such materials but full scale equivalents have yet to be developed.

The component containment (pressure boundary) materials for Section 3 will be similar to those in Section 1. The higher temperatures of Section 3 will be accommodated by the use of corrosion resistant insulating materials inside the chemical containment boundary. The heat transfer materials used in Section 1 may also be usable at the higher temperatures associated with Section 3 but this needs to be verified.

- <u>Other Uncertainties:</u> Additional uncertainties associated with the SI process are as follows:
 - Availability of sufficient iodine from existing worldwide production capabilities to support operation of multiple hydrogen production plants.
 - Efficiency at NGNP operating conditions where hot gas for the sulfuric acid decomposer may only be available at slightly above 700°C.
 - Co-location of the SI plant near the NGNP or HTGR nuclear island (safety and licensing issues).
 - Physical size of the SI plant equipment and transportation of such equipment from the fabricator to the site.
 - The potential need for a HIx membrane separation in Section 3.
 - Technology for and cost of hot helium ducts for supplying the sulfuric acid decomposer (although like the IHX, specific evaluation of this issue was not considered within the IRT scope).
 - Modeling and subsequent operation of Section 1 in co-current mode, although some published work in the area is apparently pending (University of Toulouse).
 - Sintering and deactivation of catalysts (Pt on titania) in the sulfuric acid decomposer, and need for Ir or Ru in addition to Pt.
 - Design of a manifolded bayonet decomposer, supporting modeling of both chemical and thermal phenomena.

- Methods and flowsheet designs to avoid accumulation of corrosion products.
- Demonstration of reactive distillation as part of an ILS experiment.

4.2 HTSE Process

This section summarizes the HTSE process. The description below is a synopsis of the Technology Summary prepared for the IRT by the HTSE Technology Lead.

4.2.1 Process Description

The evaluation of HTSE for hydrogen production was based on information provided by the INL, supporting information, and the expertise of IRT members. The evaluation was based on a plant configuration shown in Figure 4-6.

Baseline data presented to the IRT for the process were generated from a solid oxide electrolysis cell (SOEC) material set based on a Ni-Zirconia cermet cathode, YSZ or ScSzZ electrolyte, and lanthanum manganite anode. The planar cells were electrolyte supported (~150µm thick). Sixty-cell stacks were bundled in pairs as a basic unit by contractor Ceramatec, and packaged in an integrated laboratory scale facility at INL. The largest planar cell areas tested were 20 cm X 20 cm with about 310 cm² of active area. Production rates up to 5000 L/hr (5.6 Nm³/hr) were demonstrated in the ILS facility. The tested system included recuperation of heat from the output gases (hydrogen, residual steam, and air and oxygen stream) to heat incoming steam (and hydrogen from recycling) and sweep air streams (when used). Unattended feedback-controlled operation for 45 days was demonstrated.

Substantial computational fluid dynamics and process flow modeling were done to support the demonstration and the projected plant designs. Modeling included the effect of area specific resistance (ASR) which is a figure of merit in the sense that it controls the number of cells needed for a given required hydrogen production rate. ASR values between 0.25 and 1.25 Ohm/cm² were considered in the models; higher values were measured during service in some tests. Operating conditions approaching the thermal neutral temperature (for which the ohmic heating just matches the required process heat) were assumed in essentially all calculations.

Data included button cell testing up to about 2 W, small stack testing up to about 500 W, and integrated laboratory scale testing up to about 15 kW. The integrated laboratory scale system testing was done with planar cells having 10 cm X 10 cm dimensions, with three modules comprised of four 60-cell stacks. Electrical interconnections on every fifth cell were used to avoid stack loss in the event of a single cell failure. Heat recuperation (using an internally

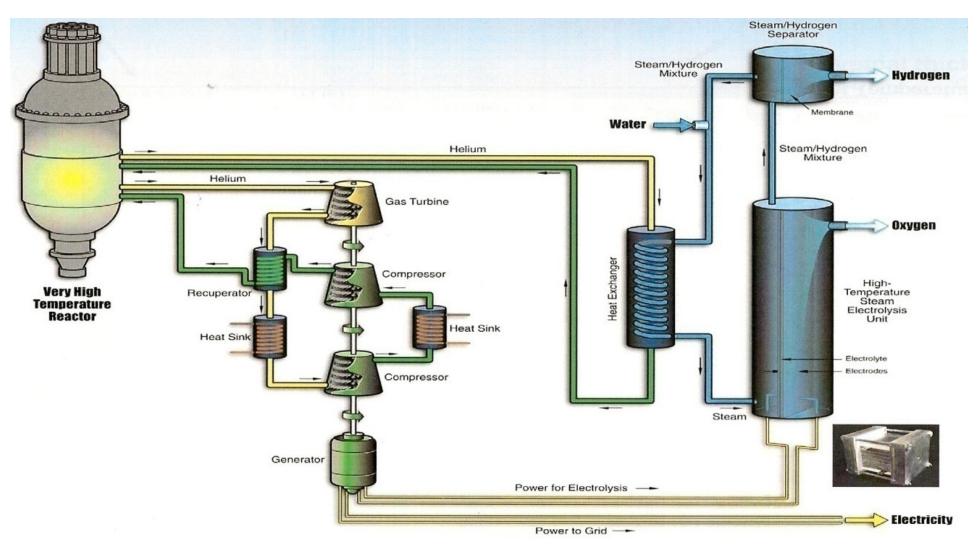


Figure 4-6. Schematic of High Temperature Steam Electrolysis (HTSE) Plant

designed, plate-fin design) reduced total electric heater power requirements. This system demonstration ran for about 1080 hours.

Coelectrolysis of CO2 (with reverse shift reaction to create syngas) was also demonstrated by the contractor. Production of syngas was in good agreement with models developed at INL. Methane production by methanation of syngas was also demonstrated by Ceramatec.

4.2.2 Key Features and Technical Uncertainties

At the most basic level, the physics and electrochemistry of the HTSE process are likely its greatest assets. Nuclear hydrogen production features high temperatures which match very well with the common operating temperatures of SOECs, especially in the 650 – 900°C range. By running on either side of the thermal neutral temperature, the SOEC can act as a pre-heater or a heat sink during operation, but the baseline system is very well matched to the temperature of the nuclear systems being considered; no major system or process cost is created in this interface. Because the SOEC is a solid state device which operates at these elevated temperatures, the electrochemical kinetics are driven by heat, and precious metal catalysts are not used, simplifying the system and reducing the cost. Additionally, the solid state ceramic device has an inherently high prospect for long-term durability, as evidenced by the semi-conductor industry. Finally, in this context, there is some developmental benefit derived in the research and development of solid oxide fuel cells, which share a similar technology, and for which there is a strong research base.

Several systems aspects also appear to be strong advantages. The systems designs considered propose keeping the SOECs at temperature, even when hydrogen is not being produced, or when power for electrolysis is varied (using nuclear heat). Under those conditions, the HTSE system has essentially unlimited and instantaneous 'load following' capability, in contrast to other processes considered. The HTSE systems appear to scale easily, and are modular by nature, allowing flexibility in design and operation. Process sensitivity studies conducted by INL indicate that HTSE shows remarkably little change in process (or plant) efficiency or product price as a function of input variables like ROT in comparison to other technologies. And finally, the possibility of using a HTSE system for coeletrolysis of CO_2 and the co-production of syngas is a unique feature.

