



Nuclear Hydrogen R&D Plan

March 2004



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March 2004

**Department Of Energy
Office of Nuclear Energy, Science and Technology**

CONTENTS

1.	Hydrogen and The Role of Nuclear Energy	1-1
1.1	The DOE Hydrogen Program	1-2
1.2	Energy Sources for Hydrogen – The Nuclear Connection	1-4
1.3	Nuclear Hydrogen Initiative (NHI)	1-5
2.	Nuclear Hydrogen Development Strategy	2-1
2.1	Objective	2-1
2.2	Approach	2-1
2.2.1	Thermochemical Cycles	2-1
2.2.2	High-Temperature Electrolysis	2-2
2.2.3	Other Production Methods	2-2
2.2.4	Systems Interface	2-2
2.3	Technology Assessment	2-2
2.3.1	General Evaluation Criteria	2-2
2.3.2	Production Process Prioritization	2-3
2.4	Process Demonstration Strategy	2-4
3.	Thermochemical Cycles	3-1
3.1	Objective	3-1
3.2	Approach	3-1
3.3	Thermochemical Cycle Assessment	3-1
3.3.1	Summary of Previous Studies	3-2
3.3.2	Conclusions from Previous Studies	3-3
3.4	Thermochemical Cycle R&D Needs	3-3
3.4.1	Sulfur-Based Cycles	3-6
3.4.2	Calcium Bromide Cycles	3-7
3.4.3	Alternative Thermochemical Cycles	3-8
3.5	Thermochemical Cycle R&D Program	3-9
3.6	Thermochemical R&D Activities	3-10
4.	High-Temperature Electrolysis	4-1

4.1	Objective	4-1
4.2	Approach	4-1
4.3	High Temperature Electrolysis Assessment	4-1
4.3.1	HTE Energy Efficiency	4-2
4.3.2	HTE Production Plant Features	4-3
4.4	High Temperature Electrolysis R&D Needs	4-3
4.4.1	System Design and Trade Studies	4-3
4.4.2	HTE Cell / Module Optimization	4-4
4.5	High Temperature Electrolysis R&D Program	4-6
4.6	HTE Milestones	4-6
5.	Systems Interfaces and Balance of Plant	5-1
5.1	Objective	5-1
5.2	Approach	5-1
5.3	Systems Interface and Balance-of-Plant Assessment	5-1
5.4	Systems Interface and Balance of Plant R&D Needs	5-3
5.4.1	Nuclear Hydrogen System Design Studies	5-3
5.4.2	Heat Exchanger Design Studies	5-3
5.4.3	Intermediate Heat Transfer Loop	5-4
5.4.4	Supporting Systems Scoping Studies	5-4
5.5	Systems Interface and Balance of Plant R&D Program	5-5
6.	Nuclear Hydrogen R&D Plan Summary	6-1

Nuclear Hydrogen R&D Plan

1. HYDROGEN AND THE ROLE OF NUCLEAR ENERGY

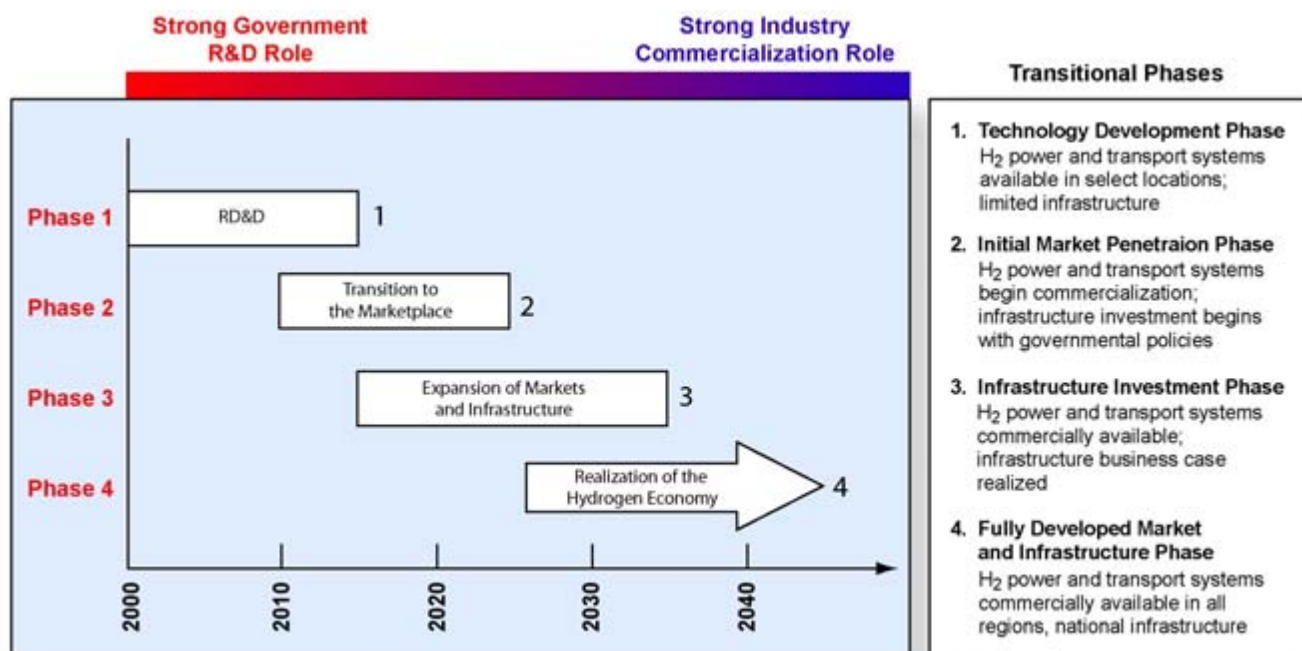
In November 2002, the U.S. Department of Energy (DOE) issued its *National Hydrogen Energy Roadmap*. The purpose of the Roadmap was to identify the activities required to realize hydrogen's potential to address U.S. energy security, diversity, and environmental needs. The Roadmap concluded that:

"Expanded use of hydrogen as an energy carrier for America could help address concerns about energy security, global climate change, and air quality. Hydrogen can be derived from a variety of domestically available primary sources, including fossil fuels, renewables, and nuclear power. Another key benefit is that the by-products of conversion are generally benign for human health and the environment."

"Despite these compelling benefits, realization of a hydrogen economy faces multiple challenges. Unlike gasoline and natural gas, hydrogen has no existing, large-

scale supporting infrastructure—and building one will require major investment. Although hydrogen production, storage, and delivery technologies are currently in commercial use by the chemical and refining industries, existing hydrogen storage and conversion technologies are still too costly for widespread use in energy applications. Finally, existing energy policies do not promote consideration of the external environmental and security costs of energy that would encourage wider use of hydrogen."

The Administration has proposed a research and development (R&D) program to accelerate the development of hydrogen technology. Successful implementation of the DOE Hydrogen Program will enable industry to make an investment decision by 2015 regarding the commercial viability of hydrogen for fuel cell vehicles. The transition to a hydrogen economy involves several phases, which are summarized in Figure 1-1.



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Figure 1-1. Phases in the development of the hydrogen economy.

1.1 The DOE Hydrogen Program

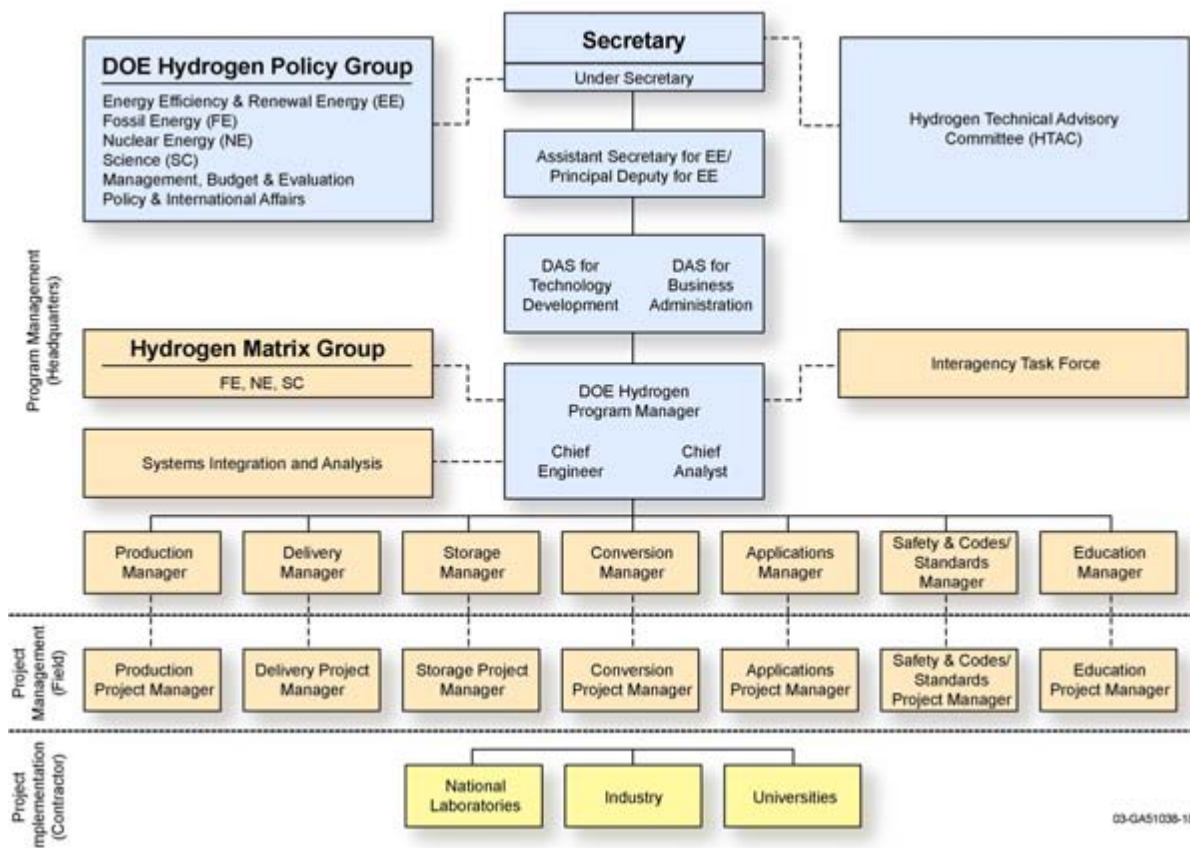
One of the advantages of hydrogen is that it can utilize a variety of feedstocks and a variety of production technologies. Feedstock options include fossil resources, such as coal and natural gas, and renewable resources, such as biomass and water. Production technologies include thermochemical, biological, electrolytic, and photolytic processes. Energy needed for these processes can be supplied through fossil, renewable, or nuclear sources. Ultimately, a spectrum of feedstocks and technologies for hydrogen production will be necessary to address energy security and climate change concerns. Therefore, the DOE Hydrogen Program will address multiple feedstock and technology options to provide effective and efficient hydrogen production for both the short and long term.

The DOE Hydrogen Program is contained within the Offices of Nuclear Energy, Science and Technology (NE), Fossil Energy (FE), and Energy Efficiency and Renewable Energy (EE), which has the lead organizational role for the DOE as shown in

Figure 1-2. These offices are now working together synergistically to accomplish the overall program goals. The immediate focus is on distributed production to meet short-term needs most cost effectively. To meet longer-term needs, there is significant effort to provide competitive renewable feedstocks and energy sources, to develop centralized production from coal with sequestration, and to develop high-temperature processes using advanced nuclear reactors.

While R&D on production technologies are carried out within various DOE offices, the DOE uses three criteria for all feedstocks and production technologies in determining which to develop. All production options must:

- Be produced from domestic resources
- Avoid the production of greenhouse gases
- Be cost-competitive with gasoline in the implementation timeframe.



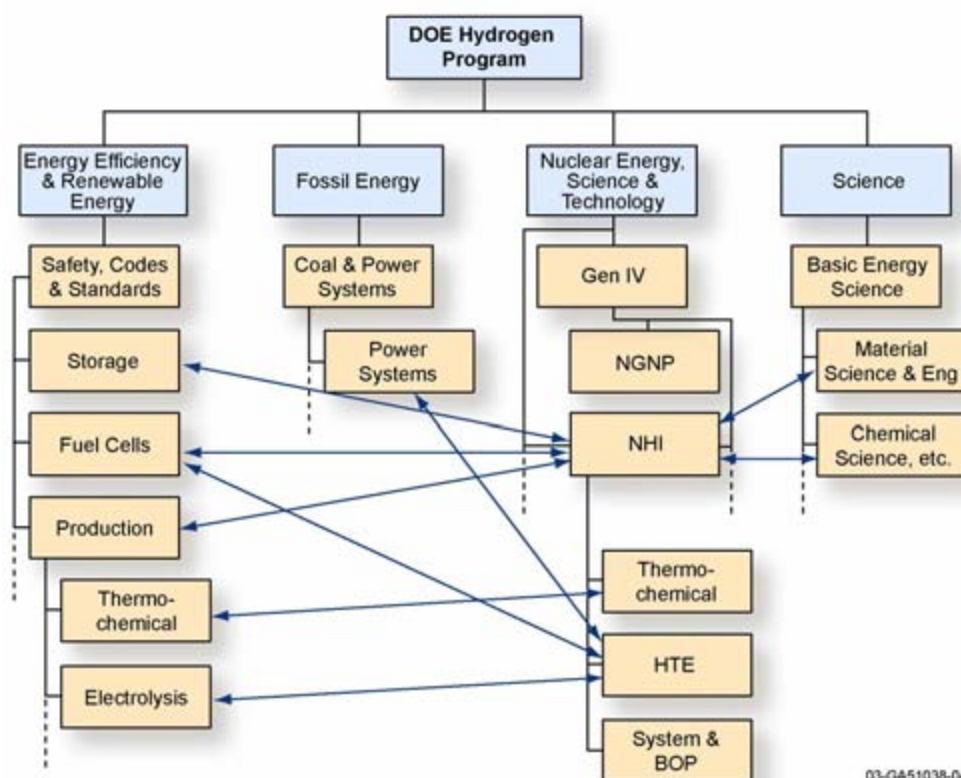
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Figure 1-2. DOE proposed organizational structure for hydrogen program.

These technical challenges and the investment required to achieve these primary goals are significant. To address the multiple challenges that face the widespread use of hydrogen, the DOE Hydrogen Program must:

- Produce hydrogen economically. Large-scale, cost-effective hydrogen production methods that make the cost of hydrogen competitive with gasoline must be developed. A significant component of this production must ultimately come from sources that are not dependent on fossil fuels or do not produce carbon emission.
- Develop storage methods for hydrogen to provide an adequate capacity for the next generation of vehicles.
- Develop technologies that use hydrogen efficiently for the wide range of industrial and transportation applications (e.g., fuel cells, internal combustion engines).
- Develop the delivery infrastructure to enable routine hydrogen use for transportation.

DOE has undertaken major research programs to address the full range of hydrogen production, distribution, storage, and utilization issues that were identified in the National Hydrogen Energy Roadmap. Hydrogen production technologies that are being addressed by FE, EE, and NE will have some technology issues in common, which will require an integrated approach to ensure that the issues are efficiently addressed. Solid oxide materials, anode and cathode materials, and sealing technologies being developed for fuel cells are relevant to high temperature electrolyzers. Thermochemical methods and basic data, and engineering materials being developed for other programs are also relevant to the nuclear hydrogen processes. Technology working groups will be formed around common research opportunities to facilitate efficient interoffice communications for these common technology areas. Figure 1-3 illustrates the flow of technology and information between DOE programs for the hydrogen production technologies being developed within the Nuclear Hydrogen Initiative (NHI).



03-GA51038-04

Figure 1-3. DOE hydrogen program elements/structure.

1.2 Energy Sources for Hydrogen – The Nuclear Connection

Hydrogen is abundant in nature but occurs primarily in stable compounds that require significant energy to produce hydrogen for use as a fuel. Hydrogen is an energy carrier, much like electricity, that requires a primary energy source to produce. Domestic energy sources that do not generate greenhouse gases and have the potential to produce hydrogen at costs competitive with gasoline will be essential components of the long-term energy supply. The DOE Hydrogen Program is investigating the potential for all of the practical energy sources for hydrogen production, including:

- Fossil sources with carbon sequestration (coal and natural gas)
- Renewable energy sources (solar, wind, and hydroelectric)
- Biological methods (biomass and biological)
- Nuclear energy.

In the long term, economics and national policy will determine the mix of energy sources that are implemented, and the technologies initially implemented may differ from those ultimately selected for long-term deployment. In any scenario, domestically based, emission-free energy sources will be high priority candidates for further development.

