II.J.1 Sulfur-Iodine Thermochemical Cycle

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Start Date: September 2002
Projected End Date: 2008
(completion of lab-scale experiments)

Objectives

- Evaluate the potential of the Sulfur-Iodine (S-I) thermochemical cycle for large-scale hydrogen production using nuclear energy.
- Perform an integrated lab-scale (ILS) experiment to demonstrate closed-loop operation of the S-I cycle.
- Provide the technical basis for DOE Nuclear Energy thermochemical selection and scale-up decisions.

Technical Barriers

This project addresses the following technical barriers for the Nuclear Hydrogen Initiative (NHI):

- Process chemistry and thermodynamic data information for evaluation of thermochemical cycle efficiency and operational characteristics.
- High-temperature, corrosion resistant materials for extended service under thermochemical process conditions.
- Heat exchanger development for the reactor-process interface.
- Hydrogen production facility costs.

Approach

- Perform flowsheet analysis of the S-I thermochemical cycle process and evaluate alternative configurations.
- Construct experiments to investigate the major reactions of the S-I cycle and perform stand-alone experiments to demonstrate the chemistry and operation of the three major reactions for the S-I cycle.
- Identify reactor-process interface heat exchange, safety requirements, system integration and control strategies.
- Develop improved materials and heat exchanger designs for service in high temperature corrosive environments.
- Based on the results of reaction experiments, design and construct an integrated lab-scale apparatus to provide a test bed for S-I cycle evaluation.
- Perform integrated lab-scale experiments to evaluate closed-loop operation.
- Develop system designs, sizing, and materials for a nominal mega-Watt pilot-scale experiment.

Accomplishments (through May 2007)

- Designed, constructed and completed initial tests for the H_2SO_4 decomposition section of the ILS experiment (SNL). This section is based on a fully integrated SiC bayonet heat exchanger design for high temperature acid decomposition. The unit has been shipped to GA for integration with the hydriodic acid (HI) and Bunsen sections later this year.
- Extractive distillation experiments for HI distillation and decomposition have been completed and construction of the HI reaction section is underway at GA.
- The Bunsen reactor section has been constructed and Bunsen reactor section experiments have been performed through the DOE-Commissariat a Energie Atomique (CEA) International Nuclear Energy Research Initiative (INERI) project.

Future Directions

FY 2007 (June-September)

- Complete assembly and testing of the H_2SO_4 decomposition and HI extractive distillation and decomposition sections for the ILS experiment.
- Assemble Bunsen section at GA and complete Bunsen section testing (CEA).
- Complete facility and safety process preparations for the ILS experiment.

- Perform system controls and integration tests to confirm readiness for integrated testing to begin in FY 2008.

FY 2008

- Conduct the initial integrated experiments in the ILS apparatus to evaluate system operation, control strategies, and performance.
- Identify and implement process improvements and complete the final experiments.
- Complete the S-I pilot-scale experiment final design.
- Assess S-I cycle performance for large scale hydrogen production using advanced nuclear reactors.

Introduction

Thermochemical cycles are promising options being investigated by the NHI for hydrogen production using nuclear energy. The NHI is investigating the sulfur-based thermochemical cycles (S-I and Hybrid Sulfur) to evaluate the potential of these cycles for hydrogen production coupled to a high temperature gas-cooled reactor. This project focuses on the S-I cycle with the objective of developing an ILS experiment to provide the technical basis for future technology selection and scale-up decisions. This project is being performed as part of a US-DOE, French-CEA INERI agreement. The CEA is designing and testing the primary (Bunsen) reaction section. GA is developing and testing the HI decomposition section. SNL is developing and testing the H_2SO_4 decomposition section. These three reaction sections were developed and tested as stand-alone experiments, and are now in the process of being assembled into an ILS experiment at GA to perform a closed-loop demonstration of the S-I cycle. Key issues that must be addressed include high-temperature materials in highly corrosive environments, process chemistry and thermophysical data uncertainties, innovative heat exchanger designs and materials to couple to the nuclear heat source, and systems evaluation of the potential performance and costs of these cycles for large-scale hydrogen production.

Approach

The approach taken in this project has been to first investigate process configurations in flowsheet analyses to determine the most promising approaches, and then conduct experimental studies to provide data on reaction chemistry, performance of materials, and catalysts. Supporting studies on material corrosion, catalyst stability, and separation membranes are also being pursued in the NHI. Information from the ILS experiment will provide the technical basis for decisions on scaling to higher power levels at a pilot-scale.

Results

The three reaction sections of the S-I cycle have now been constructed and testing is underway. The status of the three reaction sections is summarized in the following.

Sulfuric Acid Decomposition Section: Sulfuric acid decomposition experiments have been completed at temperatures up to 900°C using the SiC bayonet heat exchanger concept developed at SNL in FY 2006 (see Figure 1). This approach integrates the vaporizer, superheater, catalytic decomposer, and recuperator into a single bayonet unit. This design eliminates all high-temperature connections, allowing the use of glass- or Teflon-lined steel in lower temperature areas. This new design was developed and tested to confirm operation in the range of 200–300 l/hr of SO_2 production as required for the ILS experiment. Tests have confirmed that the SiC bayonet design minimizes corrosion issues and simplifies the operation of the acid decomposer.