Despite the aforementioned advantages, there are also some important limitations to the HTSE process. For example, degradation observed during service in the baseline systems tested to date was unacceptable. ASR values grew sharply in several cases and internal micro-cracking (and

delamination) were observed. However, there is evidence that these problems are engineering problems that can be solved in the relatively short term. The INL support contractor (Ceramatec) was able to introduce a new air-side electrode material and interconnect coating which demonstrated stable operation (no increases in ASR) after 500 hours of service. Other research partners (e.g. Versa Power) claim to have low degradation rates in long-term electrolysis mode. Other materials advances are expected as the mechanisms of degradation are more fully understood. Fundamentally new designs are also showing remarkable promise. The Bi-Supported Cell design developed by the research group at NASA Glenn was also tested in button cell format by INL. The nano-morphology of that cell is symmetric, eliminating thermal displacement gradients during temperature transients. The freeze-case microstructure also accommodates gas transport without channel plates, greatly reducing the size and mass of the cells. Current densities in the Bi-Supported Cells were some of the highest recorded, and the ASR remained at fractions of Ω -cm² during the test. Other partners in the development team are also providing support for this development.

In addition to an obvious need for research and development in the area of degradation mitigation, other needs for this technology include demonstration of pressurized operation, and scale-up of the basic planar size of the cells in the stack. More generally, manufacturing of large-geometry ceramic films, membranes, and material systems is a general challenge, albeit a familiar one. It should be mentioned that there is currently a major thrust in the additive manufacturing area that is moving related technology sharply forward at this point. This development may well be leveraged for the purpose of HTSE scale-up. Collaboration with the SOFC community may also provide support. The DOE Solid State Energy Conversion Alliance program, the largest single SOFC program in the U.S., has initiated an extensive research effort part of which is dedicated to degradation; degradation rates in SOFCs are typically ten times lower than those currently seen in many SOEC tests.

Additional uncertainties associated with the HTSE process include:

- Final design and lifetime of the cell stack interconnects.
- Design challenges associated with differences in coefficient of thermal expansion of stack components.
- Mass production and integration of 50 cm x 50 cm cells (required to improve the economics of HTSE) into reliable stacks under HTSE conditions.
- Pressurized operation of stacks.
- The need for use of sweep gas on the cathode side of the cells.
- Maintaining uniformity in temperature, flows, and gas compositions in large stack configurations.

- A full understanding of the consequences of cell/stack failures.
- Design of electrical and steam/process gas feed-throughs for large pressurized stack arrays.
- Oxygen safety issues in the event that sweep gas is not used.

4.3 HyS Process

This section summarizes the HyS process. The description below is a synopsis of the Technology Summary prepared for the IRT by the HyS Technology Lead.

4.3.1 Process Description

The evaluation of the hybrid sulfur process (HyS) for hydrogen production was based on information provided by the team led by SRNL, which included SNL, INL, U. South Carolina, Penn State, DuPont, Giner Electrochemical, Westinghouse PBMR, North West University (S. Africa), and collaborators in Korea and France. The evaluation was based on a plant configuration shown in Figure 4-7.

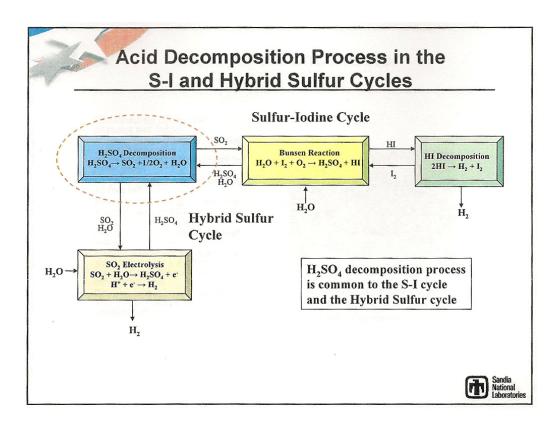


Figure 4-7. Schematic of Hybrid Sulfur Process (HyS) as Represented by Sandia National Laboratories

The integration of the HyS process with a nuclear heat source is shown schematically in Figure 4-8. In contrast to the SI process, the HyS process has only two main reaction steps, and involves only S-H-O compounds. In addition, the process uses a low temperature (PEM) water electrolyzer to generate hydrogen and sulfuric acid. The anode of the electrolyzer is depolarized by SO₂, reducing the standard cell potential from 1.23 V to 0.158 V. High temperature acid decomposition to recover the SO₂ for the next cycle of input with H₂O is required. Optimizing heat input for acid concentration and acid decomposition is a key feature of plant design. Acid vaporization and decomposition was demonstrated by SNL as part of the sulfur iodine process effort. Data reported to the team were generated primarily for the PEM electrolyzer operation for 100 hours was demonstrated. Typical running conditions were ambient up to 800°C, and up to 600 kPa. Target operating conditions are 0.6V, 500 mA/cm² at 120°C.

In addition to single cell data, through a partnership with Giner, a bi-polar 3-cell stack with 160 cm² active area per cell was designed and tested. That stack operated for 72 hours with a maximum hydrogen output of 86 L/hr, demonstrating an 8X scale up from the single cell tests. In all, 37 single cell configurations were tested.

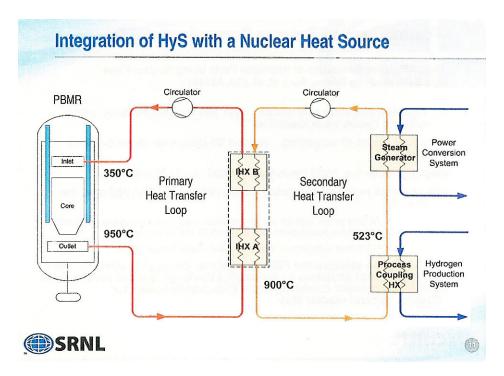


Figure 4-8. Schematic of Hybrid Sulfur Process (HyS) Plant Arrangement

4.3.2 Key Features and Technical Uncertainties

The HyS process is essentially the marriage of two technologies to achieve a simplification of the more traditional sulfur iodine process. The success of that marriage is greatly enhanced by the innovative use of sulfur dioxide to depolarize the anode of the electrolyzer, reducing the cost of the product hydrogen, by 70% according to the contractor. As in the case of HTSE, the HyS process benefits from parallel efforts by various agencies and industries to develop PEM fuel cells, especially polymer membranes and catalysts. In that context, PEM development is generally at a more advanced stage of technology readiness than the SOFC technology which is a companion of the HTSE technology.

However, barriers to the HyS process success are substantial. To achieve operational goals and in order to avoid sulfur buildup in certain elements of the chemical cycles, it is likely that a successful process would have to operate above 120°C, the nominal melting point of sulfur. The contractors were successful in identifying and testing membranes that can operate at those temperatures (and somewhat above), including a cross linked Sulfonated Diels-Alder Polyphenylene membrane (developed by SNL) for which limited data show operating conditions exceeding the targets (~1 A/cm2 at 0.7V). In the context of the overall process, however, it is not clear that 120°C (or greater) temperature operation is possible, or necessary. Gas phase operation of the electrolyzer is being considered, but would require a significant process change.

SO2 transport through the membrane is another challenge. High ionic conductivity (essential to success of the cell) is typically accompanied by increased SO₂ transport, typically resulting in substantial sulfur buildup between the electrolyte membrane and the cathode as the SO₂ reaching the cathode can be reduced by the hydrogen gas. This was mitigated in the tests by careful operating condition controls, requiring a lowered concentration of SO₂ in the anolyte. This operating change also reduces H_2S concentrations in the product hydrogen.