Among these primary energy sources, nuclear energy offers great potential for the large-scale production of hydrogen that is virtually emission-free and generated from domestic resources. The production of hydrogen represents a new mission for nuclear energy that is potentially larger than the current mission of emission-free electrical production. To accomplish this goal:

- Efficient, large-scale production methods suitable for use with advanced nuclear reactors must be demonstrated. The most promising production methods are in the early stages of development. The NHI will develop and demonstrate these hydrogen production methods.

- Very high temperatures or high-efficiency electricity is required to drive the most promising hydrogen production processes. Advanced nuclear systems must be developed that provide the necessary high heat to enable these processes. The DOE Generation IV Nuclear Systems Initiative (Generation IV) is developing options to address this need with international collaboration. The focus of this effort is the Very High Temperature Reactor (VHTR), which would provide the advanced nuclear heat source for demonstrating nuclear hydrogen and electricity production.

The DOE Office of Nuclear Energy, Science, and Technology (NE) is considering a major demonstration project, which would demonstrate the commercial potential of hydrogen production from nuclear energy at a 50 megawatt thermal (MWth) scale by 2017 and provide a basis for industry investment decisions. The Next Generation Nuclear Plant (NGNP) project would develop and demonstrate the VHTR with the most promising hydrogen production processes developed by the Nuclear Hydrogen Initiative.

For hydrogen production from nuclear energy to be sustainable, the technology and policy questions that have limited nuclear energy's contribution to the energy supply must also be addressed. Advanced reactor systems that are passively safe, more efficient, and demonstrably cost effective are essential to the wider public acceptance of the nuclear role. The advantage of more efficient fuel cycles that minimize waste volumes, toxicity, and proliferation concerns must be considered. The DOE Advanced Fuel Cycle Initiative (AFCI) has initiated development of advanced fuel, separations, and transmutation technologies to address these issues.

The relationship of these nuclear energy programs to the overall DOE Hydrogen Program is illustrated in Figure 1-4. The relative sizes and positions represented in the diagram are not intended to signify program importance. The NHI and Generation IV programs together will provide the technologies needed for emission-free transportation fuels from nuclear energy.

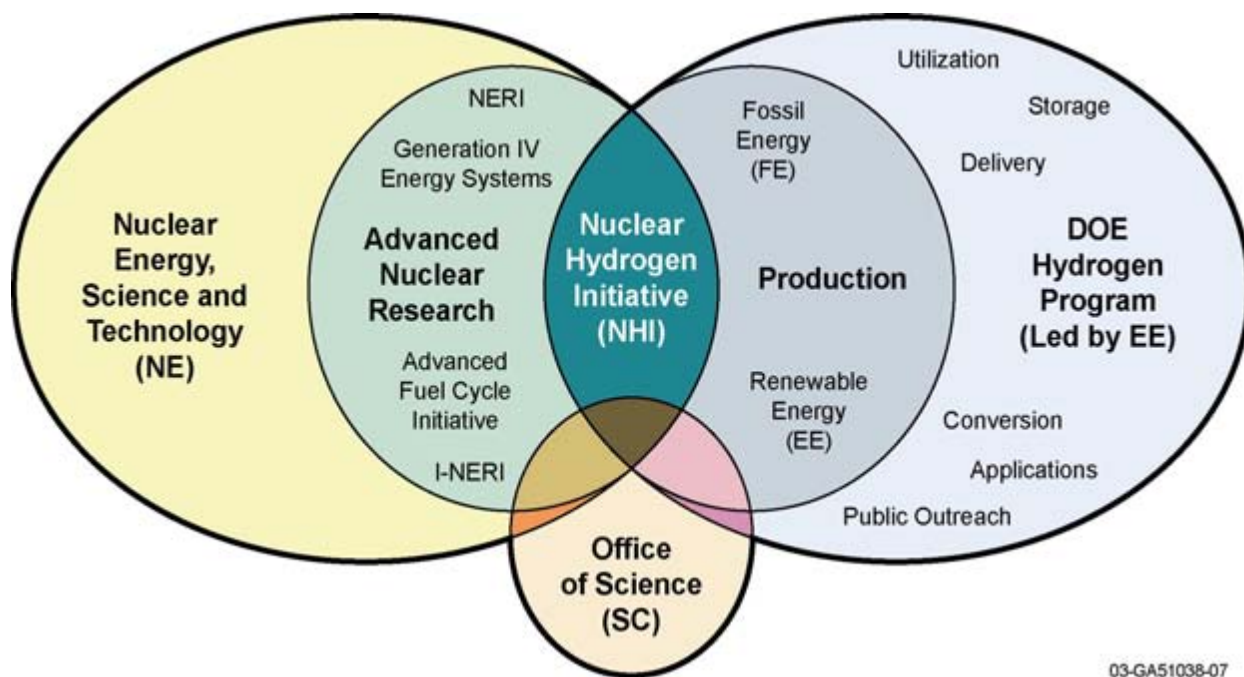


Figure 1-4. The Nuclear Hydrogen Initiative is part of an integrated DOE program to develop technologies for nuclear hydrogen production.

1.3 Nuclear Hydrogen Initiative

The NHI will focus on hydrogen production technologies best suited for use with advanced nuclear systems. Although there are already significant quantities of hydrogen produced in the United States, it is primarily produced by steam reforming of natural gas, which is already a high-quality fuel (see Appendix 1-1). Reforming one high-quality fuel to another is economically justified because of the value of hydrogen to the petrochemical industry for use in refining lower-grade crude oil to produce gasoline, and to the agricultural industry for use in fertilizer production. The current production level in the United States would be equivalent to about 100 GWth of nuclear or fossil power, assuming 50% efficiency for hydrogen production. Other fossil fuel production options based on coal, such as coal gasification, are also being developed. These production methods and the necessary carbon sequestration technologies are being pursued by FE.

Nuclear energy can be used to provide thermal energy for steam reformation, replacing approximately 25 to 30% of the natural gas requirement (or electricity equivalent for conventional electrolysis). The application of nuclear energy to these existing

hydrogen production technologies needs to be understood as a baseline for nuclear hydrogen economics. Since these technologies are currently in commercial use, they do not require significant process R&D to assess the nuclear hydrogen potential.

When nuclear energy replaces fossil fuel in the reforming or gasification process, the environmental benefit of nuclear energy is significantly reduced since carbon emissions are still generated. As such, the NHI will only address these conventional production options in system studies and economic assessments for comparison with advanced technologies.

The most attractive hydrogen production options for nuclear energy are those that utilize high temperatures or efficient electricity from a VHTR to produce hydrogen from non-fossil resources (i.e., water) and will, therefore, be the focus of the NHI. These NHI production processes will have materials and systems technology issues in common with the hydrogen production research being performed by EE, FE, and SC, and will be closely coordinated through the DOE Hydrogen Program. The NHI will collaborate, augment, and complement ongoing DOE research efforts, where appropriate, or initiate needed R&D in nuclear-specific areas to accomplish NHI program goals.

The NHI research effort will focus on:

- *Nuclear Hydrogen Production Process R&D* for production processes that most efficiently couple to advanced high-temperature reactors to produce cost effective hydrogen from nuclear energy. These processes include a range of thermochemical cycles and high-temperature electrolysis (HTE).
- *Nuclear Hydrogen Systems Interface R&D* to address the essential high-temperature thermal transport, heat exchanger, and materials issues to couple the high-temperature reactor to the process plant.

The NHI will also require independent analysis capabilities to support the assessment and evaluation of technology options and scaling decisions. Effective interfaces with other U.S. and international hydrogen research must also be established to ensure effective collaboration and integration. These NHI program functions include the following:

- *Nuclear Hydrogen System Studies* for independent analyses to support comparative technology assessments, and more comprehensive systems analyses to provide programmatic perspective on technology choices and implications. Independent analyses will provide a consistent basis for

comparison of cost and performance for alternative processes. More comprehensive systems analyses that examine the implications (e.g., economic, environmental, social/political) of nuclear hydrogen technology choices and implementation strategies will provide the necessary perspective to inform technology and policy decisions.

- *NHI Program Interfaces* to support program direction and maintain essential interfaces with related hydrogen programs. These include the necessary international and industry interfaces to conduct the most cost-effective research. Collaboration with countries performing related research (e.g., Japan, France, European Community) will expand available resources and improve the potential for success. An early, effective interface with both the chemical and nuclear industries will be essential to assessing the technical and economic viability of nuclear hydrogen options since both process development and implementation stages should have strong chemical and nuclear industry components.

These major NHI elements are summarized in Figure 1-5.

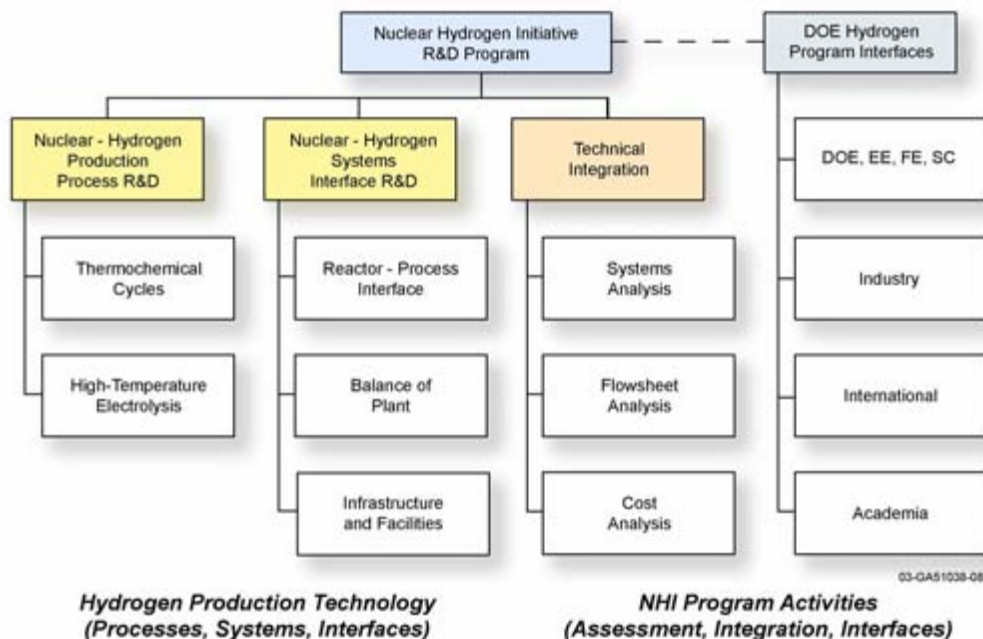


Figure 1-5. Nuclear Hydrogen Initiative program and technology development elements.

2. NUCLEAR HYDROGEN DEVELOPMENT STRATEGY

The first step in developing a nuclear hydrogen capability is the development of a coordinated research plan to define the necessary R&D and evaluation criteria to guide technology decisions. This *Nuclear Hydrogen R&D Plan* constitutes that first step and defines the path to develop the technologies necessary to demonstrate hydrogen production using nuclear energy.

2.1 Objective

The purpose of this R&D plan is to identify the candidate advanced hydrogen production technologies most suitable for nuclear energy, assess their viability, and prioritize the needed R&D to enable the demonstration of nuclear hydrogen production. This plan outlines the process and systems interface information that must be available to guide NHI R&D investment decisions.

2.2 Approach

The NHI will focus on the non-fossil-based production processes that couple efficiently to high-temperature nuclear reactors. Most of these production methods split water molecules using thermal or electrical energy. This decomposition of water takes significant energy; assuming no ohmic losses, about 123 megajoules (MJ) is required to produce one kilogram (kg) of hydrogen. (The energy content of 1 kg of hydrogen is approximately equal to one gallon of gasoline). To accomplish this with heat (thermolysis) alone requires extreme temperatures of 2500°C or more. Furthermore, current technology to produce hydrogen using radiolysis (the chemical decomposition of water by the action of radiation) does not meet minimum efficiency requirements for large-scale applications.

Although research is being conducted on techniques to reduce the temperature required for thermolysis and other possible production approaches, the most promising methods using nuclear energy are based on electrolytic or thermochemical processes. To identify the most promising hydrogen production options, available information on thermochemical cycles, high-temperature electrolysis, and other possible production methods was reviewed, and limited confirmatory analyses were performed. Processes

were evaluated using general evaluation criteria based on performance potential and demonstrated technical viability. These criteria are also part of the ongoing systematic reevaluation of process potential based on R&D results.

2.2.1 Thermochemical Cycles

Thermochemical cycles produce hydrogen through a series of chemical reactions where the net result is the production of hydrogen and oxygen from water at much lower temperatures than direct thermal decomposition. Energy is supplied as heat in the temperature range necessary to drive the endothermic reactions, generally 750 to 1000°C or higher. All process chemicals in the system are fully recycled. Thermochemical cycles were widely investigated from the late 1960's through the mid-1980s. The advantages of thermochemical cycles are generally considered to be high projected efficiencies, on the order of 50% or more, and attractive scaling characteristics for large-scale applications. However, of the more than 200 cycles that have been identified in the literature, many have been found to be unworkable, have low efficiency, or require excessive temperatures.

Thermochemical cycle technology is at a relatively early stage, and only a few cycles have been demonstrated at the laboratory-scale. Although there is greater uncertainty in the outcome of R&D, there is also potential for significant process improvement based on more recent advances in materials and chemical technology over the past two decades. Based on the information that is available, (i.e., performance potential and demonstrated technical viability) promising cycles have been identified for further investigation. Several "families" of thermochemical cycles emerged as the most promising options. The various cycles within these families were further evaluated and prioritized by factors such as theoretically achievable efficiencies, technical risk, and technical maturity. Key R&D needs for these families of cycles were developed, along with preliminary recommendations regarding decision points in cycle development.

Thermochemical cycles described in the literature indicate potential for higher-efficiency, lower-

temperature operation or other attractive cycle characteristics but are not sufficiently developed to make a clear determination. These high-risk, but potentially high-payoff, cycles are listed as alternative cycles and should be analyzed further at a lower priority. Several cycles were identified, and others may be added as more detailed evaluations are completed.

2.2.2 High-temperature Electrolysis

Electrolysis is the most straightforward approach currently available to produce hydrogen directly from water. Conventional electrolyzers are available with electric to hydrogen conversion efficiencies of 70% at a cost of about \$400/kWe installed. This gives an overall hydrogen production efficiency of 23 to 28% if electricity generation is 33 to 40% efficient. High-temperature electrolysis (HTE), or steam electrolysis, has the potential for higher efficiency. Thermal energy is used to produce high-temperature steam, which results in a reduction of the electrical energy required for electrolysis.

Electrolysis requires low-cost, efficient electricity and an energy source that provides the highest possible temperatures consistent with materials capabilities. The temperature range currently being considered is nominally up to 950°C. HTE has the potential for higher efficiency than conventional electrolysis and can be accomplished using similar materials and technology to those used in solid-oxide fuel cells (SOFC). Specific technologies used in the HTE process are being developed by other DOE offices. Therefore, the assessment of the nuclear application will integrate these results and focus on developing a conceptual design for an HTE system coupled to an advanced high-temperature reactor.

Large-scale applications would be composed of many smaller electrolyzer modules. The cost effectiveness of scaling the modular electrolysis process as compared with the scaling of thermochemical methods needs to be evaluated.

2.2.3 Other Production Methods

Additional alternative technologies or enhancements were also considered. Some of these potentially promising concepts (including thermally-assisted membrane hydrogen production and direct-contact methods) were not considered consistent with the

current demonstration schedule. Process improvements for commercial technologies, such as conventional electrolysis or steam reforming of methane process improvements, are supported by other elements of DOE and are not addressed as part of the NHI (see Appendix 1-1). Finally, while the transportation, storage, and use of hydrogen are necessary components of a hydrogen energy system, these technologies are being developed by EE and are not addressed in this R&D plan.

2.2.4 Systems Interface

Both thermochemical and HTE processes introduce new considerations into the design, licensing, and operation of combined nuclear-chemical plants that must also be considered in the technology selection process. These balance-of-plant and systems interface issues are identified as important components in the R&D program for nuclear hydrogen. R&D needs that address the essential high-temperature thermal transport, heat exchanger, and materials issues to couple the high-temperature reactor to the process plant were identified. Many supporting systems will be common to both the nuclear and hydrogen plants, while the high-temperature heat exchanger and the materials associated with thermal transfer will be specific to each production process.

2.3 Technology Assessment

Since all nuclear hydrogen production approaches being considered in the NHI avoid the production of greenhouse gases and can be based primarily on domestic resources, the primary issue for nuclear hydrogen is the development of cost-effective systems that produce hydrogen at a cost that is competitive with gasoline. Projecting costs for technologies at this early stage of development is highly uncertain for any of the approaches being considered. The criteria to be used to evaluate the benefits of the various hydrogen production methods in relation to these cost objectives include the system and performance characteristics that drive costs, and the uncertainty of the projected costs.

