### III.13 Innovative Hydrogen Liquefaction Cycle

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- Massachusetts Institute of Technology (MIT), Cambridge, MA

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**Objectives**

Reduce the cost and improve the efficiency of hydrogen liquefaction:

- Develop and model a large capacity (50,000 kg/day or greater) hydrogen liquefaction cycle that:
  - Attain efficiencies which are a 33% improvement over present state-of-the-art systems.  
  - Significantly reduce the capital expense relative to similar capacity systems.  
- Identify and develop the key components needed for the hydrogen liquefaction cycle that are not commercially available.

**Technical Barriers**

This project addresses the following technical barriers from the Delivery section of the Fuel Cell Technologies Program Multi-Year Research, Development and Demonstration Plan:

(C) High Costs of Hydrogen Liquefaction

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**Technical Targets**

This project is designing an innovative hydrogen liquefaction cycle. The resulting design will meet the DOE Fiscal Year (FY) 2012 technical targets (nearest term target) for small-scale liquefaction (30,000 kg/day) of:

- **Installed Capital Cost:** $40 million for a 30,000 kg/day system or $1,333/kg of daily output  
- **Energy Efficiency:** 75%

The results of the first year work have shown that the GEECO innovative hydrogen liquefaction cycle design results in both significantly increased efficiency (50% better) and significantly lower capital cost.

**Accomplishments**

- Designed a practical hydrogen liquefaction cycle that significantly increases efficiencies over existing technologies.  
- Identified and designed the key component – the continuous catalytic heat exchanger (CHEX).  
- Began fabrication of testing apparatus.  
- Designed a 50,000 kg/day plant using low/no risk development components.  
- Documented a significant reduction in the total cost of hydrogen liquefaction at the complete 50,000 kg/day production level.

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**Introduction**

The purpose of this project is to produce a pilot-scale liquefaction plant that demonstrates GEECO’s ability to meet or exceed the efficiency targets set by the DOE. This plant will be used as a model to commercialize this technology for use in the distribution infrastructure of hydrogen fuel. It could also be applied to markets distributing hydrogen for industrial gas applications. Extensive modeling of plant performance will be used in the early part of the project to identify the liquefaction cycle architecture that optimizes the twin goals of increased efficiency and reduced cost. The major challenge of the project is to optimize/balance the performance (efficiency) of the plant against the cost of the plant so that the fully amortized cost of liquefying hydrogen meets the aggressive goals set by DOE.

This project will design and build a small-scale pilot plant (several hundred kg/day) that will be both a hardware demonstration and a model for scaling to larger plant sizes (>50,000 kg/day). Though
an effort will be made to use commercial or near-commercial components, key components that will need development for either a pilot- or full-scale plant will be identified. Prior to starting pilot plant fabrication, these components will be demonstrated at the appropriate scale to demonstrate sufficient performance for use in the pilot plant and the potential to achieve the performance used in modeling the full-scale plant.

**Approach**

The simplest liquefaction process is the Joule-Thomson expansion cycle. The gas to be liquefied is compressed, cooled in an after-cooler, and then undergoes isenthalpic expansion across a throttle valve. If the gas is cooled below its inversion temperature in a heat exchanger, then this expansion results in further cooling – and may result in liquid formation at the valve outlet. For hydrogen, this temperature is -95ºF. The Joule-Thomson cycle alone cannot be used for liquefaction of hydrogen without any pre-cooling of hydrogen below its inversion temperature. A modification of this cycle is sometimes used in which liquid nitrogen is used to cool the gaseous hydrogen below its inversion temperature and then Joule-Thomson expansion is used to liquefy hydrogen.

Joule-Thomson expansion is inherently inefficient as there is no work done during expansion. The industrial gas industry departed from using Joule-Thomson as a primary process used in liquefaction of atmospheric gases in the 1960s. Turbo-expanders or expansion engines are now used at most industrial gas plants to provide the necessary refrigeration for liquefaction. The expansion across a turbo-expander is ideally isentropic, or in other words, some useful work is done in expansion.