Another challenge associated with the HyS process is related to catalysts used for the electrolyzer, specifically their activity and degradation. Fundamentally, sulfur is a contaminant in catalyst systems. This problem is the subject of much research, and many mitigation strategies have been developed, involving both the catalyst itself and the catalyst support materials. The addition of high melting point platinum group metals to Pt was tested by the contractors and found to improve retention of activity and stability. At high temperatures (in the present context), Pt migration and sintering can be significant, reducing the available sites for catalyst activity.

Mechanical rupture of the electrolyzer membrane was not observed or considered. In PEM fuel cells, mechanical rupture, sometimes driven by combined and interactive chemical-mechanical degradation mechanisms, is a failure mode that significantly influences design and operation. In the context of the HyS process, it is not clear what the consequences of such a rupture would be, i.e., how transport of acid through the membrane would affect operation and how such a failure mode would be managed.

Finally, scale up of cell size may be a significant challenge for the HyS process. The contractors point to the large scale operation of Chlor-alkali plants as evidence that large polymer membranes can be implemented successfully. PEM fuel cells, now widely implemented in demonstration applications including the power plants for automobiles and busses, do not have planar dimensions greater than about 0.11 m². PEM electrolyzers, which have been in use for more than 20 years, and which are more technologically similar to the projected use in HyS (e.g., operate at pressure) are usually smaller than those dimensions.

Additional uncertainties associated with the HyS process include:

- A full understanding of cell/stack failure modes and consequences (as well as mitigation) has yet to be developed.
- Extended duration testing of cells and stacks has yet to be completed. An ILS experiment is planned but not yet performed.
- Compatibility of the process with load following.
- Similar technical challenges as discussed in the description of the SI process related to the sulfuric acid decomposer (recognizing these issues may be less challenging in the absence of iodine contaminants).

5 RESULTS OF THE HYDROGEN TECHNOLOGY DOWN-SELECTION EVALUATION

This section provides a summary of the results of the evaluation of the three candidate hydrogen production technologies for NGNP performed by the IRT. The methodology used by the IRT is discussed in Section 3 of this report. The specific inputs to each of the nine evaluation criteria that were considered by the IRT are summarized below:

- <u>Quantity of Hydrogen Produced</u> The principal input to defining the quantity of hydrogen produced by each of the three processes at NGNP with 50MWth of power was the information provided in the Technology Summaries by the Technology Leads. These quantities were independently checked by either re-calculating the values or by scaling with the results presented in the Shaw Study [4]. Table 5-1 contains the production rates that were used in the evaluation.
- Purity of Hydrogen Produced A number of sources were used to assess the purity of hydrogen produced. They included information provided by the Technology Leads, open literature sources, the Shaw Study assumptions, and other INL references that had been supplied to the IRT. The purities were then summarized in term of percent hydrogen and nature of the impurities present (e.g., water, sulfur, trace iodine). The resulting purities used in the evaluation and compared to industrial process needs are summarized in Table 5-1.
- <u>Flexibility to Serve Various Applications</u> The principal input in the evaluation for each process with respect to this criterion was the flowsheet information provided in the Technology Summaries, supplemented in some cases by information obtained from the Technology Leads at the IRT Workshop. The Shaw Study was also reviewed.
- 4. <u>Waste Management</u> Information regarding the nature and quantity of waste produced by the process was obtained from the Technology Summaries and other references provided by INL to the IRT. For all three processes, confirmation of any assumptions made by the IRT was obtained at the IRT workshop during discussions with the Technology Leads.
- 5. <u>Overall Cost of Hydrogen Production</u> The evaluation of the overall cost of hydrogen production was one of the more challenging tasks undertaken by the IRT. The Team

considered published results regarding costs and inherent process efficiencies (analogous to Carnot efficiencies), and the efficiencies that might be achieved in an actual production plant producing up to 350 tons of hydrogen per day. The efficiencies of production at large plants was highly dependent on the flowsheet design, types and costs of materials of construction, economies of scale (scaling factors for both HyS and HTSE electrolyzers), maintenance costs and availability, and cost of electricity when grid power was required. The IRT also requested that Technology Insights, Inc. perform additional case studies using the same H2A model they had used as part of the Shaw Study. These case studies modified some key assumptions made in the original Shaw study based on inputs from the Technology Leads. The new cases also included not only a large plant integrated with one or more HTGRs, but the process efficiencies of the 50MWth hydrogen process at NGNP with a 750°C ROT. Large scale HTGR cases with 750°C and 950°C ROTs were included. Overall, these final analyses were used to establish as consistent a basis as possible for the evaluation of the cost criteria. The results of the LHV and HHV process efficiencies are summarized in Table 5.4. The reader is cautioned that these process efficiency results were not necessarily for the same flowsheets or plant configurations for the 750°C and 950°C cases, and as such should be considered as relative efficiencies.

- <u>Validity of H2A Cost Assumptions</u> The principal input to the evaluation of this criterion was information provided in the Technology Summaries, INL references and the Shaw Study.
- 7. <u>Development Costs</u> In the assessment of development costs, the IRT adopted a different metric from that originally proposed in the Criteria and Weighting Plan [5]. This was because the development costs presented by the Technology Leads were all significantly lower than the ranges assumed in the Criteria Matrix in Table 3-1. This was found to be the result of differences in assumptions made by the INL and the Leads regarding the scope. The IRT instead adopted a Low to High qualitative ranking.
- 8. <u>Technical Maturity-Current</u> The principal input in this evaluation was the information provided during the presentations made by the Technology Leads.
- <u>Development Risk</u> The principal input to the IRT evaluation of development risk was information provided by the Technology Leads and the TDRM references and previous TRL ratings of the technologies [6].

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Table 5-1. Case Studies for Candidate Hydrogen Process (Large Scale Generation) and the Integration with NGNP