2.3.1 General Evaluation Criteria

General evaluation criteria were based on process performance potential and development maturity. Laboratory demonstration of a process may not ensure

cost effectiveness; however, the performance and cost uncertainties can be reduced. None of the processes reviewed have been demonstrated on a scale that would be necessary for reliable cost estimates.

- Costs** – Capital cost recovery is considered to be the dominant component of nuclear hydrogen costs. Although early studies suggest that the capital cost of the hydrogen production process will be less than the nuclear heat source, the projected capital cost of the process plant is a key selection criterion. The most direct indicator of process costs is process efficiency since higher efficiency processes leverage both hydrogen and nuclear plant capital costs. Hydrogen production efficiency is defined as the lower heating value of hydrogen (~123 MJ/kg) divided by the total energy required to produce the hydrogen, including net reaction energies and energy loss mechanisms. Processes with potential for efficiencies in the range of 50% were considered promising. Other factors considered as cost indicators were process complexity (number of reactions, separations), materials (high-temperature compatibility requirements, corrosion, toxicity), level of industrial safety concern, and operational modes and flexibility.
- Uncertainty/Risk** – Assessing the probability that a particular hydrogen production option will meet expectations for performance and cost involves significant uncertainty at this stage of development. Processes demonstrated at a laboratory scale allow more reliable estimates of performance and cost. Promising processes that have not been demonstrated may still deserve further evaluation, but not as a near-term priority until additional confirmatory analyses are available. To be considered as a baseline process, all individual chemical reactions or physical process steps should have been demonstrated in laboratory experiments. An integrated process demonstration is necessary before a process can be selected for a pilot plant demonstration. Demonstrations at the pilot plant or engineering-scale have not been completed for any process. Unique industrial

safety concerns pose a different type of developmental risk that must also be included in future implementation decisions.

2.3.2 Production Process Prioritization

Performance and viability information on candidate hydrogen production processes will be needed in 2009 to provide the basis for pilot plant decisions. Integrated demonstrations of those cycles under consideration must be sufficiently complete by that time to assess performance and cost implications. To ensure that the necessary information is available for critical scale-up decisions, it is essential to establish a prioritization process that ensures that information on the high-priority processes is available for this decision, while not overlooking other potentially higher performance alternatives. The NHI will pursue a two-tier approach as described below.

Processes identified as having the highest probability of achieving program goals in the planned schedule and budget will have the highest priority. The goal of the R&D for these baseline processes will be to complete an integrated, laboratory-scale experiment by 2009 to provide a basis for pilot plant process selection. Initial process priorities will be systematically reevaluated as R&D progresses. At the same time, however, it is recognized that alternative processes, those posing higher risk but with potentially higher gain, should also be evaluated further to ensure that potentially important options are not overlooked. These analyses will be lower priority but in parallel with baseline process development. This demonstration strategy is designed to ensure that a 2017 engineering-scale demonstration is fully supported while minimizing risk and allowing flexibility by exploring potentially more efficient, yet currently immature, processes.

2.3.2.1 Baseline Processes. Two families of thermochemical processes were identified as baseline methods. These processes have potential for high efficiencies (~50%), have process steps that have been demonstrated to show feasibility, and can be developed by 2017. High-temperature electrolysis was also identified as a baseline process. Cost uncertainties remain, but preliminary projections were available to support the potential for making hydrogen at a cost competitive with gasoline.

- The first priority baseline thermochemical cycles are the sulfur-based family of cycles. The sulfur-iodine, sulfur-bromine hybrid, and hybrid sulfur cycles are sulfur-based variations that have demonstrated high performance and are the focus of ongoing research in the United States, Japan, and France. The sulfur-based cycles are first priority because they are projected to have high efficiencies at NGNP temperatures, are the most developed and supported, and much of the R&D is potentially applicable to multiple cycle variations, thus reducing overall risk.
- The second priority thermochemical baseline cycles are the calcium-bromine family. The calcium-bromine cycle (UT-3 and Star) involves lower peak temperatures and solid-gas reactions, and all process steps have been demonstrated. The calcium-bromine family is identified as second priority because it is projected to have a lower overall efficiency potential than the sulfur cycles and does not have significant ongoing research support at this time.
- High-temperature electrolysis, based on SOFC technology, is also a baseline process. HTE is also projected to reach 50% overall efficiencies at NGNP temperatures, and all physical processes have been demonstrated. Much of the basic technology work is supported by other areas of DOE. NHI research will focus on system design and cost assessment.

2.3.2.2 Alternative Processes. Several promising alternative cycles were identified (either because of greater efficiency, lower temperature requirements, or reduced complexity) as deserving further study. Sufficient information was available to identify their potential, but not to merit significant early investment. The initial list of alternative cycles will be reevaluated as analysis results become available.

2.4 Process Demonstration Strategy

Demonstrating nuclear hydrogen production by 2017 will require a systematic approach to develop the required information for each candidate process necessary to provide an adequate basis for decisions on the next stage of demonstration. Such a systematic demonstration strategy has been developed to allow the maximum amount of flexibility to reassess the promise of process technologies while still ensuring that the overall goal is met. The production process options identified in this Nuclear Hydrogen R&D Plan are currently in different stages of development. Baseline processes are closer to demonstration than the alternative processes. However, all high-priority processes will develop in a similar sequence, beginning with the demonstration of viability on a laboratory-scale. The current demonstration strategy is as follows.

- Consistent thermodynamic or systems analyses will be performed on all candidate processes to confirm performance and process viability. Once analyses have confirmed sufficient performance potential and process viability, decisions can be made as to whether laboratory-scale experiments should be performed.
- Integrated laboratory-scale demonstrations will be performed to confirm technical feasibility of the closed loop process. Laboratory-scale demonstrations of selected processes need to be complete by 2009 to support pilot plant decisions. Pilot plant conceptual designs and system interface technology requirements will also be considerations in process selection. Although work on candidate processes must be substantially complete by 2009 to support pilot plant decisions, process improvement R&D may proceed in parallel with pilot plant construction. Processes that have not been demonstrated by 2009 may also be tested at a pilot plant-scale later if there is sufficient NHI program interest.

- Processes successfully demonstrated in integrated laboratory-scale tests can then be considered for demonstration at a pilot plant-scale to confirm engineering approaches and performance. Pilot plants on the order of 1 MW would be prototypic with regard to process technology and operating conditions, but would not require nuclear heating. Design and construction of the initial pilot plant configuration is planned to begin in 2009. High-temperature heat exchanger and materials R&D must be sufficiently advanced to demonstrate viable engineering approaches for the reactor-hydrogen interface. Pilot plant operations for one or more of the processes provide the engineering and performance basis to select the production process for the NGNP demonstration. Results from operations are needed by the end of 2011 to impact final design of the NGNP engineering

demonstration. It is planned that the pilot plant-scale experiments will be nominally one tenth the size of the engineering-scale demonstration.

- The results of pilot plant operations provide the engineering and performance basis for selecting production process(es) for the NGNP engineering-scale demonstration. An engineering demonstration, nominally 50 MW, will be designed, constructed, and operated to confirm economics and commercial feasibility of the selected production process(s).

The sequence and schedule are summarized in Figure 2-1 for the major R&D areas that are identified in the R&D plan leading to a nuclear-heated engineering demonstration in 2017.

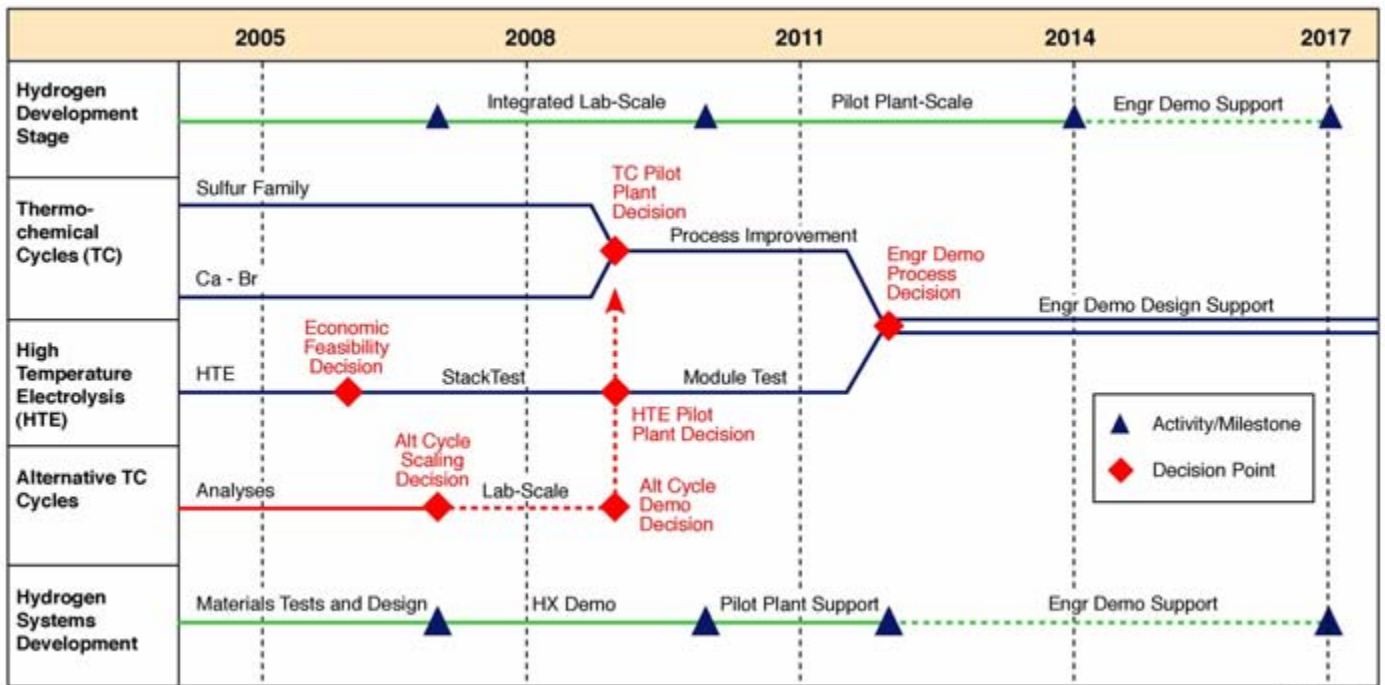


Figure 2-1. Summary of Nuclear Hydrogen R&D Plan activities and sequence.

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3. THERMOCHEMICAL CYCLES

Thermochemical cycles produce hydrogen by a series of chemical reactions where the net result is the decomposition or splitting of water, $\text{H}_2\text{O} \rightarrow \text{H}_2 + \frac{1}{2}\text{O}_2$, at much lower temperatures than can be done with heat alone (thermolysis). A pure thermochemical cycle involves a series of chemical reactions that are driven only by thermal energy. The net result of the input of heat and water is the production of hydrogen and oxygen. All other process chemicals in these systems are fully recycled. Hybrid thermochemical cycles include both chemical reaction steps and an electrolysis step of some chemical compound (not water) that usually produces hydrogen. Both thermal and electrical energy are required to complete the hybrid cycle. However, the energy requirements for the electrolysis step are much less than the requirements for electrolysis of water. The temperatures required to drive the endothermic reactions for most thermochemical cycles are challenging – generally from 750 to 1000°C or higher.

Thermochemical cycles are considered promising options for hydrogen production because of the potential for high efficiencies and scaling to large capacities. Thermochemical cycles are generally considered to have potential for lower costs than conventional electrolysis of water because the production of hydrogen by electrolysis requires conversion of heat to electricity prior to hydrogen production, whereas thermochemical cycles produce hydrogen directly from thermal energy. Chemical plant economics-of-scale (such as thermochemical processes) have historically been favorable compared to the more modular electrolysis processes. Finally, the status of thermochemical cycle technology is relatively immature (see Appendix 3-1), but there is a greater potential for improvement compared with conventional electrolysis. High-temperature (or steam) electrolysis is a variation of conventional electrolysis that has potential for increased efficiency (see Section 4).

3.1 Objective

The objective of the thermochemical cycle research effort is to develop and demonstrate thermochemical processes to produce hydrogen cost effectively using nuclear energy. Based on analyses and laboratory-scale experiments, research will develop the necessary

information on cycle performance and cost needed to support the selection in 2009 of the production process for a pilot plant. The target costs for hydrogen production are \$2.50/kg in 2011, and \$2.00/kg in 2017. These projected hydrogen cost targets assume that pilot plant operations in 2011 and the engineering-scale demonstration in 2017 provide performance and cost information that allow projection to commercial-scale production with these costs.

3.2 Approach

Performance and technical viability information on candidate processes will be needed in 2009 to provide the basis for pilot plant decisions. R&D activities to support this scaling decision will focus on laboratory demonstration of the most promising baseline cycles identified as having the highest probability of achieving program goals. These integrated baseline systems must be substantially demonstrated by 2009.

The NHI research program will also address alternative thermochemical processes that have been identified as having potential for high performance but are considered to have high technical risk due to lack of sufficient analyses and development. These cycles will be evaluated further to ensure that potentially important options are not overlooked. These analyses will proceed in parallel with baseline process development. This demonstration strategy is designed to ensure that the 2017 NGNP demonstration plant is fully supported, while minimizing risk and allowing the flexibility to explore potentially more efficient, yet currently immature, processes.

3.3 Thermochemical Cycle Assessment

The energy crisis of the 1970s initiated a major effort to develop thermochemical processes in Europe and the United States. Except for a limited amount of work in Japan, this work ended by the early 1980s. More than 200 cycles have been identified in the literature. The most comprehensive sources of information were the extensive review studies that have been published, which in some cases included evaluation and selection criteria.

3.3.1 Summary of Previous Studies

The first major program was at the European Community Joint Research Center (ISPRA), beginning in the late 1960s and continuing through 1983. The goal of this work was to identify thermochemical cycles to couple to the high-temperature, gas-cooled reactor. The three-phase program investigated 24 cycles. In Phase I, thermochemical cycles were developed based on the chemistries of mercury, manganese, and vanadium. In Phase II, nine cycles based on iron chloride chemistry were investigated. The iron chloride cycles were abandoned because of the difficulties with the thermal decomposition of ferric chloride. Later, the Gas Research Institute identified a proprietary procedure that may have resolved these issues. Phase III focused on multiple sulfur-based cycles with a laboratory demonstration of the sulfur-bromine hybrid process. Associated with these laboratory efforts were parallel activities involving corrosion testing, design of larger-scale equipment, and development of industrial flowsheets. The duration (1.5 yrs) of the sulfur-bromine hybrid laboratory test is the most extensive demonstration of any thermochemical cycle to date.

In the United States, the Gas Research Institute (now known as the Gas Technology Institute) funded a long-term program that systematically examined thermochemical cycles. Over the nine-year program, 200 distinct thermochemical cycles were examined, about 125 cycles were considered feasible based on thermodynamic considerations, 80 of the most promising cycles were tested in the laboratory, 15 of these cycles were found to be operable using batch techniques with reagent-grade chemicals, and eight cycles were operated successfully with recycled materials to achieve proof-of-principle. This work is the single most comprehensive source of information on thermochemical cycles. The heat source for these cycles was to be either a nuclear reactor or a solar system, and cycles covering a wide range of temperatures were considered.

Several conclusions were drawn from the Gas Research Institute work. The most important conclusion was that experimental verification is required to determine if a thermochemical cycle is viable. Analyses alone are not sufficient. Most proposed cycles were eliminated in the laboratory

because the chemical reactions were too slow, unwanted chemical reaction products were produced, or no efficient methods were found to separate chemical reaction products. New developments in catalysts or separations techniques could make some of these processes viable. The second conclusion was that there are very large differences between theoretical efficiencies and efficiencies based on initial process flow sheets; thus, processes that have high theoretical efficiencies may not work in practice. The conclusion of this work was that relatively few of the cycles investigated were promising for further development. The three that were most highly ranked were hybrid sulfur, sulfur iodine, and hybrid copper sulfate.

The largest single-process development effort was conducted by Westinghouse Corporation to develop the hybrid sulfur process. This effort progressed through a laboratory demonstration with the final product being a conceptual design report for a pilot plant. Two conclusions are derived from this work: first, the hybrid sulfur process with 1970s technologies could be made to work and second, there were many options to improve efficiency, given sufficient R&D resources. Process efficiencies above 40% were calculated for these hybrid sulfur experiments; however, potential improvements were also identified that could significantly increase efficiency. With the decrease in energy prices in the late 1970s, all of these efforts (except for a small effort in Japan) were abandoned.

