III.11 Advanced Hydrogen Liquefaction Process

Joseph Schwartz
Praxair, Inc.
175 East Park Drive
Tonawanda, NY 14150
Phone: (716) 879-7455
E-mail: Joseph_Schwartz@Praxair.com

DOE Managers
HQ: K. Scott Weil
Phone: (202) 586-1758
E-mail: Kenneth.Weil@ee.doe.gov
GO: Paul Bakke
Phone: (720) 356-1436
E-mail: Paul.Bakke@go.doe.gov

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Fiscal Year (FY) 2011 Objectives

Develop low-cost hydrogen liquefaction systems to produce 30 and 300 tons/day:

- Improve liquefaction energy efficiency.
- Reduce liquefier capital cost.
- Integrate improved process equipment invented since last liquefier was designed.
- Continue ortho-para conversion process development.
- Integrate improved ortho-para conversion process.
- Develop optimized new liquefaction process based on new equipment and new ortho-para conversion process.

Technical Barriers

This project addresses the following technical barrier from the Delivery section (3.2.4) of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

(C) High Cost and Low Energy Efficiency of Hydrogen Liquefaction

Technical Targets

TABLE 1. Technical Targets for Liquid Hydrogen Delivery

Characteristic	Units	2012 Target	2017 Target
Small-Scale Liquefaction (30,000 kg/day)			
Installed Capital Cost	$	40M	30M
Energy Efficiency	%	75	85
Large-Scale Liquefaction (300,000 kg/day)			
Installed Capital Cost	$	130M	100M
Energy Efficiency	%	>80	87

We are addressing the capital cost and energy efficiency targets.

Capital Cost:

- Improved process design.
- Improved process equipment.

Energy Efficiency:

- Increased equipment efficiency.
- Improved process efficiency.
- Improved ortho-para conversion efficiency with a goal of reducing energy required for ortho-para conversion by at least 33%.

FY 2011 Accomplishments

- Constructed improved test unit capable of operating over a temperature range from 77 K to about 150 K.
- Developed spreadsheet model to calculate energy requirements for hydrogen liquefaction.
- Identified problems with commercial process simulation software for modeling ortho and para hydrogen and worked with project supplier to solve the problems.
- Prepared and tested new materials for improved ortho-para conversion using recipes and equipment that produce materials similar to those that could be produced in commercial quantities.
- Developed process models for existing and proposed liquefier designs.
- Showed that overall power consumption can be reduced by about 2.5% if catalyst is used in the high-temperature heat exchanger.
- Identified a process where the demonstrated performance of the improved ortho-para conversion process is sufficient to reduce total power consumption.

- Identified a process where the performance of the improved ortho-para conversion process, even when accounting for future improvement, is unlikely to be sufficient to reduce total power consumption.
- Evaluated different compressors for small and large-scale hydrogen liquefaction.
- Calculated potential overall efficiency improvement due to improved process equipment.
- Demonstrated that the combined improvements due to using more efficient compressors (4-7%), more efficient expansion turbines (2-3%), more efficient heat exchangers (<1%), an improved liquefaction process (2%), and an improved ortho-para conversion process (3-6%) can all contribute to significantly improve efficiency and reduce power requirements, but not enough to meet the DOE goal.

Introduction

Hydrogen liquefiers are highly capital intensive and have a high operating cost because they consume a significant amount of electrical power for refrigeration. There are only a few hydrogen liquefiers in the world and only six currently operating in the U.S. These plants are not built frequently, so they have not been thoroughly optimized for today's equipment. Furthermore, many of them were built when power was much less expensive than it is today, so those plants do not have optimized efficiency.

Approach

This project focused on improving liquefier efficiency and reducing overall liquefaction cost, including reducing capital cost. The project attempted to accomplish these goals using three different aspects of an integrated approach:

- Improved process design – Develop a more efficient refrigeration process including ortho-para conversion and refrigeration using available streams and equipment.
- Improved process equipment – Integrate improvements made in process equipment since the most recent liquefier design to take full advantage of the increased capabilities and improved efficiency. Project the impact of further improvements in process equipment, including novel devices currently being developed.
- Improved ortho-para conversion process – Ortho-para conversion consumes a significant amount of refrigeration energy because it requires cooling at low temperatures. Improvements in ortho-para conversion can lead to a significant reduction in power requirements.

This project built on previous work done at Praxair, some of which was part of a project funded through Edison Materials Technology Center (EMTEC). The previous project demonstrated that the improvements in ortho-para conversion were possible, but developing the complete optimized process design was beyond the scope of that project.

Results

The material screening test system used during the EMTEC project was recommissioned to perform additional material testing and test new materials. This system can test materials at the boiling point of the cooling fluid, such as liquid nitrogen. The system can test ortho-para conversion at pressures up to 400 psig and has the advantage of being a simple system that is excellent for preliminary screening of materials. Figure 1 shows this system. New materials work proceeded from the EMTEC project with new recipes and methods developed to provide samples with properties that more closely approximate those that would be obtained using commercial-scale materials manufacturing.