			HTSE HyS						SI			
			SHAW	(Note 1)	INL (Note 2)	SHAW	(Note 1)	SRNL (Note 2)	SHAW	(Note 1)	GA (Note 2)	
			NC	DAK	NGNP	NC	DAK	NGNP	NC	NGNP		
Key Assumptions	Reactor Thermal Rating (MWth)		550	600	Up to 600	550	600	Up to 600	550	600	Up to 600	
	No. of Units at Site (N)		1	1	1	2	3	1	3	2	1	
	Site Thermal Rating (MWth)		550	600	Up to 600	1100	1800	Up to 600	1650	1200	Up to 600	
	ROT (°C)		950	750	750	950	750	750-800	950	750	750	
	Efficiency of Elec. Prod. By HTGR (η)		40%	40%	40% (Note 3)	34%	40%	40%	N/A	40%	40%	
	Assumed Capacity Factor (HTGR)		94%	94%	90%	94%	94%	NR	94%	94%	NR	
	HTGR Energy: PH, Electricity or Both		Both	Both	Both	Both	Both	Both	PH	Both	Both	
	Ultimate Heat Sink Temperature (°C)		20	20	20	20	20	20	20	20	20	
Process Capabilities	Hydrogen Production	Hydrogen (kg/hr)	14400	14400	535	14400	14400	433	15840	14400	483	
		Hydrogen (kg/day)	343500	343500	12842	343500	343500	10400	380160	343500	11600	
		Oxygen (kg/hr)		115200 (Note 4)	4281 (Note 4)	115200	115200	3467	126720	115200	3867 290	
	Hydrogen Product Conditions	Temp (°C)	-	26	26		77	40 2		115		
		Pressure (MPa)		.0	4.95		4			.85	12	
		Purity (%)		.94	99.917	-	00	99.98	9	99.953		
		Principal Impurit(ies)	W	ater	Water			H ₂ S, SO ₂ , Water	Wat	Water		
	Flexibility	Turndown (x to 100%)	Good				Limited		Limited			
		Load Following (Ease)	Good			Limited				Limited		
		Base/Peaker		Either			Primarily Base		Primarily Base		1	
Process Inputs	From NGNP/HTGR	Heat (hot gas) (Mw _t)	88	83	3.1	712	1239	18.7	1650	1077	36.4	
	(Total for N-Pack)	Elec. Plant (Mwt)	440	517	46.9	388	561	31.3	0	123	13.6	
		[Elec Plant Mw _e]	176	207	18.8	133	224	12.5	0	49	5.5	
		Elec. Import (Mwe)	365	328	0	198	12	0	330	157	0	
		Percent Process Heat	6%	6%	6%	46%	68%	37%	67%	68%	73%	
	Gas Temperatures/Excess Energy	T _{supply} (°C)	910	710	700 (Note 6)	910	710	700 (Note 6)	910	710	700 (Note 6)	
	(if HPS returns hot He at >350C)	T _{return} (°C)	829	629	238 (Note 9)	522	522	523	344	269 (Note 9)	350	
	(,	Excess T (Energy) Use	Elec.	Elec.	None	Elec.	Elec.	Elec.	None	Elec.	None	
		Excess T (Energy) (MWe)	176	207	N/A	133	224	12.5	N/A	49	N/A	
	NGNP Topping Cycle	Туре	None	None	None	None	None	RH	None	None	None (Note 7)	
		Size [MWth]	N/A	N/A	Unknown	N/A	N/A	13.3	N/A	N/A	N/A	
		Needed	N/A	N/A	Unknown	N/A	N/A	Unknown	N/A	N/A	Unknown	
		Desired T (°C)			TBD			TBD			TBD	
	Feedstock	Water (kg/s)		34	1.34	34		1.08		7.4	1.21	
		Purity (µS/cm)	Not Sp	pecified	Not Specified	Ultra	a-Pure	Not Specified	Ultra	-Pure	Not Specified	
		Chemicals		one	None		ote 8)	Not Specified		te 8)	Not Specified	
Process Analysis	Efficiency (LHV or HHV) (Note 5)	HHV	38.8%	40.0%	42.2%	35.6%	31.0%	34.2%	25.2%	35.6%	38.1%	
(0.4 grid η)		LHV	32.8%	33.8%	35.7%	30.1%	26.3%	28.9%	21.4%	30.2%	32.2%	

Note 1: NOAK numbers taken from Shaw Study Report and evaluations performed by INL subsequent to the Shaw Study (750°C cases).

Note 2: NGNP numbers are taken or adapted from INL, SRNL and GA Technology Summary Reports and/or presentations for HTSE, HyS and SI, respectively.

Note 3: The actual power cycle proposed for the HTSE power plant is a Brayton cycle with projected thermal effiency of 53% - this will significantly increase the hydrogen production efficiency over the 40% efficiency assumed here.

Note 4: Oxygen generated from the HTSE process will be diluted with air if sweep gas is used in the process.

Note 5: Note that the difference in efficiency between LHV and HHV will be accentuated at lower temperatures where total energy input is lower.

Note 6: Based on a 750°C ROT.

Note 7: The lower ROT is compensated for by increased sulfuric acid recycle such that the same conversion is obtained.

Note 8: For S-I and HyS there will be catalyst replacement, acid make-up and caustic for acid blowdown neutralization. For S-I there is additionally iodine make-up.

Note 9: Proposed return temperature potentially below that expected for NGNP.

5.1 Technology Evaluations

Table 5-2 through Table 5-4 summarize the results of the evaluations of each of the three hydrogen production processes. Each table contains the following:

- 1. The technical issues for each process identified by the IRT.
- 2. Technical uncertainties that had been described previously in the open literature or in past INL studies that, based on the judgment of the IRT, were considered not to be significant.
- 3. Major positive attributes of the process.
- 4. Recent accomplishments in process R&D or demonstrations.
- 5. Process features that match well with NGNP.
- 6. Potential limitations on the process given the NGNP operating conditions including ROT of 750 to 800°C.
- 7. Short term R&D needs.
- 8. Opportunity cost of not pursuing.

5.2 Evaluation of Process Efficiencies

As discussed earlier, Table 5-1 presents three reported results of the HHV and LHV efficiencies of the three processes. The three cases were:

- 1. The original HTGR Shaw Study result with ROTs of 950°C.
- 2. A revised HTGR Study using the H2A model with an ROT of 750°C. A number of other assumptions adopted in the original Shaw Study were modified in this revision to reflect current conditions, including:
 - a. Nuclear heat costs were reduced from \$30/MWth to \$20/MWth.
 - b. Electric power reduced from \$75/MWe-hr to \$60/MWe-hr.
 - c. Capital cost for conventional equipment reduced to account for the down-turn in commodity prices since the 2008 Shaw Study.
 - d. The HTGR plant size(s) was changed from 550 MWth in the Shaw study to 600 MWth.
 - e. Process plant heat and electric power consumption per unit output (i.e. process efficiencies) from the IRT workshop were used, including updated data on the SI process.
 - f. HI section equipment in the SI system and bulk materials costs were lower for reduced tantalum content.
 - g. HTSE cell cost increased to $2,000/m^2$ Approximately doubled from the Shaw study and 33% more per unit area than the HyS electrolyzer cells.

- h. Higher resistance in the electrolysis stacks which in turn required higher voltage for the same hydrogen output and hence greater electric power is consumed in the electrolysis. This also increases the ohmic heating in the cells and correspondingly reduces the nuclear heating requirement.
- 3. A summary of the efficiencies reported by the Technology Leads for each of the three processes.

As can be seen in Table 5-1, there is a non-intuitive trend in the efficiencies as a function of temperature and plant size in these three cases, with NGNP being the smallest plant but in some cases having a higher efficiency. This highlights that the calculation of process efficiencies is highly dependent, as expected, on the assumptions used in the analyses.

The IRT reviewed these results and concluded that because of discrepancies in such items as the manner in which the flowsheets were designed, assumptions on return temperatures, and other factors in the calculations, general efficiency trends were more important than the specific values.

Also provided in Table 5-1 are assessments of:

- Hydrogen production rates.
- Hydrogen product conditions proposed by the Technology Leads (recognizing changing of these delivery conditions is quite achievable).
- Nominal hydrogen purity and form produced by each process (further recognizing additional purification processes could be implemented to satisfy specific end-users).
- Compatibility of the process with turndown (operation at varying production rates), load following, and its suitability as a large base load central hydrogen generation facility versus operation under peaking conditions (say for example at night when electricity costs are reduced).
- Percent of the energy provided by NGNP or one or more HTGRs that is in the form of electricity or process heat (or steam).
- The amount of electricity that must be supplied from the grid to support the hydrogen production.
- The potential need for a small topping cycle at NGNP to operate the hydrogen production process at temperatures above the planned ROT.
- Feedstock requirements.
- Hydrogen production efficiencies shown in Table 5-1 based on LHV are also shown graphically in Figure 5-1. Here, the general trend showing HTSE with the highest efficiency is apparent.