More recently (1999), based on the renewed interest in hydrogen production, General Atomics, Sandia National Laboratories, and the University of Kentucky jointly conducted a literature evaluation of thermochemical processes. The Nuclear Energy Research Initiative (NERI) study reviewed available information for 115 cycles, which were ranked by complexity (reactions, separations, elements, and corrosiveness), development maturity (demonstration level and publications), and performance (efficiency and cost). The four leading processes were hybrid sulfur, sulfur-bromide hybrid, UT-3 (calcium bromine), and sulfur iodine. The new process was the UT-3 process developed by the University of Tokyo since the 1970s. The work on this new cycle was initiated to provide a lower-temperature process that would be compatible with lower-temperature heat sources.

3.3.2 Conclusions from Previous Studies

The general conclusion of these studies is that there are currently only a small number of processes that can currently be considered as candidates for large-scale development. Given the scope of research in the 1970s, it is judged relatively unlikely that a significant number of new cycles with more promising chemistry will be identified in the near term. It is recognized that new technologies (catalysts and separations techniques) may make previously unattractive thermochemical cycles viable. Although many cycles should be reevaluated in light of these new technologies, they cannot yet be considered candidates for near-term development. Therefore, the cycles that were examined in the previous efforts and that survived the laboratory confirmation process formed the starting point for prioritizing these cycles for the NHI. Collectively, these studies identified several cycles, or groups of cycles, that had promising performance and were confirmed as viable after laboratory experiments. Relatively few of the more than 200 cycles met these basic requirements.

The sulfur-based cycles were commonly identified in all studies: sulfur-iodine, hybrid sulfur, and sulfur-bromine hybrid. These cycles were demonstrated to have high efficiencies and were among the least complex. They have also been extensively demonstrated at a laboratory-scale to confirm performance characteristics. A calcium-bromine (Ca-Br) cycle (UT-3), developed later in Japan, has also been demonstrated and appears as a promising cycle. Researchers demonstrated high efficiencies at lower temperatures than the sulfur-based cycles. The gas-solid reactions involved in this cycle present both substantial technical challenges and some potential advantages. Recent Argonne National Laboratory (ANL) work has proposed an alternative version of this cycle.

These studies also identified many other cycles that had potentially promising features (higher efficiency, lower temperature, less complex configurations) that either had not been or could not be demonstrated as workable at the time. In most cases, there is a process step that makes the process noncompetitive with the leading sulfur processes. Examples of these potentially promising but high-risk cycles include: the hybrid copper sulfate cycles and the iron-based and copper-based cycles (with lower temperature and less toxic materials).

3.4 Thermochemical Cycle R&D Needs

The NHI R&D Plan assessment examined this distilled group of candidate cycles in light of the general evaluation criteria for NHI described in Section 2 to define initial priorities for NHI research. These general evaluation criteria focused on the projected cost of hydrogen produced by that cycle and the technical and development risk (i.e., Can the cycle be made to work and meet performance expectations in the NHI time frame?). For the thermochemical cycles, the most useful indicators of hydrogen production cost are the cycle efficiency and complexity (number of reactions and separations). The indicator of acceptable technical risk was that all cycle reactions and physical processes have been demonstrated in the laboratory. Cost and technical risk perception is also influenced by other factors (e.g., excessive temperature or pressure requirements, highly toxic or corrosive materials). The potential for performance improvement, although not easily quantifiable, was also considered as a factor in prioritizing these cycles for NHI. In cycles where performance improvements had been suggested based on new technologies (i.e., membranes and catalysts; see Appendices 3-2 and 3-3), the return on research investment could be increased, such as in the recent sulfur cycle membrane and catalyst proposals. This trend should increase as visibility and interest in hydrogen production increases.

Based on the preliminary list of cycles derived from the literature on thermochemical cycles and on the NHI general evaluation criteria, a short list of cycles was identified that met the NHI efficiency and cycle characteristics criteria and had been experimentally demonstrated in the laboratory. These cycles are listed in Table 3-1 and grouped in order of priority for the NHI (see Appendix 3-1). Of the laboratory-demonstrated processes, four (sulfur-iodine, hybrid sulfur, sulfur-bromine hybrid, and calcium-bromine cycles) have been tested in small, laboratory-scale, integrated experiments with fully developed flowsheets. The other cycles are promising but at an earlier state of development with significant uncertainties associated with the flowsheets and estimated efficiencies. Definitive calculations of efficiency exist only for the four demonstrated baseline processes. Confirmatory analyses for the two leading sulfur processes, hybrid sulfur and sulfur-

iodine, were performed to confirm earlier results using modern flowsheet analysis methods.

The sulfur-based cycles (sulfur-hybrid, sulfur-iodine, and sulfur-bromine hybrid) are the highest priority candidates for NHI. These cycles have the highest calculated efficiencies (>40%) of demonstrated cycles, and improvements have been proposed that could result in significant improvements in cycle efficiencies, projected to be as high as 60%. The hybrid sulfur cycle is among the least complex process of any thermochemical cycle. The high-temperature sulfuric acid decomposition reaction (see Section 3.4.1) and the associated materials requirements are common to all cycles in the family. Thermochemical research programs worldwide have increasingly emphasized sulfur cycles as work has progressed, and R&D on these cycles is ongoing in the United States, Japan, and France.

Calcium-bromine cycles also have potential for high performance, with lower temperature requirements than the sulfur-based cycles, and the cycle configuration has potential for further simplification. High performance has been demonstrated in a continuous operation, and there is recent R&D interest in Japan and the United States. It is considered a lower priority due to lower potential efficiency and issues with integrity of the solid-gas reaction beds.

Other cycles were identified that should be pursued due to the promise of higher efficiency, lower temperatures, or better cycle characteristics. Examples of these cycles are listed in Table 3-1 as alternative cycles that should be analyzed further before significant work is implemented. As interest and activity in thermochemical research increases, other cycles could be added to these examples.

Table 3.1. Prioritized thermochemical cycles for hydrogen production.

	Peak Temp (°C)	Number of Reactions	Reported Flowsheet Efficiency %*	Cycle Status	Advantages	Key Issues
Sulfur Cycles						
Sulfur-iodine	827-900	4	42-51	Continuous Demo 48 hr	Projected >60% efficiency All thermal (no electrolysis step) International R&D ongoing	Hydrogen-iodine distillation High temp materials Iodine hazard and cost
Hybrid sulfur	Not Given	3	41-53 Depends on cell voltage	Continuous Demo 200 hr Conceptual Plant Design	Projected >60% efficiency No halogens Least complex sulfur cycle	Economic scaling of hybrid processes High temp materials Higher efficiency electrolysis
Sulfur-bromine hybrid	Not Given	4	39	Continuous Demo 1.5 y	Longest duration demonstration No major instabilities	Economic scaling of hybrid processes High temp materials Higher efficiency electrolysis Bromine hazard and cost
Ca-Br Cycles						
UT-3	750	4	40-50	Continuous Demo 10 hr	Lower maximum temperature	Durability of the solid chemicals through multiple cycles Gas-solid reactions
Ca-Br-Star (ANL)	750	3	44		Variant of UT-3 with fewer steps	Not fully demonstrated
Promising Alternatives						
Copper-chlorine hybrid	550	4	46 (Est.)	Rxn Demo	Low peak temperature	Higher efficiency electrolysis
Iron-chlorine	650	3	47-49 (Est.)	Rxn Demo	Low peak temperature	Need to suppress competing chemical reactions ⁸
Copper-sulfur hybrid	827	5	68-73 (Est.)	Rxn Demo	Potential for high efficiency	Economics of scaling hybrid processes Higher efficiency electrolysis
Vanadium-chlorine	925	5	40.5-42.5	Rxn Demo	Potential for improvement (O ₂ membrane)	Very-high temperature O ₂ membrane Conflicting data on one reaction

* Reported efficiencies and peak temperatures have been obtained from literature from the 1970s to 2003 using a range of assumptions and methods. See Appendix 3-1 (table and accompanying references) for details on the efficiency calculations. Cycles defined as promising alternatives are those that were not sufficiently developed in the past but may merit further investigation based on estimated performance and improvements from incorporation of that new technologies may that may significantly improve performance.

3.4.1 Sulfur-Based Cycles

The sulfur-based thermochemical cycles have been sufficiently developed to extrapolate with reasonable confidence that, given sufficient resources, the processes would be technically viable. No other cycle or group of cycles was identified that had the combination of performance potential, technical maturity, and demonstrated viability. These cycles have the highest probability of success for NHI and will be the focus of the NHI thermochemical R&D program. An integrated program will be pursued that explicitly recognizes common R&D areas. Similarly, pilot plant facilities will be designed to test the family of cycles. A schematic representation of the sulfur

family of cycles is shown in Figure 3-2. Note that the common high-temperature reaction shown on the left can be used with any of the three lower temperature reactions shown on the right.

The decomposition of sulfuric acid is common to all of these sulfur-based cycles. The sulfur-iodine and sulfur-bromine hybrid cycles involve primary reactions that produce hydrogen-iodine (HI) or hydrogen-bromine (HBr) in solution, which must be separated and decomposed to produce hydrogen. Hydrogen is produced in the sulfur-iodine cycle by thermal decomposition of hydrogen-iodine, while the hybrid sulfur and sulfur-bromine hybrid cycles produce hydrogen in an electrolytic step.

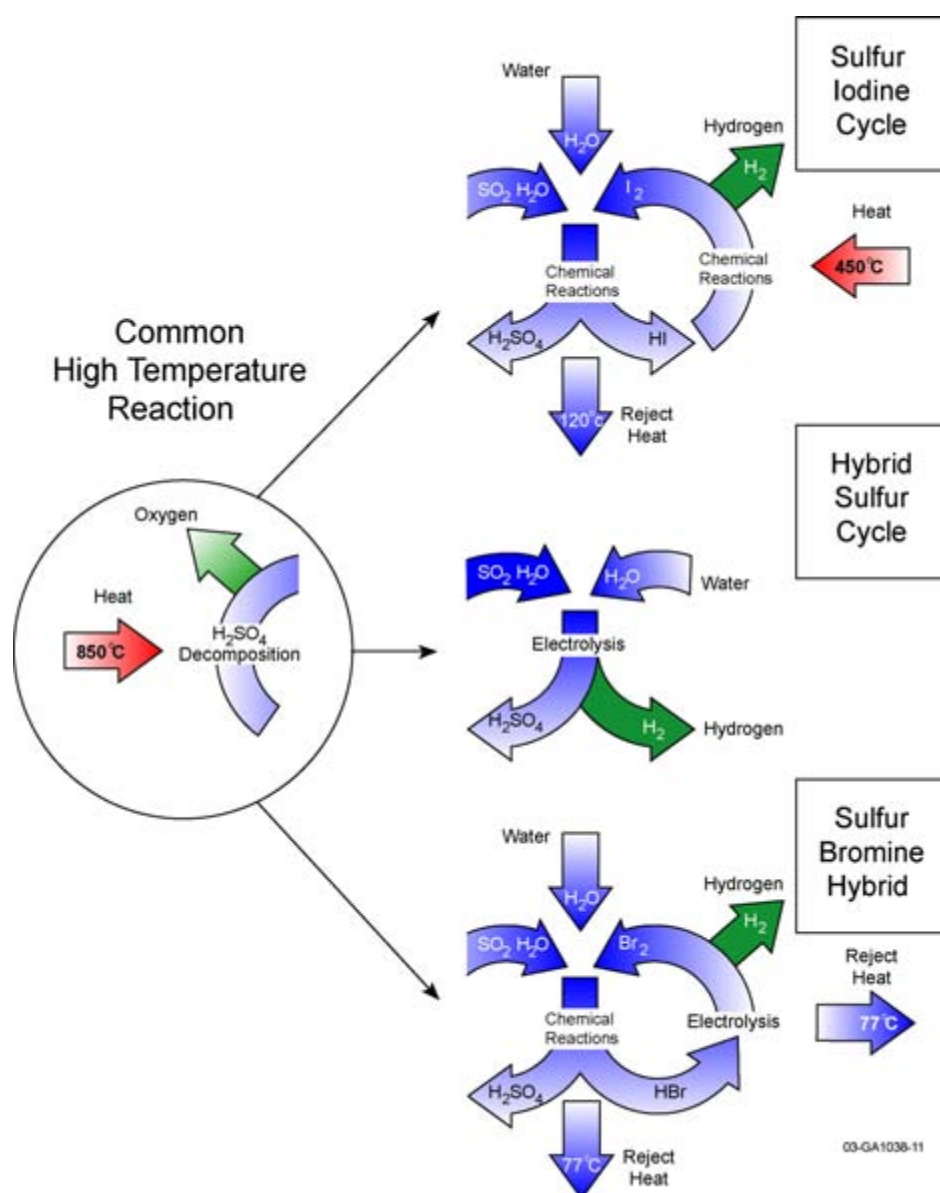


Figure 3-2. Sulfur family of thermochemical cycles.

3.4.1.1 Common R&D Areas. The sulfur-based thermochemical processes have a common oxygen-generating, high-temperature step (the dissociation of sulfur trioxide into sulfur dioxide and oxygen at temperatures in the 850 to 1000°C range). Therefore, the high-temperature materials and heat exchanger (HX) designs required to interface the sulfur cycles with the high-temperature reactor is common to all sulfur family cycles. The materials issues are challenging and different in the boiling and decomposition regimes. R&D requirements for this stage include long-term materials tests and innovative heat exchanger design studies, which are described in Section 5. The incorporation of a high-temperature (inorganic) membrane in this common stage of the sulfur cycles has the potential to increase efficiency or lower heat requirements by enhancing the decomposition reaction. This could significantly reduce the engineering challenges or improve process efficiency. R&D to evaluate the potential of high-temperature inorganic membranes should be performed early to determine the potential impact on sulfur-based cycle design.

The R&D necessary to perform an integrated laboratory-scale demonstration of sulfur-iodine and hybrid sulfur will be the highest priority for NHI. The sulfur-bromine hybrid cycle benefits from much of this research, but due to lower projected performance potential, process-specific work on sulfur-bromine hybrid will be initially limited to confirmatory analyses. The process-specific R&D areas for sulfur-iodine and hybrid sulfur are summarized as follows.

- *Sulfur Iodine* – This all-fluids-and-gases cycle involves three primary thermochemical steps. Unique technical issues associated with this specific cycle include efficient separation of hydrogen iodide, minimizing the recycle rates of chemicals within the process per unit of hydrogen produced, and reducing the inventories of iodine within the process, which, although not consumed, is expensive and toxic. Multiple alternative technical solutions (primarily using membranes) have been proposed to address these challenges. The distillation of hydrogen-iodine from solution is the most difficult process issue for this cycle. The NHI work will examine both reactive and extractive (H_3PO_4) distillation and investigate recent Japanese proposals for

the use of electrodialysis and membrane reactors as alternative technologies.

- *Hybrid Sulfur (also called Westinghouse, GA-22, and ISPRA Mark 11)* – This all-fluids cycle involves a single thermochemical step and a single electrolytic step. Westinghouse Electric Corporation demonstrated this process on a scale of 150 l/h in 1976, and a conceptual plant design has been developed. As a two-step process, it is the simplest process that has been demonstrated, involving only sulfur compounds, water, hydrogen, and oxygen. The unique R&D needs for the hybrid sulfur cycle focus on the electrolytic cell design and optimization. Reducing the electrical losses and requirements in the electrolysis step and designing improvements to allow cost effective scale up are the highest initial priorities. Anode, cathode, and membrane materials need to be reevaluated in light of the extensive R&D in those areas over the last several decades (electrical consumption of these cells is small in comparison with traditional water electrolysis).
- *Sulfur Bromine Hybrid* – This all-fluids cycle involves two thermochemical steps and one electrolysis step. It was demonstrated in a laboratory-scale experiment operated in ISPRA for 1.5 years producing 100 l/h. However, the projected efficiencies are slightly lower than the hybrid sulfur cycle. The hybrid sulfur cycle is chosen relative to this cycle because the process is more efficient, primarily because the electrolytic cell power consumption is less (0.6 vs. 0.8V), and it is a simpler process. Reduction of energy requirements for the electrolytic step is the most important R&D area. Initial work on the sulfur bromine hybrid cycle will focus on updated flowsheet analyses.

3.4.2 Calcium Bromide Cycles

The calcium-bromine cycle has been demonstrated at 1 l/h for ~100 h. The primary incentive to develop this cycle is that the peak temperature is lower than for the sulfur cycles, typically 750°C. Efficiencies have been estimated between 40 to 50%. Major work

has been done in Europe and Japan, with continuing work in Japan and in the United States under the NERI program. The key R&D areas that must be addressed for this cycle are associated with the solid-gas reactions that characterize the cycle. The CaO , CaBr_2 , and Fe_3O_4 , and FeBr_2 reactants in fixed beds or other configurations undergo volume changes in each reaction of the cycle. Research efforts to date have not been able to demonstrate the integrity of these reaction beds after many cycles. While industrial processes have been commercialized with this characteristic, such processes present a significant engineering challenge because of multiple degradation mechanisms (fines formation, slow chemical reactions, etc.).