The project as originally proposed was intended to use an optimized combination of the Reverse-Brayton expansion cycle (or a modified Claude cycle) with the Joule-Thomson expansion cycle. At the beginning of the project, the scope was expanded to look at a broader range of alternate cycles. The resulting innovative cycle is shown in Figure 1. It is a once-through cycle that uses a helium-based refrigeration cycle employing Reverse-Brayton turbo-machinery. The heat removal from the hydrogen stream is performed by standard two- and three-channel heat exchangers. The baseline modeling assumes that the catalytic heat exchangers are isothermal, though additional modeling showed the added efficiency gain by using continuous catalytic heat exchangers throughout the cycle.

**Results**

The following is a summary of the major work efforts and accomplishments to date.

- Completed development and validated accuracy of CHEX numerical model.
- Through modeling, demonstrated that typical heat exchanger channel dimensions are satisfactory for pressure drop, heat exchange, and catalyst reaction rate criteria.
- Completed design of CHEX article test apparatus.
- Fabricated sensor for measuring para/ortho make-up and verified sensor performance.
- Verified through testing that the selected hydrogen and helium compressor will support the planned testing of the CHEX.
- Designed auxiliary heat exchangers.
- Designed test article “cold box.”

The results of the first year work showed that the innovative liquefier cycle design results in both significantly increased efficiency (30% better) and significantly lower capital cost. During the second year, a numerical model was developed for the catalytic heat exchanger and validated against previous testing performed on simplified catalytic heat exchangers. The continuous catalyst numeric model was created and successfully tested against known data. The model uses MATLAB for controlling inputs, outputs, and referencing thermo-physical properties, and COMSOL Multiphysics for the finite element computations. The model breaks the heat exchanger into alternating lengths of adiabatic catalytic conversion and “normal” heat exchange. This numerical model was then applied to a physical configuration that uses “typical” shell and tube heat exchanger dimensions (1/8 inch inside diameter, 5 inch length). The heat exchanger tube design is similar to the existing adiabatic catalytic heat exchanger test data examining the para to ortho transition (cold to warm). A satisfactory step size was determined so that the model acceptably simulates a continuous and simultaneous catalytic and heat exchange process. The model was then exercised by comparing it to known data and test conditions. The model results were in good agreement with the results from existing data. In addition, several parametric runs using the model confirmed the very weak pressure dependence expected for the process.

During the second year, the basic design of the test facility was completed. This is shown in Figure 1. Figure 1 also indicates the detailed design and fabrication responsibility for the test article system split between GEICO and MIT. Work has been started on the fabrication of the test apparatus to test scaled-down versions of the heat exchangers. The test apparatus para to ortho measurement devices were built and successfully tested, catalytic material acquired, and compressors to run the helium and hydrogen flow loops were identified and tested (Figure 2). In addition, sizing the auxiliary heat exchangers (recuperators for the independent hydrogen and He loops) has been
completed (Figure 3). The CHEX test article cold box has been designed to use an existing cryostat. The cold box has been sized to accept cryogenic recuperators and heat exchangers. The tubing and instrumentation will pass through the cryostat upper lid (Figure 4).

**FIGURE 1.** Helium-Based Refrigeration Cycle Employing Reverse-Brayton Turbo-Machinery and CHEX Test Apparatus

**FIGURE 2.** Helium Compressor Test Results

**FIGURE 3.** Auxiliary Heat Exchanger Design for CHEX Test Article
Conclusions and Future Directions

Based on simulations run using the model it was found that several configurations of “practical, buildable” heat exchangers can be loaded with catalytic material and effectively used in the liquefaction cycle developed during the first year. GEECO is proceeding with the 2010 work plan outlined as follows:

- Finish design and build of test apparatus
- Build adiabatic catalyst bed
- Test adiabatic catalyst bed
- Build CHEX
- Identify full-scale compressor
- Test CHEX
- Assess and report