Table 5-2. Evaluation of the SI Process

Topic		Technical Issue	Comment/Description	Risk	Potential Resolution
		Flowsheet development and optimization	After 30 years of study, no concurrence on optimum process design or flowsheet	М	Improve understanding of intrinsic chemical reaction engineering; optimize flowsheet; laboratory studies; dynamic modeling (startup and shutdown)
		Overall complexity	While complex, not judged to be significantly more difficult that many large chemical plant unit operations	М	Improve flowsheets; continue to solicit input from AEs in the flowsheet development.
		Design, optimize, couple three large chem processes	Integration issues may be challenging, but degree of challenge no greater than that faced by many large scale chemical or petrochemical sites.	М	Improve flowsheets and further testing at ILS level.
		Cross contamination (e.g. S in HIx)	Should be resolved by conventional separation processes	М	Likely separation by conventional distillation.
		Materials (general)	Excessive need for Ta can be resolved with glass lined components; Ta coating technology for smaller fitting, instruments, parts; welding techniques for Ta- 10%W demonstrated.	н	Continued ILS level testing; fundamental electrochemical corrosion studies; collaboration with International efforts (India, Italy, Japan)
		Process Chemistry and Side Reactions	Issue such as exothermic reaction (Bunsen reactor); however, many of the process chemistry issues have been resolved, especially through ILS.		Laboratory studies, ILS experiments
		Startup and shutdown Solidification of iodine	Not fully modeled/studied for large scale plant. Potential process complication	M	Requires further dynamic modeling - testing at ILS Likely to be resolved through proper process design and operations.
		"Steep" efficiency curve	While curve slope depends on flowsheet, steeper efficiency dependence on temperature than other processes		Higher H_2SO_4 recycle - more electric power, larger components
		Electrical demand (in addition to Q)	Potential penalty on process efficiency if grid electricity used.	L	Not considered a major impact on process.
	General	Limited operating time/results from ILS	Programmatic issue	М	Further advancements may be possible in ILS with cooperation from International organizations
	G	Design of ILS (no reactive distillation)	Existing ILS could be modified for reactive distillation demonstration; alternative is to replicate past experience with reactive distillation on smaller scale.	M/H	Continued funding of ILS experiments.
		Fundamental corrosion studies (electrochemistry)	Considered required, especially for code development		Standard electrochemical techniques can be used.
		Decision on "train size" e.g., 3 trains/HTGR Potential reliance on membrane separations	Flowsheet issue Potential for electro dialysis	L L/M	Refine flowsheets Probably not required, but may be a desirable feature of HIx separations
		Materials cost	Special components (glass lined components), manifolded bayonet heaters.	М	Continue to work with suppliers on material selection.
		I&C	Not yet fully developed (needs, type, accuracy, etc.)	M	I&C needs for NGNP may be different (expanded).
		Proposed Matls of construction (glass lined, Ta lined, etc)	Vendor has proposed alternatives to "all Ta" construction. Alternatives, while not fully proven, appear reasonable.		Continue to develop concepts for components and equipment.
		Ceramic lined HIx Rxn vessel	Not yet proven; however, similar approach taken in many other chemical processes.	М	Moderate risk of development. Continue to develop concepts.
ategory		Greater safety challenges	Hazards associated with use, spills, transport of especially sulfuric acid, SO ₂ , SO ₃ .		Conduct both a standard HAZOPS analysis and USNRC SRP/Reg Guide evaluations (NUREG-0800)
		Embrittlement issues	Not fully investigated		Potentially a significant issue. Initiate laboratory scale studies and modeling of phenomena.
		Limited Load following Heat transfer	Limits process flexibility for some applications; base loading has advantages in some respects (economics) Designs yet to be optimized	M	Expand number of trains in various sections, recognizing losses in th economics of scale. Likely to be resolved with further engineering and understanding of
			Testing has been proposed and should be pursued at least lab scale.	Н	Receipto be resolved with initial engineering and understanding of reaction kinetics, fluid, and heat transfer. Would require further lab scale or ILS experiments.
	nsen	Integration of boost reactor	Potential optimized design proposed by CEA	L/M	Not considered a significant hurdle if required.
	Bu	Heat, chemical rxn, fluids modeling	Will be required to ultimately develop and optimize flowsheets.	М	Pursue laboratory scale investigations/experiments to improve understanding in these areas.
		Limited kinetics and thermo theory for some reactions/processes	Some new work expected from International organizations (University of Toulouse, Italy, India)	М	Review international efforts and develop plans for further improvements in this area.
		Reactive distillation (possible?)	Potential key to developing process and optimizing flowsheet.	Н	Lab scale followed by ILS experiments.
		Generation of corrosion products	Rates of generation (corrosion), accumulation, blowdown, removal, and periodic cleaning not yet fully defined.	н	Evaluate these issues early in the next flowsheet revisions; collect lal data as needed to understand generation, transport and removal of corrosion products.
	HIX	Corrosion	Few fundamental corrosion studies other than coupon test have been completed.	Н	Develop plan for an execute electrochemical corrosion studies; coordinate with NACE/ASTM.
		Lack of thermodynamic data	Lack of thermo data compromises flowsheet development, especially in Bunsen section	М	Continue to work with US and International labs to obtain data.
Ļ		Activated carbon catalyst (I2 absorption)	Can be resolved by HI/H2O wash		Verify proposed approach
		Catalyst life/selection/loading	Further work required - economics of need for more precious metal (other than Pt) catalysts effects economics		Continue lab scale testing; integrate to ILS; define plans for bayonet NDE, changeout, regeneration.
		Decomposer modeling/optimization/kinetics Feasibility of or need for bayonet at T<750°C	Initial results promising; extend to manifold type bayonet arrangement. Need to prove flowsheet requires bayonet. At low temperature, conventional decomposer HX) may be feasible.	M H	Modeling and mechanical design studies. Important design decision for NGNP.
	poser	Decomposer manifolding/design refinement Incomplete modeling of decomposer	Key requirement to validate bayonet concept. Modeling may be limited by range of issues including heat transfer correlation,	M M	Coordinate with HyS program Continue to improve.
	Decomposer	Regeneration of catalyst	fluids behavior, reaction kinetics, fouling assumptions, etc. Decide on regeneration versus replacement may be key to design and flowsheets.	М	Continue catalyst develop and investigate mechanisms of degradation
		FMEA	No complete FMEA exists including compensatory measures, means on detecting	М	deactivation and poisoning. Pursue FMEA analyses (along with HAZOPS)
		"Metal scavenger" technique for metals	failures, etc. Proposed, but not clear how this would be achieved	L/M	Continue to develop concepts; assess value or need.
		removal			

Table 5-2. Evaluation of the SI Process (continued)

Topic	Technical Issue	Comment/Description	Risk	Potential Resolution
	I2 requirements for 2400 MWth plant could exceed current	GA provided data to support worldwide reserves not significantly taxed by	L	
Non-Issues	world supply	up to 300 large scale HPS based on SI		
	Large scale potential (low scaling factor) K~0.7			
M · D ···	High theoretical efficiency			
Major Positives	Highly studied			
	Less stringent water quality specifications			
	Danfoss coated (Ta) components			
Recent	Bayonet concept			
Accomplishments	Ta welding, machining, joining			
	Potential support of Code Issues for Ta			
As a Technology for	Maximizes use of process heat	Potentially well suited for HTGRs as a means of large scale, central		
NGNP (Positives)		generation of hydrogen.		
	50 Mwt (not optimum for an SI process - too small)			
Limitations due to	Process inherently suited for T>800°C (even 900-950°)	Consider topping cycle to explore operations at more appropriate SI		
NGNP Constraints		temperatures (up to 950°C)		
(e.g. 750C)	Intermediate He loop cleanup	Use of three chemicals may complicate requirements for IHX intermediate		
		loop helium cleanup.		
	Kinetics of decomposer reactions	Required to optimize flowsheet and decomposer designs		
	Membrane issues	Investigate as needed		
	Thermodynamics	Required to optimize flowsheet and decomposer designs		
	Modeling	Refine models, including transients and complex multi-component equilibria		
R&D Needs (short	Reactive Distillation	Test at lab scale as a minimum		
term) [possible	Co current Bunsen	Test at lab scale as a minimum		
program funding]	Complete an ILS with reactive distillation	Plan and implement		
	Catalyst refinement	Continue current investigations		
	Material and corrosion testing	Perform electrochemical studies		
	Refined 750°C flowsheet	Required form NGNP		
	Revise flowsheets to support an ILS	May include 750°C operation of ILS (with or without topping cycle)		
Opportunity Cost	Reduced opportunity for alternate uses (solar power use)			
of Not Pursuing	Potential for losing our domestic leadership position			