The SiC bayonet design provides a more compact and robust acid decomposer system for the S-I cycle. The unit has been shipped to the GA test site for integration with the other S-I sections later this year.

HI Decomposition Section: The development of the HI decomposition section is based on an extractive distillation approach which separates I_2 from the HIx (blend of hydrogen iodide, iodine, and water) feed stream before decomposition. Experiments during the past year have quantified the liquid-liquid extraction step where the H_3PO_4 strips the HI and water from the

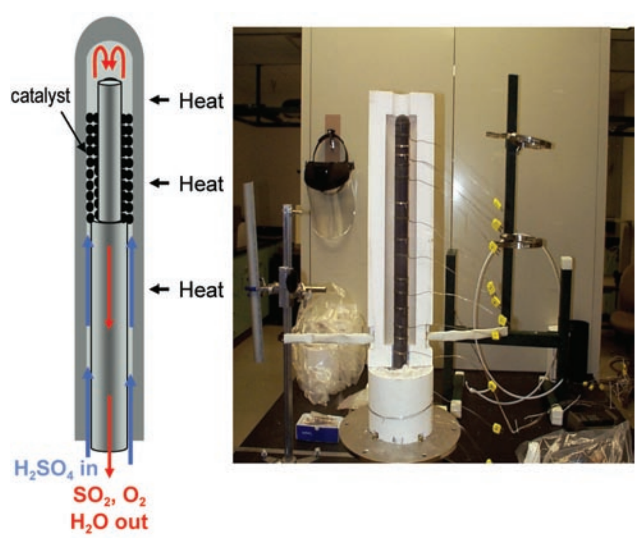

FIGURE 1. Sandia SiC Bayonet Acid Decomposer

HIx feed solution and defined the desired operating conditions for effective distillation and extraction (see Figure 2).

Bunsen Section (CEA): The CEA is developing the Bunsen section as part of a U.S. DOE–CEA INERI project (see Figure 3). The CEA has completed construction of the Bunsen section and conducted initial testing in France prior to shipment to the U.S. for integration with the other S-I sections.

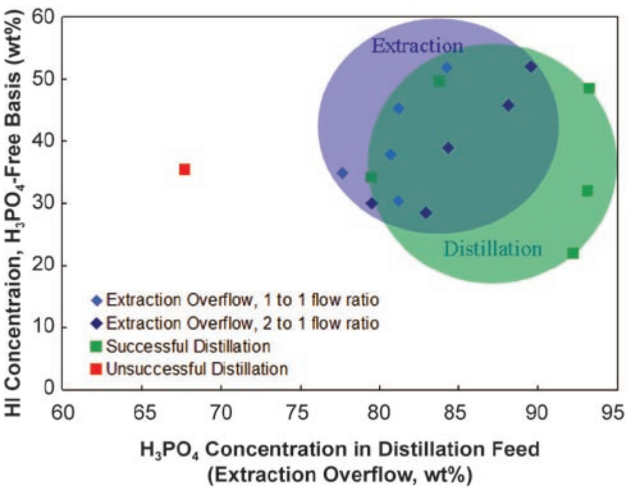

FIGURE 2. H_3PO_4 Operating Space for the Extraction and Distillation for the HI Decomposition Section

FIGURE 3. The Bunsen Section for the S-I ILS Experiment (CEA)

Conclusions

The three reaction sections of the S-I cycle have been constructed and initial testing is underway. The H_2SO_4 acid decomposition section constructed and tested at SNL has been shipped to GA in preparation for integration with the Bunsen and HI sections.

The acid decomposition section has been tested to confirm SO_2 outputs greater than 200 l/hr. Experiments at GA have defined the operating regime for H_3PO_4 extraction of HIx to allow efficient distillation and decomposition of HI. The CEA Bunsen section construction has been completed and the section will be shipped to GA in June, 2007. After completing assembly of the integrated experiment, shakedown testing will be performed in late FY 2007. Closed-loop, ILS experiments will begin in FY 2008.

Publications

1. Fred Gelbard, Robert C. Moore, Milton E. Vernon, Edward J. Parma, Dion A. Rivera, James C. Andazola, Gerald E. Naranjo, Carlos E. Velasquez, and Andrew R. Reay, “Pressurized Sulfuric Acid Decomposition Experiments for the Sulfur-Iodine Thermochemical Cycle” World Hydrogen Energy Conference, June 2006, Lyon, France.

2. Fred Gelbard, Robert C. Moore, Milton E. Vernon, Edward J. Parma, Dion A. Rivera, Howard B. J. Stone, James C. Andazola, Gerald E. Naranjo, and Paul Pickard, “Sulfuric Acid Decomposition with Heat and Mass Recovery using a Direct Contact Exchanger,” AIChE Annual Meeting, San Francisco, California, November, 2006.

3. Bunsen Wong, et.al., "Construction Material Development in Sulfur-Iodine Thermochemical Water-Splitting Process for Hydrogen Production," AIChE Spring Meeting, April 2005, Atlanta.

4. Benjamin E. Russ, et.al., "HI Decomposition-A Comparison of Reactive and Extractive Distillation Techniques for the Sulfur-Iodine Process," AIChE Spring Meeting, April 2005, Atlanta.

Presentations

Numerous presentations on the S-I project have been given by project participants including:

1. NHI Semiannual Review Meetings (November 2006, March 2007).

2. U.S. - French INERI Review Meetings (France, June 2007).