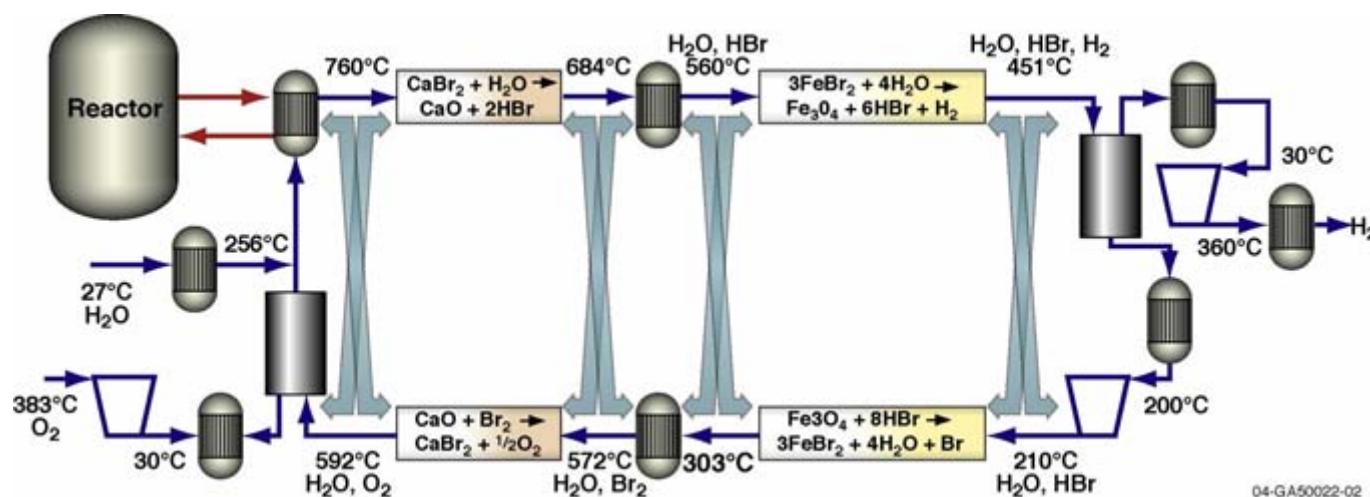
A schematic representation of the calcium-bromine UT-3 cycle is shown in Figure 3-3. The UT-3 cycles reactions occur in pairs of solid reaction beds. One pair contains calcium-bromide and calcium-oxide and the other pair contains iron-oxide and iron-bromide. The initial reaction steps form hydrogen-bromine from a high-temperature steam reaction with the calcium-bromide bed and hydrogen from the iron-bromide bed. A second (lower) series of reactions regenerates the calcium and iron-bromide reactants. When the initial reaction beds are fully converted, the flows are switched in each pair of beds and the same reactions occur in the opposite flow direction. Work at ANL has identified a variation of this cycle that eliminates the iron reaction beds and introduces an alternative electrolytic or cold plasma step for hydrogen generation from hydrogen-bromine. R&D

to develop stable reaction beds or alternative reaction stages will be the focus of the initial NHI research on this process.

3.4.3 Alternative Thermochemical Cycles

In addition to these sulfur and calcium-bromine families of baseline thermochemical cycles, several other cycles were identified as being promising due either to lower temperature requirements, high advertised efficiencies, or other positive cycle characteristics. Examples of these cycles are given in Table 3-1 (copper chloride, iron chloride, and copper-sulfur). Additional analysis of these cycles will be performed at an early stage to assess the potential of these cycles to provide a more cost effective process. Because almost three decades have passed since there has been significant work in this field, there are multiple new technologies that may enable improvements in previously considered thermochemical cycles. These cycles have not been fully demonstrated in the laboratory. In most cases, there is insufficient documentation to determine whether a cycle has potential or whether a fatal flaw in the process was discovered. The lack of a common database has resulted in significant duplication of work and has hindered progress.

Evaluation of alternative cycles will initially focus on performing consistent thermochemical flowsheet analyses to improve estimates of efficiency and complexity and provide a basis for assessing the potential for an economic cycle. If these analyses



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Figure 3-3. Schematic of Ca-Br thermochemical cycle (UT-3).

confirm cycle potential above baseline cycles, decisions will then be made whether to test key reactions in the laboratory. Laboratory results demonstrate the effects of chemical kinetics and provide data that are essential to the final assessment. The evaluation of experimental data and a preliminary flowsheet will provide the basis for a decision on further development. This evaluation and the systematic documentation of the results could be done effectively in cooperation with universities, industry, and international partners.

3.5 Thermochemical Cycle R&D Program

The R&D program for thermochemical cycles will develop the necessary information on candidate thermochemical processes to support pilot plant decisions in 2009. The decision to demonstrate a thermochemical cycle at a pilot plant-level (500 kWth to 1 MWth) requires that the candidate processes be demonstrated in integrated, laboratory-scale experiments that successfully confirmed technical viability and projected cost estimates. The objective of the NHI thermochemical research is to complete integrated, laboratory-scale demonstrations for the baseline cycles to provide the technical basis for pilot plant decisions in 2009. The research will also provide sufficient information on alternative thermochemical cycles to confirm that promising options have not been overlooked. The thermochemical R&D program priorities will focus on:

- *Integrated laboratory-scale demonstration of sulfur-iodine and hybrid sulfur baseline cycles* – Both of these cycles are sufficiently well defined to proceed to integrated testing. These demonstrations will utilize engineering materials and be performed under prototypic pressure and temperature conditions. The component reactions that must be demonstrated to support these integrated tests are:
 - *H₂SO₄ decomposition* – H₂SO₄ decomposition is common to all sulfur-based cycles. R&D will focus on high-temperature materials compatibility, heat exchanger design, and incorporation of catalysts as an

integral part of heat exchanger design.

- *Sulfur-iodine reactions* – These reactions consist primarily of prime (Bunsen) reaction and hydrogen-iodine distillation. R&D will address reduction of recycle volume in the prime reaction and determination of optimum technique for hydrogen-iodine distillation (reactive, extractive [H₃PO₄], and electrolysis options) to maximize efficiency.
- *Hybrid sulfur* – Hybrid sulfur involves an electrolytic step for hydrogen production. Research focuses on improved anode-cathode materials and fabrication methods and on improved models of cell operation (voltage, acid concentration, and temperature characteristics) to optimize the cell design. Improved membrane options will be developed based on recent membrane research results.
- Flowsheet and thermochemical data development will be performed to support the sulfur-based cycle R&D program.
- *Calcium-bromine cycle* – Flowsheet analyses and system design studies will be performed to assess performance and cost. The initial focus will be on analyses, materials, and thermodynamic data experiments. Integrated demonstration decisions will be made after key reaction bed or electrolytic issues have been addressed.
 - *R&D on calcium and iron reaction bed materials integrity, (materials and coating technology)* – R&D will focus on developing thermodynamic property and kinetics data to support flowsheet analyses.
 - *Electrolytic step (STAR variation)* – Cell design and materials assessment for the hydrogen-bromine electrolytic step will be performed. Models of

hydrogen-bromine dissociation and cell processes will also be developed to support improved performance studies.

- *Alternative thermochemical cycles* – Initial focus will be on consistent flowsheet analyses of promising cycles to confirm potential. Studies will examine membrane and catalyst options to resolve key issues in selected alternative cycles.

The proposed organization of the R&D for the thermochemical cycles is shown in Figure 3-4. These assessments will identify if calcium-bromine or an alternative cycle should be considered a higher priority in 2005. The demonstration of all component reactions in 2006 supports the start of integrated

demonstrations of the high-priority sulfur-iodine and hybrid sulfur in 2007.

Integrated demonstrations provide performance confirmation and a basis for cost projections that support pilot plant decisions in 2009. Pilot plant preliminary design activities will be performed as R&D progresses and provides a basis for comparative cost estimates for scaling between hybrid and pure thermochemical cycles. This development strategy is summarized in Figure 3-5.

3.6 Thermochemical R&D Activities

Major thermochemical R&D activities for sulfur, calcium-bromine, and alternative cycles are summarized in Table 3.2.

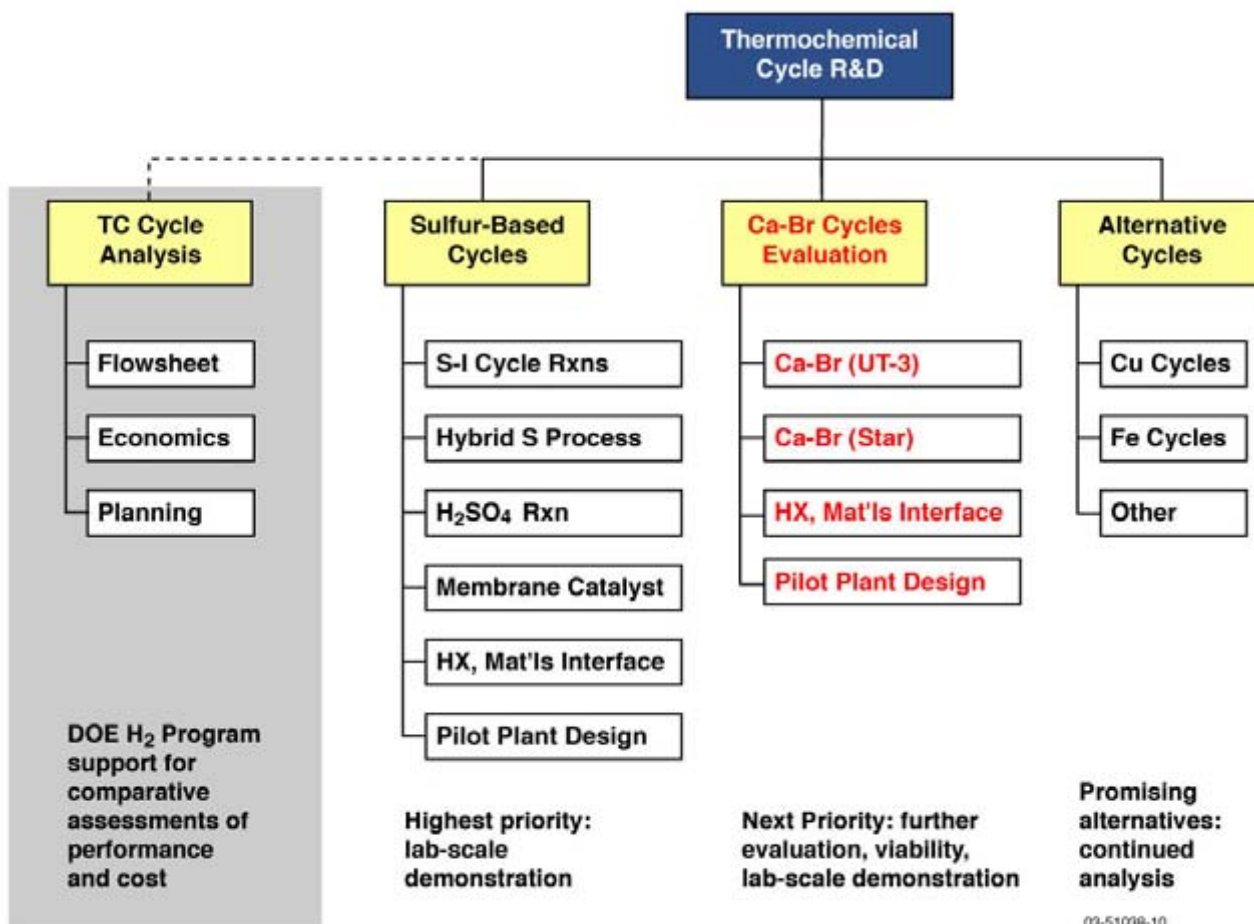


Figure 3-4. R&D program structure for the development of thermochemical processes for NHI.

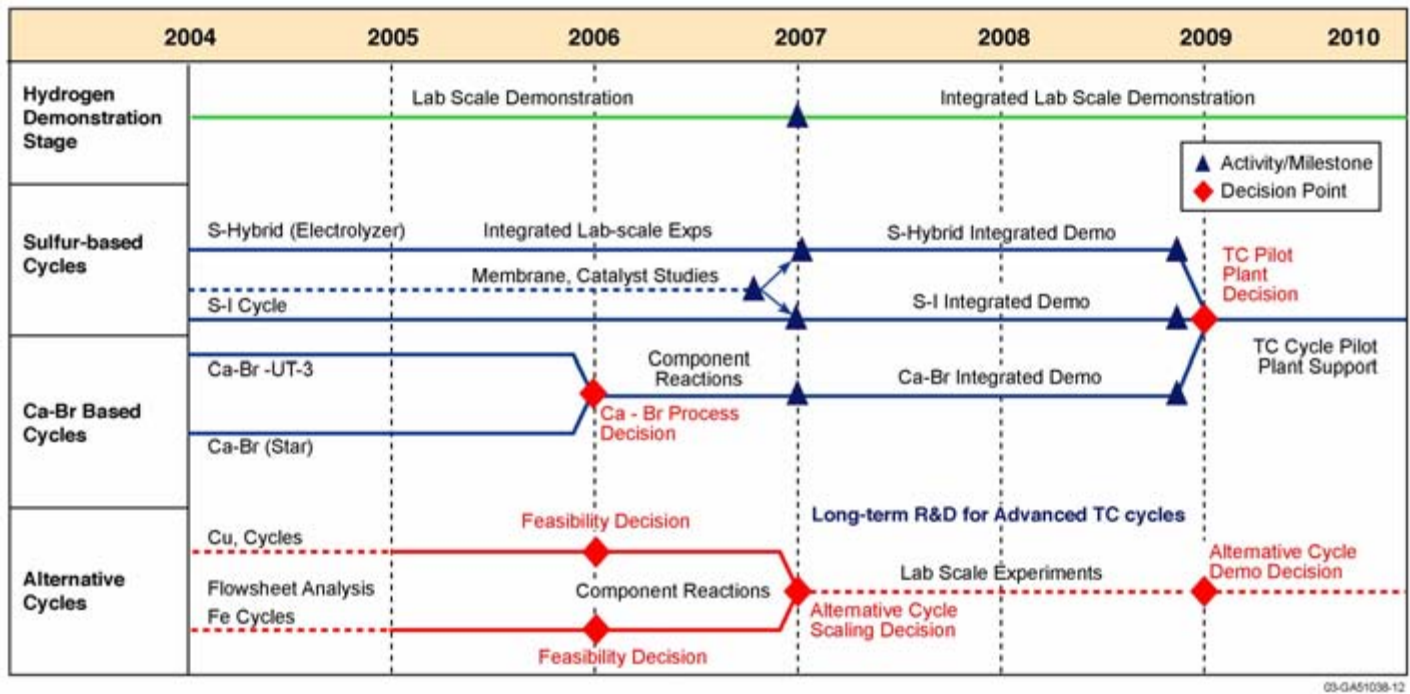


Figure 3-5. Thermochemical cycle major research areas and R&D sequence.

Table 3.2. Thermochemical R&D milestones.