Table 5-3. Evaluation of the HTSE Process

Topic	Technical Issue	Comment/Description	Risk	Potential Resolution
	Achievable ASR (current density)	Result of specific electrolyte used to date (Ceramatec)	L	Alternate designs being pursued; issue likely can be solved
	Performance Degradation	Same as ASR related issue (can be solved)	Н	Alternate designs being pursued (also other vendors VERSA)
	Interconnect lifetime	An issue; engineering not yet complete	М	Not clear exactly how will be solved, but highly probable workarounds exist
	Manifolding stacks (steam and power)	An issue; engineering not conceived	М	Concepts need to be developed
	CTE and materials issues for steam/gas	Code issues and design challenges	М	Concepts need to be developed
	Edge sealing	An issue.		Confidence exists that issue is solvable (e.g. non- Si seals)
	Cost of cells	Not yet fully known.	М	Continue to follow SOFC trends as a guide. Solicit feedback from manufacturers.
Category Category	50cm by 50cm (or larger) SOECs	Trivial to make, may not be trivial to stack; cells of 1m by 1mhigh risk	L	Technology for 50cm by 50cm likely to be available and support HTSE.
č	Pressurized Operation	Has not been demonstrated (pressure boundary as opposed to cell DP)	М	Complete tests in pressurized stack arrangement
	Sweep gas use	Not clear if this is a positive or negative. Currently an artifact of lab scale control system, handling pure O2	L	Evaluate need for sweep gas; resolve safety issues with O2 if not required/desirable.
	Uniformity of conditions (T, flows, gas comp)	Greater challenge at 50 x 50+ (ή, electrolysis, CTE, ect.)	М	Continue test and modeling
	Techniques for replacing cells/stacks (planned)	Not defined	М	Evaluate RAMI and lifecycle costs.
	Thermal cycling	Has not been demonstrated	М	Include in future test programs.
	Stack life	Uncertainties	M	Not a showstopper
	Consequence of cell/stack failure	Not clearly defined	L	RAMI analysis
	Failure analysis and modes	Not clearly defined	L	FMEA/PACT/RAMI Analysis
	Final recuperator design/licensing	Engineering issue	L	Evaluate locating within pressure boundary or as an external component.
	Scale up of integral recuperator	If external, potential issue with heat transfer/supply	L	Continued design optimization
	Insulation	Not clearly addressed	L	Validate through test/modeling.
Non-Issues	"H2-O2 Hazard"	SOFC experience, fundamentals of hydrogen hazards	L	Verify through HAZOPS
Ton-issues	Proportionally little process heat	See NGNP limitations.	L	Not clear if pro or con.
	Progress to date (incl. recent)	Partially due to funding levels as compared to others		
	No chemicals	Safety and potential cost advantage. Potential licensing advantage.		
	Leverage SOFC technology	Significant potential advantage.	-	
	Good intrinsic physics and activity (Hi Temp)	Significant potential advantage.		
	Recuperator concept Load following	Issues with scaling to larger pressurized stacks. Very good (especially with Q retention)		
Major Positives	Startup (<24 hours)	Good		
	Start stop operation	Good		
	"Q retention" behavior	Resilient or robust against RX trip		
	"Flatness" of Efficiency Curves	Advantageous should HTGRS be limited to <800°C		
	Potential "SOFC Revolution"	Strong potential benefit (e.g., manufacturing)		
	Smaller IHX	Potentially significant NGNP/HTGR advantage		
	Upgradable	Capability to incorporate technology improvement and components		
	"New" electrode performance and durability	Encouraging - resolves issues that existed even in early 2009		
Recent	Progress toward 2500 hour run	Encouraging results to date.		
Accomplishments	ILS	While not perfect, does represent progress		
	NDE and destructive exams	While additional work need, NDE has been integral to progress		
	NASA and St. Gobain cell (potential) Technical maturity	Potential additional advancement possible. Partially due to funding levels.		
As a Technology for	Potential for meeting "down-select plan" criteria	Partially due to funding levels. Strong positive for NGNP Project		
NGNP (Positives)	Benefits of SOFC investments	Strong positive for NGNP Project		
	Power at 50 MWt	Not an issue		
Limitations due to	Efficiency	Moderate reduction in value ("flat curve based on T")		
	Intrinsic ops at 750°C acceptable	Not known to be an issue		
(e.g. 750C)	Potential Ohmic heating to achieve >750°C	Not expected to be a significant issue		
	Lower use of process energy (long term)	Advantage if IHX cannot be designed or licensed.		
	Cell/stack degradation and mitigation	More testing and identification of mechanisms/physics		
	Pressurized operation	Stack loads, CTE, transients, HT, gas flows		
R&D Needs (short	Stack longevity under pressure	Needs to be proven in short term		
term)	"Extended Operations" (>2000 hour)	Underway		
[possible program	Engineering	Penetrations, pressurized design concept		
funding]	Sweep gas issue	Study (impact on eff., feasibility, safety)		
	Modeling Additional ILS Runs	Solid state issues and multiphysics		
	Operations at 700°C	As a minimum, plans should be formulated Validate curves		
	Reduced opportunity for alternate uses (e.g., co-	Not a specific IRT evaluation criteria.		
Opportunity Cost of Not Pursuing	electrolysis)			
	Potential for losing our domestic leadership position	Similar to other concepts.		