A pilot-scale system to conduct process testing on desired materials over a range of temperatures was built (Figure 2). This new test system is fully automated to allow for both remote control and material life testing. The system consists primarily of a series of pressure vessels, each of which houses a material bed. The annular space between the inside of the pressure vessel and the outside of the material bed contains a liquid coolant that can be pressurized. The ortho-para conversion process is conducted at very low temperatures (<150 K) and therefore a liquid coolant such as liquid nitrogen or liquid argon is required to achieve temperatures in this range. Each pressure vessel contains a vent line equipped with a back pressure control valve to allow for control of the liquid boiling pressure. By controlling the liquid boiling pressure, temperature can be controlled indirectly over a range of 77 K to 126 K using liquid nitrogen as the coolant and temperatures up to about 150 K using liquid argon.

This new test system was used to determine performance characteristics of the ortho-para conversion

FIGURE 1. Material Screening Test System

FIGURE 2. Pilot-Scale Test System

system. These results were used in the process model to determine the overall performance of a system using the improved ortho-para conversion process. These results are shown in Figures 3 and 4.

Modeling

Process simulations were developed for hydrogen liquefaction processes that used the improved ortho-para conversion process to compare those to processes that did not. Of the processes that did not use the improved ortho-para conversion process, it was found that adding ortho-para conversion catalyst to the high-temperature heat exchanger reduced overall energy consumption by about 2.5%.

Several different processes have been conceived that use the improved ortho-para conversion process. The modeling portion of the program compared these processes to hydrogen liquefaction processes with standard ortho-para conversion. Concept α and Concept β were evaluated. Figures 3 and 4 show the results.

The point in the center is the target based on ortho-para performance meeting the base case power target. The y-axis represents the power required for hydrogen liquefaction as a percentage of the base case power target. The x-axis shows ortho-para performance as a percentage of demonstrated performance in the laboratory. The target is based on demonstrated performance (100% of current ortho-para performance) meeting the base case power target. In this case, about 150% of the current demonstrated performance is required to meet the base case power target. Although there has been steady improvement in demonstrated performance throughout the project, marginal gains are lower than they were earlier in the project. It is unlikely that Concept α will meet the target required to be an economically viable process.

Figure 4 shows the same results for Concept β. In this case, 100% of the current demonstrated performance

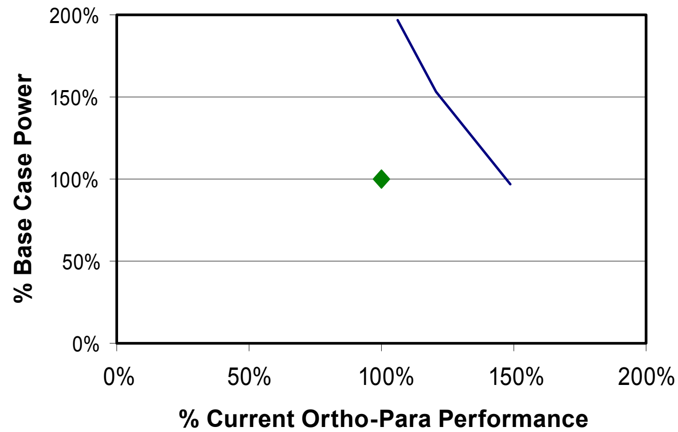

FIGURE 3. Ortho-Para Performance for Concept α

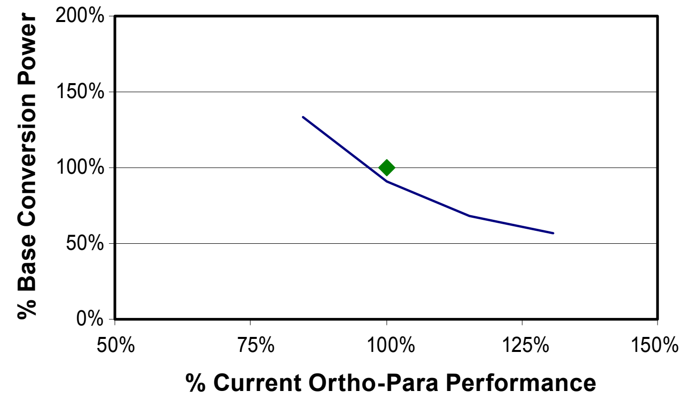

FIGURE 4. Ortho-Para Performance for Concept β

results in total power consumption below the base case power target. Concept β met the target required to be an economically viable process on an energy basis.