Year	Sulfur Cycles	Ca-Br Cycles	Alternative Cycles
FY2004	Complete comparative analysis of S-I cycle based on recommended NHI methodology Complete final component reaction section flowsheets incorporating alternative HI and H ₂ SO ₄ flowsheet results Complete final design for the S-I thermochemical laboratory-scale system	Complete preliminary flowsheet analysis for candidate Ca-Br cycle approaches, estimate performance potential and identify the most promising approaches for the Ca-Br thermochemical cycle Define lab scale demonstration approach for the selected Ca-Br cycle approach	
FY2005	Construct S-I component reaction systems Assess high temp membrane for H ₂ SO ₄ decomposition High-temperature materials test assessment Hybrid sulfur system concept definition.	Complete flowsheet analysis for UT-3 and Star versions of Ca-Br Complete cost/performance assessment Star approach evaluation	Flowsheet analyses of Cu and Fe cycles Catalyst assessment
FY2006	Complete demonstration of S-I component reactions Construct S-I integrated loop Hybrid electrolyzer tests High temp HX tests for H ₂ SO ₄	Initiate component reaction experiments Design integrated demonstration loop UT-3 and Star experiments	Flowsheet and economic analyses of alternative cycles Design lab-scale experiments
FY2007	Start S-I integrated demo (100 l/hr) Start hybrid S integrated demo (100 l/hr) Engineering HX tests Conceptual pilot plant designs for S-I, hybrid	Ca Br scaling scaling decision Construct Ca- Br demo loop for Ca-Br cycle Initiate integrated lab scale exps	Lab-scale experiments for alternative cycles
FY2008	Integrated S-I, demonstration tests Integrated hybrid S demonstration tests Preliminary pilot plant designs for S-I and/or hybrid cycles	Perform Ca-Br integrated lab scale tests Conceptual Ca- Br pilot plant design	Lab-scale experiments on alternative cycles
FY2009	S based cycle pilot plant decision Design sulfur cycle pilot plant Process improvement R&D	Ca- Br pilot plant design	Integrated demonstration for alternative cycles
FY2010	Construct sulfur cycle pilot plant Conceptual design for engineering demo	Ca-Br pilot plant construction	Conceptual pilot plant design for alternative cycles

4. HIGH-TEMPERATURE ELECTROLYSIS

Electrolysis is the most straightforward approach currently available to produce hydrogen directly from water. Conventional electrolyzers are available with electrical-to-hydrogen efficiencies of around 70% at a cost of about \$400/kilowatt electric (kWe) installed. For electrical conversion efficiencies of 33 to 40%, the overall energy efficiency of hydrogen production is 23 to 28%. Conventional electrolysis allows distributed production of hydrogen in limited amounts. Since conventional electrolyzers are generally limited by membrane characteristics to relatively small outputs (kW to MW range), a large-scale production application would be composed of many electrolyzer modules. Although production efficiencies for large quantities of cells can lead to reduced costs per cell, the cost effectiveness of the modular scaling for electrolysis in comparison with the scaling of thermochemical methods is one of the issues to be evaluated.

High-temperature electrolysis (HTE), or steam electrolysis, has the potential for higher overall efficiency. Thermal energy is used to produce high-temperature steam, which results in a reduction of the electrical energy required for electrolysis and, therefore, a reduction in the total energy required for hydrogen generation. HTE can be accomplished using materials and technology similar to those used in solid oxide fuel cells (SOFCs). Electrolysis approaches allow the use of off-peak electricity for hydrogen production, and the economic analysis of the HTE approach will consider the potential for a dual-use plant based on the high-temperature fuel cell/electrolyzer. This potential cost advantage can be negated if the high-temperature cell remains too costly for large-scale application. Conventional electrolysis (a commercially available technology) serves as a baseline for the evaluation of the cost and benefit of advanced electrolysis methods. High-temperature methods are expected to result in lower hydrogen costs than conventional electrolysis.

4.1 Objective

The objective of HTE research is to assess the potential of an HTE production plant coupled to a high-temperature nuclear heat source for the large-scale production of hydrogen. This assessment

requires the development of an HTE system design and the demonstration of improved efficiency and projected costs. Advanced high-temperature reactors can provide both the high-temperatures needed for superheated steam and the efficient electricity required for electrolysis. The projected target costs for hydrogen production are \$2.50/kg in 2011, and \$2.00/kg in 2017. These projected hydrogen cost targets mean that high-temperature electrolysis will be demonstrated at a scale and efficiency that allows projection to a commercial facility with these production costs.

4.2 Approach

Since electrolysis and related SOFC materials and technology are being developed in other parts of the DOE Hydrogen Program, the scope of the NHI R&D activity is to develop an HTE system design optimized for use with a high-temperature reactor and to demonstrate cell and module performance. The primary tasks for HTE development are as follows:

- Develop a conceptual design for an HTE system coupled to an advanced high-temperature reactor to provide a basis for cost and performance assessment.
- Demonstrate promising cell and module approaches in a series of scaling experiments to confirm performance for conditions relevant to an advanced high-temperature nuclear reactor.

The research performed in the NHI will be closely coordinated with EE and FE to identify interfaces with ongoing research relevant to HTE development.

4.3 High-temperature Electrolysis Assessment

High-temperature electrolysis uses a combination of thermal energy and electricity to split water in a device very similar to an SOFC. Fundamentally, the electrolytic cell consists of a solid oxide electrolyte (usually yttria-stabilized zirconia) with conducting electrodes deposited on either side of the electrolyte, as illustrated in Figure 4-1 (see Appendix 4-1 for

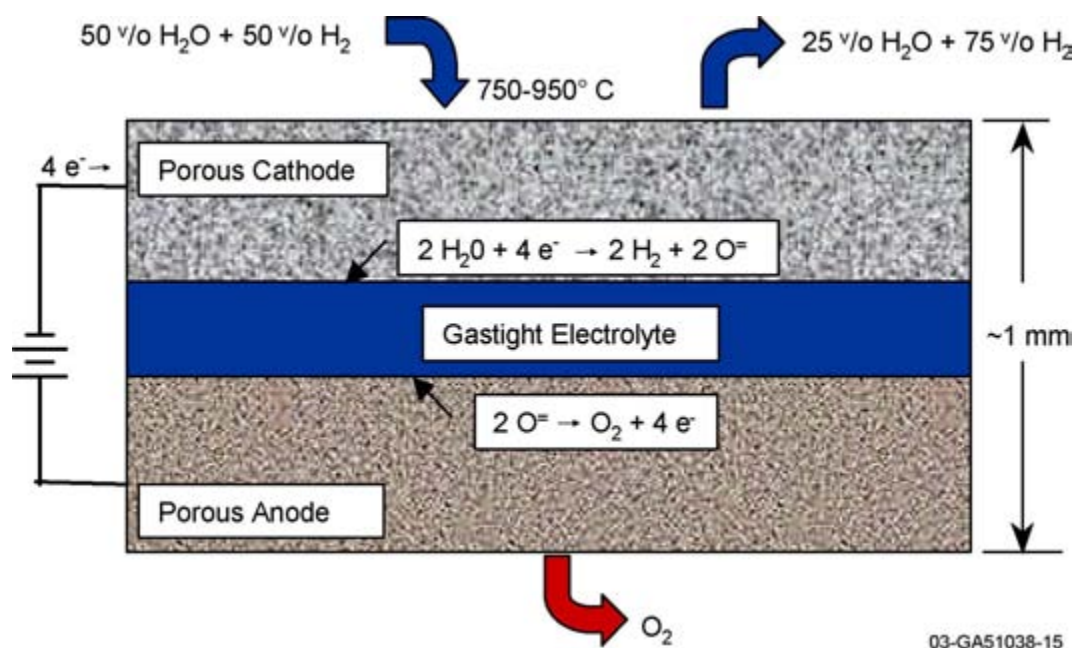


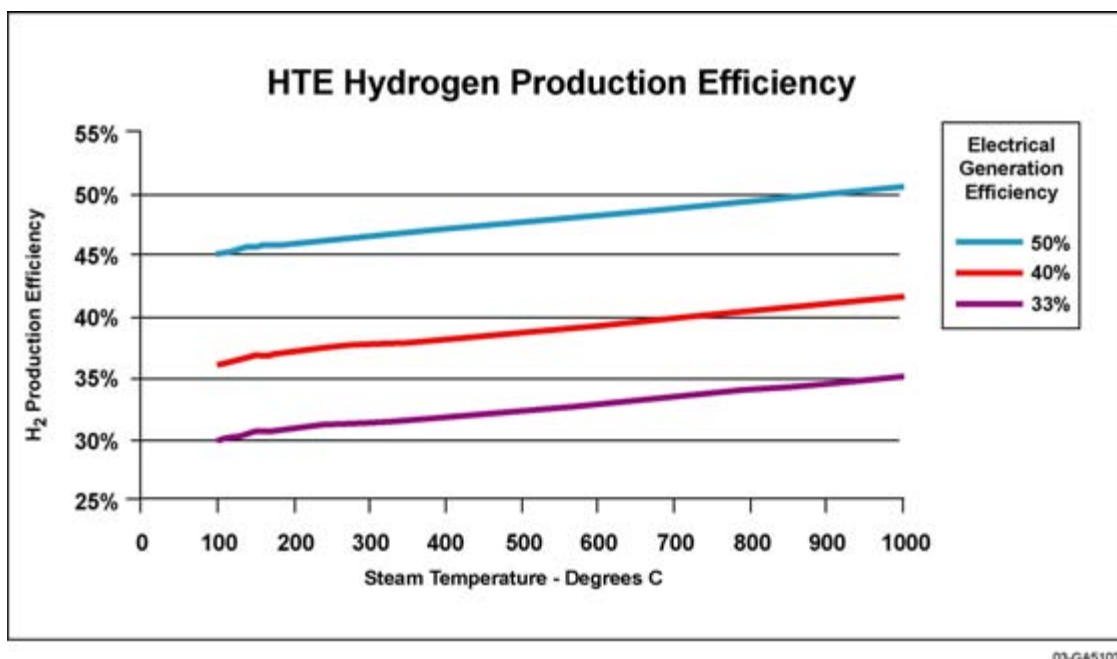
Figure 4-1. Typical high-temperature electrolysis cell configuration.

details). A nominally 50:50 (volume percent) mixture of steam and hydrogen at 750-950°C is supplied to the anode side of the electrolyte. Oxygen ions are drawn through the electrolyte by the electrical potential and combine to oxygen on the cathode side. The steam-hydrogen mixture exits at about 25:75 volume ratio, and the water and hydrogen gas mixture is passed through a separator to separate hydrogen. While present experiments and fuel cells operate near atmospheric pressures, future cells may operate at pressures up to 5 MPa.

Because of shrinkage during sintering in current manufacturing processes, the size of individual cells is limited to about 15 cm². Therefore, a high-temperature electrolysis plant powered by a reactor would consist of an array of relatively small modules connected together with the necessary high-temperature gas manifolding, electrical, and control connections. Costs for SOFCs are currently high (~\$10k/kWe), primarily due to small-scale manufacture. Ongoing SOFC research is investigating approaches to reduce both materials and manufacturing costs. Current estimates are that large-scale manufacturing could potentially reduce costs by an order of magnitude.

4.3.1 High Temperature Electrolysis Energy Efficiency

To be an attractive option, the potential for increased overall efficiency of HTE over conventional electrolysis must offset any increases in capital costs that might be incurred due to the more complex, high-temperature, electrolytic cell configuration. The improved efficiency is achieved through the substitution of thermal energy for electrical energy. A less efficient electrical generation process results in a greater overall benefit of using thermal energy directly. The magnitude of the potential efficiency improvement is illustrated in Figure 4-2 as a function of steam temperature and a range of electrical conversion efficiency assumptions. The plotted lines show the efficiency of the electrical generation process supplied to the cell. The largest efficiency improvements occur for the lowest electrical conversion efficiencies. For current light water reactor electrical efficiencies (33%), the improvement is on the order of 20%. This efficiency improvement is potentially significant since it reduces not only the size and capital cost of the hydrogen plant, but also reduces the size and cost of the nuclear plant required to produce a given amount of hydrogen.



03-GA51038

Figure 4-2. Efficiency improvement for high temperature electrolysis as a function of steam temperature.

4.3.2 High-temperature Electrolysis Production Plant Features

A schematic diagram of a nuclear hydrogen plant using HTE is shown in Figure 4-3 (see Appendix 4-2). The reactor (in this case an HTGR) supplies both electricity and steam to the electrolytic cell. The steam generator supplies superheated steam to the cells at a temperature of 750 to 950°C, and at a pressure of 1 to 5 MPa. The input gas contains both steam and hydrogen to maintain reducing conditions. A scoping system design that defines initial system parameters for an HTE plant is summarized in Appendix 4-2.

In addition to high-temperature electrolyzer technology, the development of a cost-effective HTE process poses challenges for heat exchangers and materials (750 to 950°C steam), thermal distribution and recovery systems (manifolds and heat loss), and cell engineering (materials and sealing). The development of HTE units for use with nuclear reactors will make use of the materials and configurations of SOFCs. The development of SOFCs has been supported by FE through the Solid State Energy Conversion Alliance (SECA), an industry-DOE collaboration. The NHI research needs identified for high-temperature electrolysis are those issues specific to the development and integration of

solid oxide electrolysis cells (SOECs) for nuclear application. The NHI will coordinate with ongoing research in the SECA program and other DOE SOFC programs.

4.4 High-temperature Electrolysis R&D Needs

The primary categories of nuclear-specific HTE R&D that must be addressed are as follows:

- System design studies to support cost and performance assessment for HTE nuclear plants
- HTE cell and module optimization to support technology demonstration.

4.4.1 System Design and Trade Studies

The most pressing research need for HTE is the development of a conceptual design to identify the component needs specific to nuclear applications of the technology. The NHI design study will identify the changes in present SOFC materials, configurations, and operating modes necessary for HTE. For example, the tubular SOFC design is the

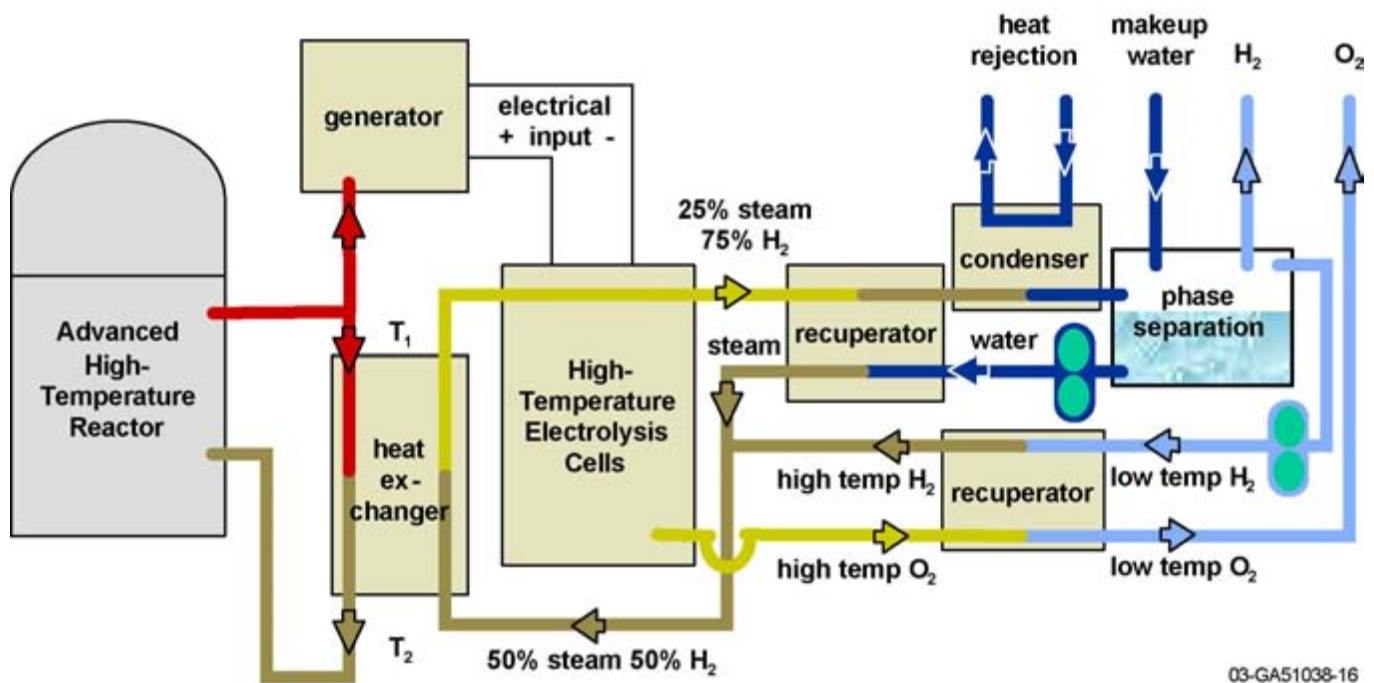


Figure 4-3. Major components of a conceptual nuclear high-temperature electrolysis plant.

most developed of the SOFC designs to date. However, the tubular configuration requires approximately ten times more hot volume than a planar configuration for the same hydrogen production rate.

Trade studies are needed to determine the economic and engineering impacts of the cell configuration, arrangement of the steam generator, need for reheating the steam and/or heating the oxygen carrier gas, methods for steam-hydrogen separations, and the choice of operating pressure. The overall design of the HTE plant can be optimized in relation to the electrical grid requirements, peak power management, and grid stability.

Models of HTE processes and systems performance are also required to support the conceptual design and assess HTE cell performance. These analyses include cell and module thermal and structural models. These models address the behavior of the electrodes, electrolyte, interconnection, and seals in the face of thermal and chemical challenges. Electrochemical models are also needed to describe materials flows through the individual cells. These analyses will define temperature and current distributions, particularly in response to changes in materials properties. Thermal models of the HTE system and reactor interface, including heat exchangers,

separators, and flow-control devices are needed to optimize overall plant performance.