Table 5-4. Evaluation of the HyS Process

Topic					
	T	Technical Issue Sulfur diffusion/accumulation	Comment/Description Recent results suggest success path	Risk H	Potential Resolution Verify recent results
		Is 120°C+ operation achievable?	PBI and SNL (SDAPP) Membranes encouraging	М	Continue testing/evaluations. Replicate Giner
		is 120 C+ operation achievable:	1 D1 and SIAE (SDALL) Weinbraites encouraging	191	results at SRNL.
		Is 120°C operation necessary?	System efficiency/sulfur removal	L/M	
		1 m x 1 m PEMs?	Manufacturability/assembly questionable	Н	To be determined
		1 m x 1 m necessary?	Not clear	L	Perform a pro-con/alternatives study
		Failure modes (understanding/mitigation)	Not clearly understood	M	Complete FMEA with current knowledge base -
					continue to evaluate
		Extended duration testing	Not yet performed	Μ	Plan and complete
		"Pin holes"/breaches	Not yet addressed	Μ	Investigate consequence. Test with simulated
	ser				breach.
	Electrolyse	Consequence of membrane rupture	Not yet addressed	Μ	Postulate and evaluate
	čtr	Cost of membranes	Not clear - widely varying estimates	L	Continue to refine for leading candidate membra
	Ele				materials
		Stack configuration (series parallel)	Not yet defined	Μ	Continue to evaluate - propose optimum
					configuration.
		Testing at design pressure (stack)	Lab limitation.	L	Resolve lab limitation (safety) and pursue testing
		Shunt currents	Potential path forward - not proven or demonstrated	M	Continue to resolve
		Membrane catalyst	Still developmental	M	Continue studies
		Migration and sintering of catalyst	Not clear if significant issue	M	Continue studies
		Nafion not part of future plan	Catalyst incorporation/sticking (e.g. PBI)	Н	Switch focus as soon as practicable to leading
Category		A shinning design meltanes at I (m A (sm2))	S::	т	candidates (higher T, P)
		Achieving design voltage at I (mA/cm2)	Significant progress	L	Continue verification
		Sulfur impurities in product	Probably not a show stopper Decide on regeneration versus replacement may be key to design and	L M	Verify removal process. Continue catalyst develop and investigate
		Catalyst life/selection/loading	flowsheets.	IVI	mechanisms of degradation, deactivation and
	1		no nonceto.	l I	poisoning.
		Decomposer manifolding/design refinement	Key requirement to validate bayonet concept.	М	Coordinate with SI program
	er	Feasibility of or need for bayonet at T<750°C	Need to prove flowsheet requires bayonet. At low temperature,	H	Important design decision for NGNP.
	bos	reasionity of or need for outjoiner at 1 4/50 C	conventional decomposer (HX) may be feasible.		important design decision for restrict.
	E	Decomposer modeling/optimization/kinetics	Initial results promising; extend to manifold type bayonet arrangement.	М	Modeling and mechanical design studies.
)ec	Regeneration of catalyst	Decide on regeneration versus replacement may be key to design and	М	Continue catalyst develop and investigate
	-	-9	flowsheets.		mechanisms of degradation, deactivation and
					poisoning.
		FMEA	No complete FMEA exists including compensatory measures, means to	М	Pursue FMEA analyses (along with HAZOPS)
			detect failures, etc.		, , , , , , , , , , , , , , , , , , , ,
		Impurity ingress, generation, collection, removal	Rates of generation (corrosion), accumulation, blowdown, removal, and	Η	Evaluate these issues early in the next flowsheet
			periodic cleaning not yet fully defined.		revisions; collect lab data as needed to understan
	5				generation, transport and removal of corrosion
	ati				products.
	Integration	Lack of Integration of PEM/H2SO4	Continue to refine concepts	Μ	Refine flowsheets
	Ē	ILS tests to date	Not expected to be limiting issue (ILS seems achievable)	Μ	Continue to pursue ILS after pressurized operation
					at T~120°C have been completed.
					Model studies/flowsheet refinements.
		Load following	Large sulfuric acid decomposer more amenable to base load generation.	M	
Non-Issues		Metal to ceramic seal in decomposer	Manifolding issues not yet addressed	М	
Non-Issues		Metal to ceramic seal in decomposer Low pressure (No pressure vessel)	Manifolding issues not yet addressed Still may require "confinement"	M	
Non-Issues		Metal to ceramic seal in decomposer Low pressure (No pressure vessel) Light weight of stacks	Manifolding issues not yet addressed Still may require "confinement" Advantage	M	
Non-Issues	<u> </u>	Metal to ceramic seal in decomposer Low pressure (No pressure vessel) Light weight of stacks Single chemical step	Manifolding issues not yet addressed Still may require "confinement" Advantage Advantage relative to SI	M	
Non-Issues		Metal to ceramic seal in decomposer Low pressure (No pressure vessel) Light weight of stacks Single chemical step Crosscutting on membrane catalysts	Manifolding issues not yet addressed Still may require "confinement" Advantage Advantage relative to SI Potential benefit to development	M	
Non-Issues		Metal to ceramic seal in decomposer Low pressure (No pressure vessel) Light weight of stacks Single chemical step Crosscutting on membrane catalysts Benefits of incorporating membrane tech.	Manifolding issues not yet addressed Still may require "confinement" Advantage Advantage relative to SI Potential benefit to development Strong potential advantage	M	
Non-Issues Major Positiv		Metal to ceramic seal in decomposer Low pressure (No pressure vessel) Light weight of stacks Single chemical step Crosscutting on membrane catalysts Benefits of incorporating membrane tech. Lower fluid recirculation (108 v. 1640 for SI)	Manifolding issues not yet addressed Still may require "confinement" Advantage Advantage relative to SI Potential benefit to development Strong potential advantage Advantage in equipment sizing/power	M	
		Metal to ceramic seal in decomposer Low pressure (No pressure vessel) Light weight of stacks Single chemical step Crosscutting on membrane catalysts Benefits of incorporating membrane tech. Lower fluid recirculation (108 v. 1640 for SI) Thermo basis for modeling H2SO4 (v. HIx)	Manifolding issues not yet addressed Still may require "confinement" Advantage Advantage relative to SI Potential benefit to development Strong potential advantage Advantage in equipment sizing/power Advantage over SI, but still not fully understood	M	
		Metal to ceramic seal in decomposer Low pressure (No pressure vessel) Light weight of stacks Single chemical step Crosscutting on membrane catalysts Benefits of incorporating membrane tech. Lower fluid recirculation (108 v. 1640 for SI) Thermo basis for modeling H2SO4 (v. HIx) Collaboration	Manifolding issues not yet addressed Still may require "confinement" Advantage relative to SI Potential benefit to development Strong potential advantage Advantage over SI, but still not fully understood Advantage, especially recent International interest	M	
		Metal to ceramic seal in decomposer Low pressure (No pressure vessel) Light weight of stacks Single chemical step Crosscutting on membrane catalysts Benefits of incorporating membrane tech. Lower fluid recirculation (108 v. 1640 for SI) Thermo basis for modeling H2SO4 (v. HIx) Collaboration Current density	Manifolding issues not yet addressed Still may require "confinement" Advantage Advantage relative to SI Potential benefit to development Strong potential advantage Advantage in equipment sizing/power Advantage over SI, but still not fully understood Advantage, especially recent International interest Potential smaller footprint	M	
		Metal to ceramic seal in decomposer Low pressure (No pressure vessel) Light weight of stacks Single chemical step Crosscutting on membrane catalysts Benefits of incorporating membrane tech. Lower fluid recirculation (108 v. 1640 for SI) Thermo basis for modeling H2SO4 (v. HIx) Collaboration	Manifolding issues not yet addressed Still may require "confinement" Advantage Advantage relative to SI Potential benefit to development Strong potential advantage Advantage in equipment sizing/power Advantage over SI, but still not fully understood Advantage, especially recent International interest Potential smaller footprint Somewhat of an advantage, but technology now far beyond 1970's work at	M	
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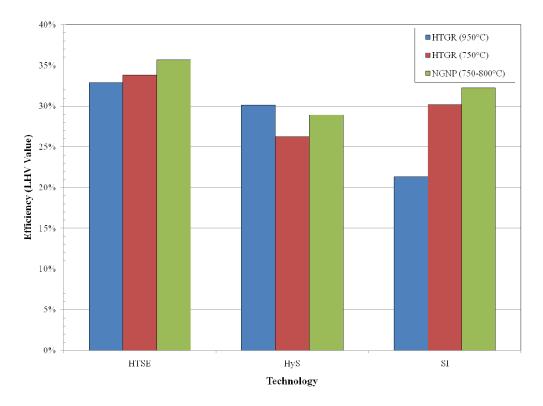


Figure 5-1. Example Hydrogen Process Efficiencies (LHV Values)

5.3 Hydrogen Production Costs for NGNP and HTGRs at 750°C and 950°C ROTs

Figure 5-2 summarizes example evaluations of hydrogen production costs in \$/kg for a HTGR plant(s) with ROTs of 750°C and 950°C respectively. These costs were generated using the H2A model based on revised inputs recommended by the Technology Leads and the IRT. As was the case with the efficiencies, the production costs were sensitive to the model inputs. As such, the IRT considered trends seen in a number of reports and analyses as opposed to absolute costs per kilogram produced.