Process Equipment

Another way to improve the efficiency of hydrogen liquefaction is to improve the efficiency of the equipment used in the process. This includes compressors, turbines, and heat exchangers. Compression accounts for most of the power consumed by hydrogen liquefaction, so improvements in compressor efficiency can provide a significant benefit as discussed in the following. The impact of improving turbine or expander efficiency is small, but can reduce total power by about 2%. The impact of improving heat exchangers by reducing the pinch between hot and cold streams is even smaller than the impact of improving turbines.

Several different types of hydrogen compressor were evaluated. Reciprocating, screw, and centrifugal compressors were identified as being the most likely to apply to large-scale liquefaction. Metal hydride compressors were determined to be too small. Guided rotor and ionic liquid compressors were determined to be in the developmental stage. Diaphragm compressors were unlikely to meet the capital cost target. Axial and shock wave compressors were determined to be a poor fit for a low molecular weight gas like hydrogen.

Table 2 shows some characteristics of the different types of compressor considered.

Screw compressors were expected to offer the lowest capital cost, but were also likely to have smaller capacity and lower efficiency. Reciprocating compressors offer larger size and higher efficiency, but at a higher initial cost and higher maintenance cost. Centrifugal compressors are expected to offer higher flow, lower maintenance cost, and similar efficiency compared to reciprocating machines.

Figure 5 shows the potential impact of improving compressor efficiency on the power consumption of the entire process assuming a base case with 80% adiabatic efficiency for all compressors. The main recycle compressor, which has three stages, consumes much more power than the low pressure recycle compressor, so improving its efficiency has a bigger impact on the total power. Improving the efficiency of each stage from 80 to 89%, the upper limit expected in Table 2, reduces total power by about 10%. Using an adiabatic efficiency value of 85%, closer to the middle of the range, reduces total power by about 6%.

Total Efficiency Improvement

The gains from each potential improvement discussed are additive; one does not reduce the ability to implement another. The total efficiency improvement is shown in the following:

TABLE 2. Hydrogen Compressor Options

Technology	Flow Range	First Cost	Relative Maintenance	Adiabatic Efficiency	Number of Units required	
	CFM				30 tpd	300 tpd
Screw	**20,000**	**Low**	**Medium**	**70 to 75%**	**2**	**20**
Reciprocating	**40,000**	**High**	**High**	**83 to 89%**	**1**	**10**
Centrifugal	**80,000**	**Medium**	**Low**	**80 to 89%**	**1**	**5**

tpd – tons per day

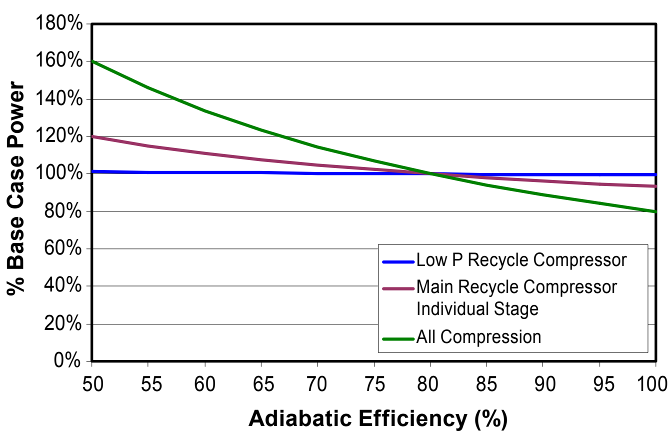

FIGURE 5. Effect of Compressor Efficiency on Total Power

Factor	Total Efficiency Improvement
Compressor Efficiency	6%
Turbine Efficiency	2%
Heat Exchanger Efficiency	<1%
Improved Liquefaction Process	2%
Improved Ortho-Para Conversion Process	5%

Total Efficiency Improvement	15%

The total efficiency improvement possible through using all of these methods is only 15%, less than the 20% goal of the project. Furthermore, each of these improvements potentially increases capital cost, making the goal of a 20% reduction in capital very unlikely to be possible for the highest efficiency process. Because of this result, the project was stopped after Phase II before completing the capital cost estimate.

Conclusions and Future Directions

- Improvements in overall process efficiency are possible through improved process equipment, improved process design, and an improved ortho-para conversion process.
- The total improvement is less than the project goal of 20%.
- Improvements are likely to add capital cost, making the other project goal of a 20% reduction in capital very unlikely.
- Ortho-para conversion performance was measured using laboratory and pilot reactors.
- The demonstrated performance is sufficient for at least one identified process concept to show reduced power cost when compared to hydrogen liquefaction processes using conventional ortho-para conversion.
- The impact of improved ortho-para conversion can be significant, but ortho-para conversion uses only about 20-25% of the total liquefaction power.
- Most of the energy used in liquefaction is for gas compression. Improvements in hydrogen compression will have a significant impact on overall liquefier efficiency.

FY 2011 Publications/Presentations

1. DOE Annual Hydrogen Review Meeting

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