The costs of power electronics are an important output of these trade studies. Rectifiers for the conversion of alternating current to direct current are a major cost in the overall HTE plant. Proper choices of stack voltage and current and the use of standard components may substantially reduce the cost of power conditioning and control.

4.4.2 High-temperature Electrolysis Cell / Module Optimization

Although the development effort for solid oxide materials for high-temperature fuel cells has been substantial, the engineering and optimization of SOFCs are at an early stage. Several engineering issues must be addressed as part of the development of the cell/module for an HTE hydrogen production plant.

- *Interconnections* – The use of metallic interconnection between planar cells would result in lower ohmic losses, improved resistance to thermal and mechanical shock, and reduced manufacturing costs. The choice of interconnection material is closely related to the choice of electrolyte, since the ionic

Table 4-1. Summary table of high-temperature electrolysis research needs.

R&D Area	Technical Issues	HTE Requirement	Ongoing Research	Needed Research
System Design Studies	HTE System Configuration	100 – 300 MW Hydrogen	NE Lead	Design and trade studies for materials and component selection. Cell-level and plant-level modeling necessary.
	Plant Design Trade Studies	Engineering, Economics		
	HTE Process Models	Reheat of Steam		
	Power Electronics Costs	Rectifiers AC to DC		
	Material Demands	HTE Large Materials Req's		
Cell / Stack Optimization	Interconnections	Thermal Cycling	FE Lead	Monitor FE research.
	Cell Sealing	Compressive or Steam Sealing		
	Electrolyte Performance	YSZ Satisfactory at High Temps		
	Cathode Material	HTE Less Challenging		
	Anode Materials	Oxygen Diluent		
Cell/Stack Demonstration	Performance for NHI Temp, Pressure Conditions	Technology Demo by 2006	NE Lead	Test program for candidate HTE options.

- resistivity of the electrolyte is temperature dependent, dropping by a factor of two between 750 and 850°C. Metallic interconnections would have to operate at lower temperatures than present day ceramic interconnections.
- *Cell Sealing* – Sealing between SOFC cells is a major issue in the choice of tubular versus planar configurations. The sealant must withstand both the oxidizing environment of the anode and the reducing conditions of the cathode. Tubular designs allow separation the oxidizing and reducing environments, while edge sealing in a planar configuration is more difficult. Design studies and laboratory tests are needed to address these issues.
- *Electrolyte Performance* – Research in the electrolyte material is seeking higher conductivity materials with comparable cost and chemical advantages. Advanced materials (e.g., scandium doped zirconia and lanthanum gallate with strontium doping) are being developed and will be examined for this application.
- *Cathode Materials* – The use of nickel-nickel oxide or nickel zirconia cermets will be satisfactory as long as a steam-hydrogen mixture (rather than pure steam) is used to

maintain reducing conditions. Cathode conditions are less challenging for HTE than for SOFC conditions, for which these cathode materials have been developed. Issues in the use of nickel with coal combustion gases pose more difficult problems due to the presence of sulfur and other contaminants.

- *Anode Materials* – Lanthanum strontium manganite (LSM) has been the most frequently used material in SOFCs, both tubular and planar. Perovskite-structured ceramics have been developed for applications at lower temperatures. The consensus of experts is that initial demonstrations of HTE could rely on LSM.
- *Materials costs* – The operation of the SOFCs at high temperatures requires the use of expensive alloys through the balance of plant. Therefore, the development of materials for lower temperature operation may result in overall reduction in the hydrogen cost due to a reduced capital investment in the plant. The impact of reduced materials costs is a central consideration in the trade studies described above.

The R&D needs for the development of the nuclear HTE system are summarized in Table 4-1.

4.5 High-temperature Electrolysis R&D Program

The sequence and interaction of the major HTE areas are summarized in Figure 4-4. The planning assumptions for HTE include early testing of candidate cells (button) based on materials being developed in the DOE fuel cell program. The NHI activities will focus on cell/stack testing to provide performance information for pilot plant decisions. The design and operation of a pilot plant at the nominal 200kW level could be initiated at a smaller power level and expanded as improved materials and electrolyzer cells become available.

The highest priority for nuclear HTE development is to complete a conceptual design that can be used to support cost and performance assessments in FY 2004. With confirmation of performance and cost

effectiveness, the NHI research effort will focus on the demonstration of HTE cell performance based on the most promising materials and technology available from the FE program. The cell engineering efforts will support the demonstration of SOFC modules in FY 2005-2006, while the NHI technology demonstrations will provide the basis for pilot plant decisions in FY 2009. The EE and FE technology research and the NHI system design and cell demonstration activity will be closely coordinated to ensure the most efficient use of R&D investments from all DOE offices.

4.6 High-temperature Electrolysis Milestones

The major R&D milestones associated with HTE are summarized in Table 4.2

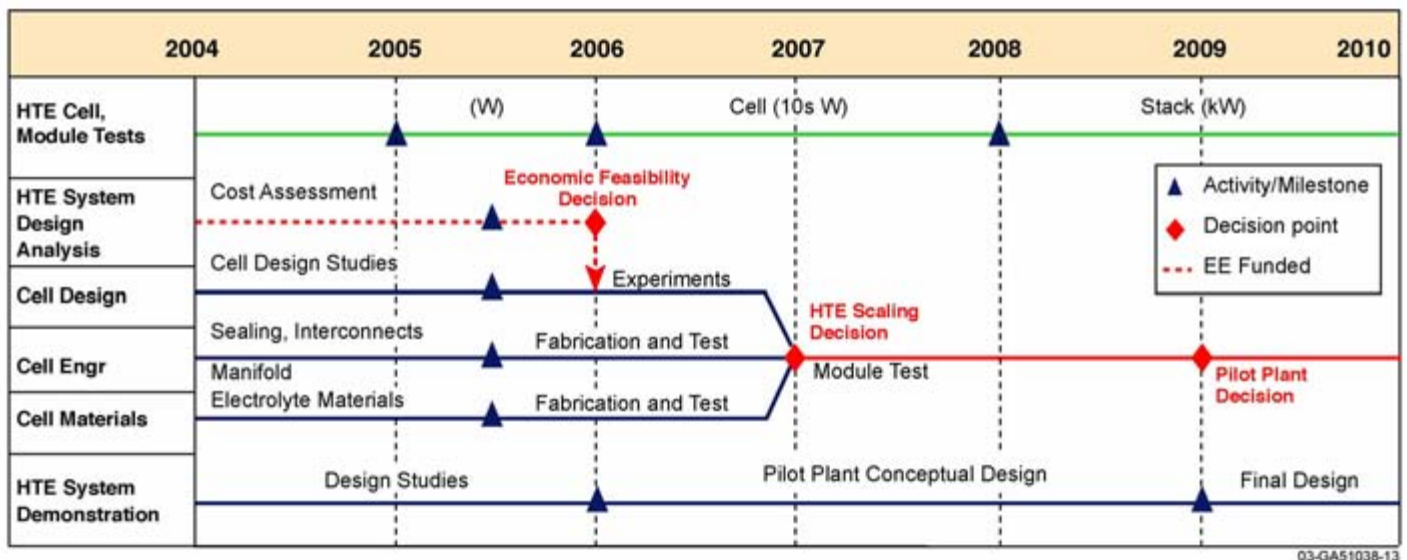


Figure 4-4. High-temperature electrolysis R&D plan.

Table 4-2. High-temperature electrolysis R&D milestones.

Year	HTE Systems Analysis	Cell Optimization
FY2004	Perform engineering analysis to support the conceptual design of HTE scaling demonstration experiments, including electrolyzer cell and module configurations, and steam distribution systems Complete a conceptual system design for a high-temperature electrolysis system coupled to a high temperature reactor Complete preliminary designs for HTE scaling experiments at pilot and engineering scales	Construct the HTE cell stack testing capability to evaluate electrolyzer characteristics and performance Complete single (button) cell experiments on candidate electrolyte materials for HTE
FY2005	Complete HTE preconceptual design Complete HTE system cost assessment (with EE) Develop engineering model for HTE system performance evaluation (thermal, structural, thermoelectric process)	Establish technology interface on materials, cell engineering and design options (with FE, EE) Define HTE cell/module options, and develop cell and module test plan (FY05-07) Button cell experiments (with EE)
FY2006	Complete thermal/structural analyses of module/stack and manifold options Develop conceptual HTE pilot plant design Design integrated lab-scale HTE demo	HTE cell testing for highest priority cell options Construct stack /module arrays, initiate testing Develop conceptual pilot plant module design
FY2007	HTE pilot plant preliminary design Conduct integrated lab-scale demo experiments	Complete HTE cell testing Conduct HTE stack / module tests Candidate pilot plant module tests
FY2008	Conduct integrated lab-scale demo experiments	Complete HTE stack / module tests Implement cell/module technology improvements
FY2009	Pilot plant decision HTE pilot plant final design Pilot plant operation	Cell technology support
FY2010	Conceptual design for Engineering demo Pilot plant construction	

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5. SYSTEMS INTERFACES AND BALANCE OF PLANT

Hydrogen production systems coupled with a high-temperature nuclear reactor introduce new considerations and requirements into the design of a combined nuclear hydrogen plant. Both high-temperature electrolysis and thermochemical systems must be co-located with the nuclear heat source. The interface between the reactor and hydrogen production system involves potentially long heat transfer paths at elevated temperatures, heat exchangers that are subject to both elevated temperature and corrosive chemical environments, new safety and regulatory issues, and supporting systems for chemical processes and hydrogen and oxygen transfer and storage. Although some of these issues will be common to any nuclear hydrogen plant, many will depend on the specific hydrogen production process selected.

5.1 Objective

The objective of the systems interface and balance-of-plant research effort is to develop the high-temperature interface components and materials required to couple the thermochemical or high-temperature electrolysis system to the high-temperature heat source. Interface requirements are defined by the characteristics of the thermochemical and HTE processes, and these activities are focused on developing the components needed to demonstrate selected processes.

5.2 Approach

The NHI systems interface research will focus on the heat exchanger and materials required for the high-temperature components and materials needed for the baseline processes, namely, the sulfur and calcium-bromine thermochemical cycles and high-temperature electrolysis. Development of technology for the sulfur cycles will be the initial focus, but priorities will track the production process research priorities. R&D will also address the intermediate loop

requirements at the production process interface and the more general supporting systems and safety issues that are associated with the coupled nuclear hydrogen plant.

It is recognized that significant R&D activities are being pursued in other DOE offices, government agencies, and industry. These systems interface activities will complement or collaborate with other ongoing DOE or industrial system research efforts to avoid any potential duplication. Therefore, this report will focus on the R&D issues unique to nuclear hydrogen production that are not currently being planned or pursued under other programs.

5.3 Systems Interface and Balance-of-Plant Assessment

The primary balance-of-plant systems and interfaces are shown schematically in Figure 5-1. The high-temperature reactor provides nuclear process heat to the hydrogen plant through an intermediate heat transfer loop. The intermediate loop working fluid and materials of construction are important engineering issues at these elevated temperatures. Physical isolation, either separation distance or an acceptable physical barrier, must be sufficient to eliminate the propagation of accident consequences from one plant to the other. Additionally, numerous supporting systems are required for gas handling, storage, and process support. High-temperature heat exchangers that are required to transfer heat from the reactor primary systems to the hydrogen process are critical elements for efficient coupling of the nuclear hydrogen plant.

The major balance-of-plant/plant interface systems are summarized in Table 5-1 along with the key issues to be addressed in the design phase of the project. Appendix 5-1 provides additional information on major system interfaces and balance-of-plant technology status.

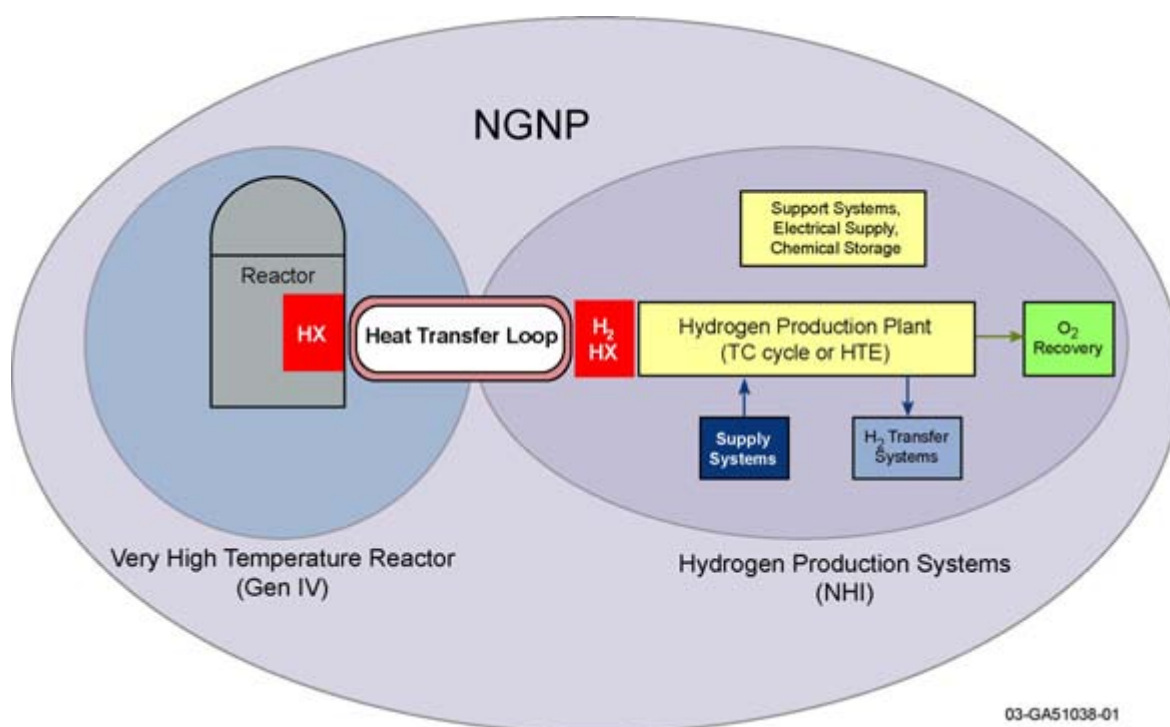


Figure 5-1. Schematic of the major reactor-hydrogen interfaces and balance-of-plant systems.

Table 5-1. Major hydrogen plant interfaces and balance-of-plant systems.

Systems/Components	Design /Interface Considerations
High-temperature Heat Exchangers	HX interface to production process, HX design options, operational conditions (temperature and pressure) High-temperature materials compatible with heat transfer medium and production process chemical species
Intermediate Loop	
Heat Transfer Medium	Heat transfer conditions – temperature, pressure, pumping power, heat loss requirements, working fluids
High-temperature Transfer Lines	Isolation configuration, materials, operating conditions (temperature, pressure, and fluids), insulation, seals, auxiliary heating
Support Systems	
Oxygen Recovery/Disposal	Oxygen inventory, recovery, or disposal approach, storage, safety issues
Oxygen Safety Systems	Oxygen hazards (combustion and health), materials corrosion, ventilation, fire systems, diagnostics, environmental control
Hydrogen Safety Systems	H ₂ diagnostics, storage, combustion, ventilation systems, explosive hazards,
Hydrogen Transfer	Transfer system to interface with pipeline, storage systems
Chemical Support Systems	Chemical feed, storage, purification, diagnostics, process controls
Instrumentation and Control	H ₂ process diagnostics and controls, interface with reactor systems,
Electrical	Hybrid process, grid, station power systems
Alternative Heat Source	Interim process heat requirements for testing or production

5.4 Systems Interface and Balance-of-Plant R&D Needs

The primary systems interface and balance-of-plant research areas are as follows:

- Nuclear hydrogen system design studies to define configurations and component requirements
- High-temperature heat exchanger design and materials
- Intermediate heat transfer loop and materials
- Hydrogen plant support systems.

5.4.1 Nuclear Hydrogen System Design Studies

System design studies for the system interface and balance-of-plant areas address plant configuration options for both thermochemical and high-temperature electrolysis systems, safety and isolation issues for the coupled plants, and assessment of applicable codes and standards. Since these studies define fundamental options for the nuclear hydrogen plant configuration and nuclear interface, they must be addressed early in the FY 2004 to 2006 time frame.

- *Hydrogen plant configuration studies* define configuration options and the operational conditions and requirements for the hydrogen plant subsystems. The isolation approach for nuclear and hydrogen production systems, both thermochemical and electrolytic processes, has performance and economic implications that need to be addressed at the earliest possible stage. These studies define options and tradeoffs for the optimum coupling of the nuclear and chemical systems.
- *System interaction studies* define safety and isolation issues arising from system level considerations and from functional and physical coupling of the nuclear and hydrogen plants. Nuclear hydrogen production systems will require a new or modified framework for regulation. Identification of information on combined plant operational requirements that could

impact system design and preliminary definition of accident categories may be important in process evaluation. The tasks that should be addressed include assessment of regulatory requirements, definition of separation and isolation criteria, and preliminary plant safety studies.