5.4 Scoring Results for the Hydrogen Technologies

Table 5-5 summarizes the scores obtained for each of the three technologies. Based on the scoring, the IRT recommends that the DOE focus on HTSE for integration with the NGNP, with HyS as the technology that should be considered as the principal backup. If possible, short term R&D efforts associated with the SI process should also be pursued (see more discussion below).

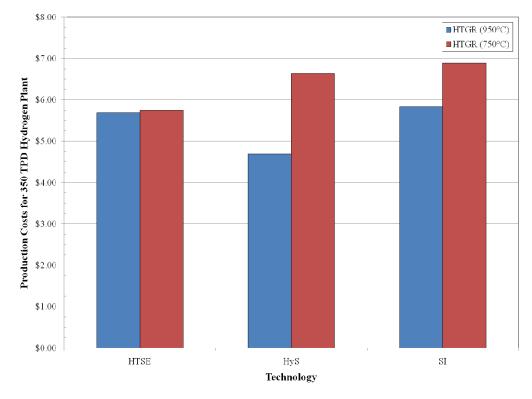


Figure 5-2. Example Hydrogen Production Costs for 350 TPD Hydrogen Plant

Table 5-6 briefly summarizes other assessments performed by the IRT that did not directly affect the scores for each. This included the potential that the hydrogen process could be used with other energy sources (concentrated solar), as well as whether there are other potential applications for the process, such as co-electrolysis in the case of HTSE.

5.5 Description of Short Term R&D Needs

This section describes short term R&D needs that the IRT recommends be considered by the DOE for each of the three technologies, with an obvious emphasis on HTSE since this is the technology recommended for continued development.

- HTSE R&D Areas:
 - Refine the understanding of cell/stack degradation modes and mechanisms.
 - Demonstrate pressurized cell/stack operation at a laboratory scale.
 - Continue to evaluate NASA and St. Gobain cells as alternatives to the current preferred cell supplier.
 - Perform more thorough destructive examinations (DE) of ILS stacks and recuperator components used to date in the ILS experiments.

Table 5-5.	Results of Scoring	g the Hydro	gen Processes
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				Weight	ings (%)		Worse <	- Scoring -	> Better		I	Ratings	6		Score	
Category	NGNP Project Goals	Number	Criteria	Goal	Criteria	1	2	3	4	5	HTSE	SI	HyS	HTSE	SI	HyS
			Quantity of H2 Produced	35%	10%	<10	10-12	12-15	15-20	>20	3	2	2	0.30	0.20	0.20
1	Performance	3.2	Purity of Hydrogen		5%	None	Almost none	Some	Most	All	5	4	3	0.25	0.20	0.15
	Perfor		Serve Various Applications		15%	Useless	Almost None	Some	Most	All	4	3	4	0.60	0.45	0.60
		3.4	Waste Management		5%	Extreme	Significant	Typical	Modest	None	4	2	3	0.20	0.10	0.15
		3.5	Cost of Production	30%	10%	>9	7-9	5-7	3-5	<3	3	2	3	0.30	0.20	0.30
2	Cost	3.6	Cost Uncertainty		10%	Unrealistic	Optimistic	Consistent	Conservative	Very Conservative	3	2	3	0.30	0.20	0.30
2	ŭ	3.7	Development Cost		10%	>1200	1000-1200	800-1000	600-800	<600	4	2	3	0.40	0.20	0.30
			RELATIVE COST			Very High	High	Med	Low-Med	Low	Low	High	Med			
3	Risk	3.8	Technical Maturity (TRLs)	35%	15%	<2.5	2.5-3.4	3.5-4	4.1-4.5	>4.5	3	1	2	0.45	0.15	0.30
	Ri	3.9	Development Risk		20%	Insurmountable	High	Medium to High	Low to Medium	Low	3.5	2	3	0.70	0.40	0.60
														3.50	2.10	2.90

3.50 2.10 2.90 Out of 5.00

Table 5-6. Additional Considerations

Supplemental Criteria	HTSE	SI	HyS
Alternative Sources of Energy	Y	М	М
Alternate Applications	Y	N	N
Cross-cutting	Y	Ν	М

Y = Yes

M= Moderate

N= No

- Resolve the design/operating issue associated with the use of high temperature gas (up to 950°C) for future HTGR integration – develop a plan for periodic operations at NGNP with "topping cycle" to evaluate higher temperature operations.
- Complete preliminary designs for an ILS experiment that uses pressurized stacks.
- Identify degradation modes under pressurized operation.
- Complete extended operations of existing stacks (>2000 hours).
- Develop concepts for pressure boundary penetrations.
- Perform fundamental modeling of physical and chemical processes in cells/stacks.
- Demonstrate operation of cells at 700°C (NGNP conditions) characterize efficiencies of the devices at these temperatures.
- Resolve need for sweep gas including hazards analyses associated with pure oxygen byproduct streams (document issues related to efficiency, safety, and feasibility).
- Continue development of co-electrolysis as a companion to hydrogen generation (although not specific to NGNP, development in parallel is likely to benefit NGNP objectives).
- Demonstrate load following and cyclic operation of cells and stacks at ILS scale.
- HyS R&D Areas:
 - Investigate "control" of SO₂ gas dynamics and distribution in manifolded stacks.
 - Model "solid state" processes in electrolyzer.
 - Complete pressurized experiments of most promising cell designs for electrolyzer at temperatures up to 120°C or higher.
 - Continue testing/analyses of high temperature membrane and catalysts.
 - Finalize decision on use of gaseous or liquid phase at anode (SO₂).
 - Continue to verify/demonstrate high current operation of cells.
 - Confirm sulfur diffusion and accumulation issue has been resolved and that proposed approach to resolving the sulfur issue is achievable in larger scale cells and stacks.

- Pursue FMEA and RAMI analyses for issues such as membrane breach.
- Develop design concepts for multiple bayonet decomposers.
- Refine concepts for sulfuric acid decomposer bayonet NDE, catalyst recharge or regeneration, and bayonet replacement.
- Refine modeling of bayonet heat and mass transfer (extend work already completed)
- Complete small-scale heat transfer experiments required to support modeling of decomposer.
- Develop plans for the scope and approach for an ILS demonstration.
- SI R&D Areas:
 - Continue fundamental chemical reaction engineering and chemical kinetics studies of the chemical processes occurring in each of the three process sections.
 - Follow-up on the potential for membrane separation processes (HI).
 - Complete work on thermodynamics of processes in Bunsen reactor.
 - Test at up to ILS scale the co-current operation of Bunsen reactor.
 - Complete further experimental studies in sections of the ILS.
 - Refine selection and investigation of catalysts for both the sulfuric acid decomposer and the HI reactor.
 - Perform laboratory-scale materials and corrosion testing including electrochemical studies.
 - Develop an optimized 750°C flowsheet with high HI section recycle.
 - Refine concepts for bayonet NDE, catalyst recharge or regeneration, and bayonet replacement.
 - Refine modeling of bayonet heat and mass transfer (extend work already completed, conduct any and all small scale heat transfer experiments required to support modeling)

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