- *Applicable code assessment* – Numerous codes, standards, and guidelines are applicable to hydrogen and oxygen, but no definitive compilation was identified. The R&D effort should include a compilation of applicable codes, standards, and guidelines for the nuclear plant and the hydrogen production processes of interest, and identify any issues.
- *BOP interface with hydrogen production process and NGNP* – Definition of design and performance requirements determined by hydrogen production process characteristics and high-temperature reactor capabilities will be required to establish conditions for balance-of-plant systems.

5.4.2 Heat Exchanger Design Studies

High-temperature heat exchangers that transfer thermal energy to chemical processes or steam at elevated temperature are critical components. Operational conditions for the interface heat exchangers are challenging. The heat exchanger for the sulfur-iodine and hybrid sulfur processes require decomposition of sulfuric acid at more than 900°C and up to several megaPascals (MPa). Heat exchangers for the high-temperature electrolysis process will actually be steam generators producing steam at up to 950°C and up to 5 MPa. The primary R&D areas that must be addressed include:

- *Thermal analyses* between intermediate loop and process conditions to define operational requirements for heat exchangers for the range of candidate cycles to support heat exchanger design studies.
- *Heat exchanger design studies* to explore innovative heat exchanger design approaches that could potentially mitigate temperature or lifetime requirements. A range of heat exchanger design approaches can be

considered (tube/shell and printed circuit) that involve differing materials and fabrication issues. Heat exchanger viability is an essential consideration in the selection of a pilot plant hydrogen process.

- *High-temperature materials test programs* are required to demonstrate performance and lifetime under prototypic conditions. Based on the operating conditions and requirements for process-specific heat exchangers, this task will identify and initiate materials testing programs for the candidate cycles. This activity also addresses fabrication issues for heat exchanger designs and will require industrial participation in their design and fabrication. Scaled demonstration testing is required for selected heat exchanger systems. Although no current material has demonstrated capabilities in the full temperature and composition ranges being considered for hydrogen production processes, considerable information is available on materials that have potential and have been demonstrated for most conditions.

5.4.3 Intermediate Heat Transfer Loop

Intermediate loop studies will define the interface with the reactor heat source, including operational conditions, materials, and controls. The selection of the heat transfer medium and transfer line configuration and materials are key technologies that impact both reactor and process design choices.

- *Heat Transfer Medium Materials* – The selection of the medium/fluid for the intermediate heat transfer loop impacts plant configuration, separation distance decisions, heat exchanger design, and materials selection. The medium must be compatible with temperatures of approximately 1000°C, pressures of several MPa, and reasonable pumping/circulating requirements, as well as chemical compatibility with heat transfer loop materials.

Molten salts, helium, and liquid metal are potential candidates for the heat transfer medium. The commonly used fluoride salts

meet the high-temperature and low-pressure requirements, and their viscosities are close to water at the temperatures being considered, which translates to relatively low pumping costs. Helium is also a logical heat transfer medium, but the gas heat capacity results in extremely large volumes of medium needed to transfer heat, resulting in large line sizes and high blower operating costs. Liquid metals can also meet the temperature and pressure range with different material compatibility issues. Liquid metals are also potentially expensive systems, have purity and pumping component issues, and have high densities and viscosities that will result in higher pumping costs over long distances.

- *High-temperature Transfer Lines* – The primary R&D activities for the intermediate heat transfer loop transfer lines focus on thermal analyses and materials development. Materials development will be required for piping and seals, circulator (pumps or blowers), valves, and structural materials (900 to 1000°C range). Materials for molten salt structural materials, pumps, and valves have a technology gap between previously demonstrated use at approximately 700°C and the required range of 900 to 1000°C. The technology gaps for use of helium involve equipment sizes and blower operating costs. The technology gaps in the use of liquid metals, which include pumping, high-temperature valve development, and potentially material compatibility issues, lie between 450°C and the required range of 900 to 1000°C.

5.4.4 Supporting Systems Scoping Studies

Preliminary design studies of the supporting subsystems for hydrogen, oxygen, and chemical systems are required for the hydrogen production process and preliminary designs that support pilot plant decisions. Operational conditions and requirements for the range of NHI production options will be defined, and scoping studies of the engineering configuration and associated costs will be developed.

5.5 Systems Interface and Balance-of-Plant R&D Program

Systems and balance-of-plant activities are directly coupled to the development and schedule of the hydrogen production processes. High-temperature materials and heat exchanger development and demonstration are essential to a successful demonstration of hydrogen production processes.

The heat exchanger design and materials testing activities must be sequenced to support production process R&D and pilot plant design and construction. A summary level diagram of the major elements and interactions of the balance-of-plant program is shown in Figure 5-2.

A summary of the major systems interfaces and BOP milestones that will be accomplished in support of the production process demonstrations is shown in Table 5.2.

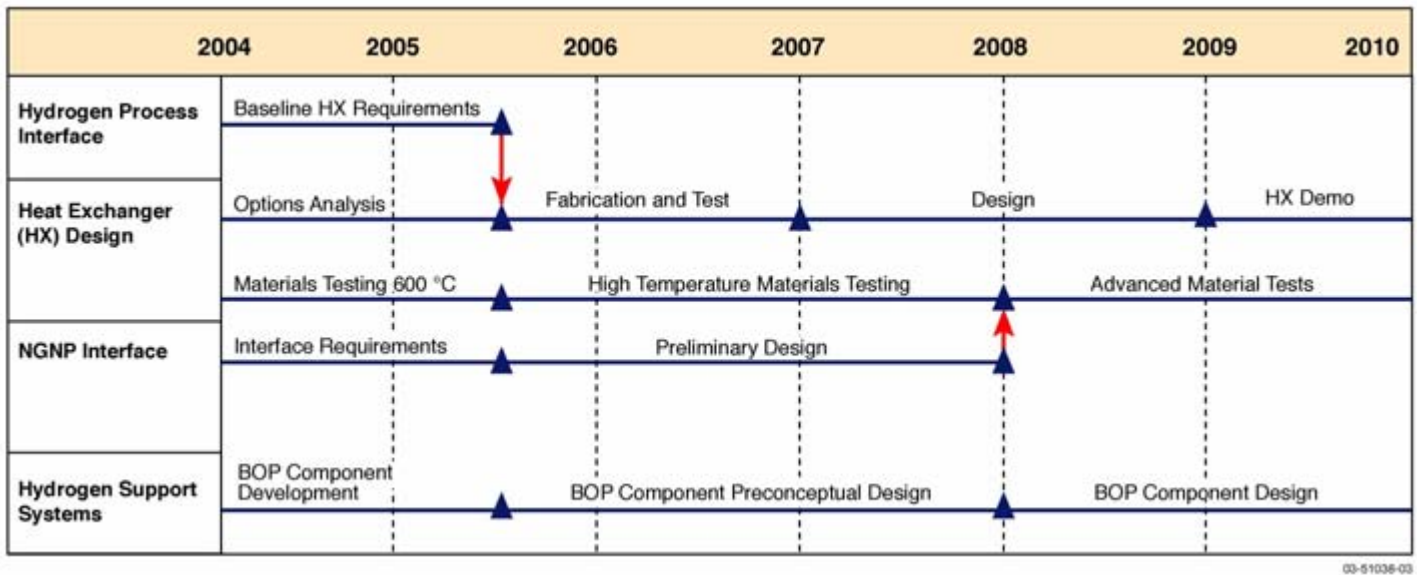


Figure 5-2. Systems interface and BOP R&D activities.

Table 5-2. Summary of major systems interface and BOP milestones.

Year	Systems Studies	High-temperature HX	Intermediate Loop	Supporting System Design
FY2004	Complete initial configuration studies to define balance of plant components and systems for thermochemical or high-temperature electrolysis systems.	Identify key technical issues for intermediate loop candidate designs and develop research requirements and priorities	Evaluate candidate coolants and develop conceptual designs for promising approaches	Complete high level infrastructure and facilities requirements assessment for pilot scale experiments
FY2005	Develop H ₂ plant configuration options for baseline processes Perform initial isolation assessment Compile applicable codes and standards	Develop process specific HX requirements for baseline processes Develop conceptual design options for high H ₂ SO ₄ decomposition HXs Initiate materials testing for Sulfur cycles	Perform thermal, flow analyses for intermediate loop options	Assess external (to nuclear hydrogen plant) interface requirements
FY2006	Complete preliminary H ₂ system cost studies for baseline processes	Initiate materials testing for Ca-Br or alternative cycles Scaled HX design and fabrication for Sulfur cycles and HTE	Intermediate loop preliminary design	Conceptual design for H ₂ system components and subsystems (pilot plant scale)
FY2007	Interface and BOP design for pilot plant Thermal, structural, flow for baseline pilot plant designs	Long-term materials tests Pilot plant HX design	Intermediate loop materials testing	Component development for pilot plant processes
FY2008	Pilot plant systems support	Long term materials tests High-temp HX demo tests High-temp HX tests		Pilot plant construction support
FY2009	Pilot plant systems support	Pilot plant support		
FY2010	Conceptual design for baseline engineering demo	Engineering demo HX design Pilot plant support		Engineering demo conceptual design

6. NUCLEAR HYDROGEN R&D PLAN SUMMARY

This Nuclear Hydrogen R&D Plan defines the research necessary to develop hydrogen production options for the demonstration of hydrogen production from nuclear energy by 2017. The technical challenges to the development of large-scale, cost-effective production options are significant, but promising approaches have been identified, all of which meet the DOE objectives of emissions-free production based on domestic resources. The fundamental challenge is to focus finite research resources on processes that have the highest probability of producing hydrogen at costs that are competitive with gasoline. Both thermochemical cycles and high-temperature electrolysis methods have been identified as having the potential to achieve this objective.

Though early in the development stage of these promising methods (none have been demonstrated at a pilot plant-scale), this Nuclear Hydrogen R&D Plan identifies baseline processes that meet NHI performance criteria and are sufficiently demonstrated to provide reasonable confidence that the processes would be technically viable in a large plant. Two thermochemical cycles (sulfur family and calcium-bromine) and HTE (based on fuel cell technology) were identified as the highest priority production processes for further development. The recognition of several common R&D issues applicable to more than a single cycle further leverages the research investment.

Initially, a broader research effort is required. The planned R&D effort will include laboratory-scale demonstrations, where justified by the available information, in addition to analytical evaluations of

those processes with promise but whose viability is uncertain. This approach will provide a more consistent and complete assessment upon which to base future R&D funding and scaling decisions. NHI research will follow a systematic approach to developing the required information for the sequence of scaling decisions. Confirmation of performance potential based on consistent thermodynamic analyses of candidate cycles will be confirmed in laboratory-scale tests to support pilot plant scaling decisions. Pilot plant demonstrations of the selected processes confirm engineering viability and establish a basis for process cost estimates. Pilot plant performance and cost information provides a basis for selecting the NGNP nuclear-heated engineering demonstration.

Both thermochemical cycles and HTE processes challenge current capabilities for high-temperature heat exchangers and materials. R&D for the nuclear and hydrogen systems interface and balance-of-plant systems and components will be required to provide viable engineering options for the range of processes being evaluated.

The NHI program office will establish the capability to provide consistent and independent analyses of performance and costs to support the comparative assessments required for technology selection and scaling decisions. The magnitude of the nuclear hydrogen research effort also suggests that the NHI program office establish effective interfaces with industry and international partners to take advantage of the significant expertise in hydrogen production.

This nuclear hydrogen R&D strategy is summarized in Figure 6-1.

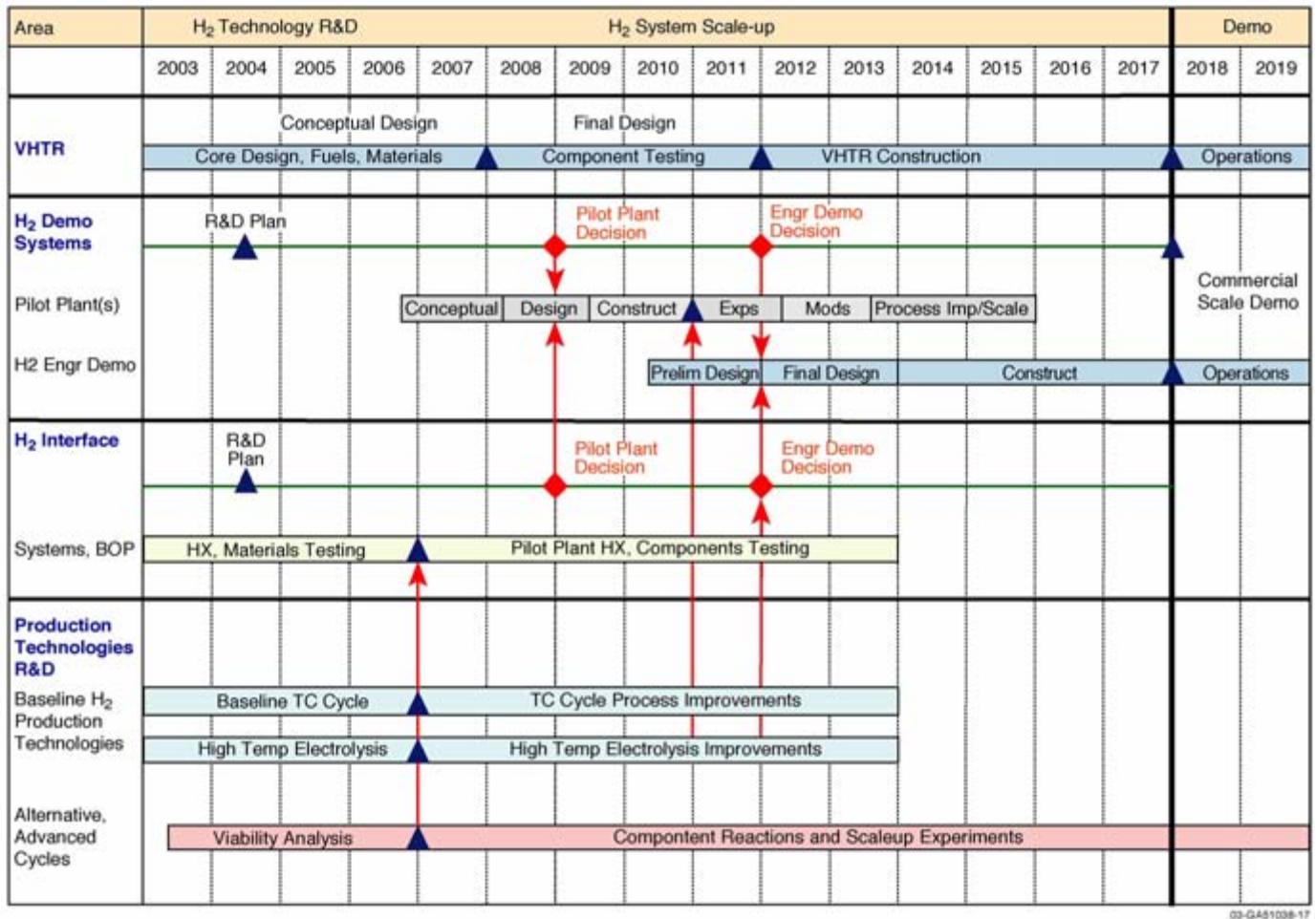


Figure 6-1. Summary of NHI R&D plan for the development and demonstration of a nuclear hydrogen production capability by 2017.

APPENDICES

[THE APPENDICES ARE NOT INTENDED TO BE PRINTED AND BOUND WITH THE R&D PLAN, BUT WILL BE ATTACHED AS A CD FOR SELECTED DISTRIBUTION, AND AVAILABLE TO ALL OTHERS UPON REQUEST]

Appendix for Chapters 1 and 2 – Introductory Chapters

Appendix 1-1. Current Hydrogen Production Processes: Steam Methane Reforming

Appendices for Chapter 3 – Thermochemical Cycles

Appendix 3-1. Thermochemical Process Descriptions and Status

Appendix 3-2. Membrane Applications for Hydrogen Production Using Nuclear Energy

Appendix 3-3. Catalyst Requirements for S-I

Appendices for Chapter 4 – High-temperature Electrolysis

Appendix 4-1. Hydrogen Production through High-temperature Electrolysis in a Solid Oxide Cell

Appendix 4-2. Conceptual Design of a High-temperature Electrolysis Plant

Appendix for Chapter 5 – Systems Interface and Balance-of-Plant

Appendix 5-1. System Interfaces and Balance-of-Plant