# V.D.2 Cost-Effective High-Efficiency Advanced Reforming Module (CHARM)

Michael Leshchiner (Primary Contact), Darryl Pollica, Christian Stanley, Changsik Kim, Atul Sharma, Kara Gilhooly

Nuvera Fuel Cells 129 Concord Rd., Building 1 Billerica, MA 01821 Phone: (617) 245-7614

E-mail: mleshchiner@nuvera.com

DOE Technology Development Manager:

Kathi Epping Martin

Phone: (202) 586-7425; Fax: (202) 586-9811

E-mail: Kathi.Epping@ee.doe.gov

DOE Project Officer: David Peterson Phone: (303) 275-4956; Fax: (303) 275-4788 E-mail: David.Peterson@go.doe.gov

Technical Advisor: John Kopasz Phone: (630) 252-7531; Fax: (630) 972-4405

E-mail: Kopasz@anl.gov

Contract Number: DE-FC36-02AL67618

Start Date: January 1, 2002

Projected End Date: September 30, 2008

# **Objectives**

Develop an advanced reforming module for stationary applications:

- Develop a 1,000 scfh (56 kg/day) fuel processor with low product life-cycle cost. Minimize capital, operating and maintenance costs over a 40,000-hour life.
- Develop a scaleable technology from 1,000 to 10,000 scfh (56 to 560 kg/day).
- Achieve a lowest life-cycle cost, namely a costeffective balance between efficiency and durability.
- Perform a lifetime assessment through modeling of stresses and through accelerated aging of the fuel processor.
- Demonstrate fuel processor durability by achieving 5,000 hours and 250 cycles.
- Validate performance of a 1,000 scfh fuel processor by a third party: Argonne National Laboratory.
- Apply Design for Manufacturing and Assembly (DFMA®) principles to guide cost comparisons and the ultimate mechanical specification of the hardware.

#### **Technical Barriers**

This project addresses the following technical barriers to developing a natural gas (NG) fuel processing system from the Fuel Cells, section 3.4 and Hydrogen Production, section 3.1of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan [1]:

Fuel Cells:

(A) Durability

Hydrogen Production:

- (A) Reformer Capital Costs
- (B) Reformer Manufacturing
- (C) Operation and Maintenance (O&M)
- (E) Greenhouse Gas Emissions
- (F) Control and Safety

## **Technical Targets**

Status versus the DOE technical targets for fuel processors [1] is presented in Table 1. The H2A Forecourt Production model [2] was used for the cost modeling.

# **Accomplishments**

- Performed computational fluid dynamics-finite elements analysis (CFD-FEA) coupling and predicted the reformer's capability for over 1,000 cycles.
- Selected Inconel 625 material for reformer tube caps to provide the lowest life-cycle cost after considering capital, operating, and maintenance costs over the 40,000-hour lifetime.
- Collaborated with strategic catalyst partner in development of highly durable base metal steam reforming catalyst.
- Found new supplier and implemented a new smaller and less expensive superheater.
- Applied DFMA® principles and reduced the fuel processor cost by over 20%.
- Adopted new desulfurization material with higher sulfur capacity and reduced maintenance cost.
- Successfully implemented a monolithic water-gas shift reactor for startup time reduction.



TABLE 1. Status of CHARM versus DOE Targets

Technical Targets: Stationary Fuel Processors (equivalent to 5-250 kW) to Generate Hydrogen-Containing Fuel Gas				
Characteristic	Units	2005 Status	2011 Target	CHARM Status
Cost	\$/kWe	1,000	220	1,450
Cold start-up to rated power at -20°C ambient	min	<90	<30	120
Transient response time (for 10% to 90% power)	min	<5	1	15
Durability	hours	20,000	40,000	Designed for 40,000 Validation is ongoing
Survivability (min & max ambient temperature)	°C °C	-25 +40	-35 +40	-20 (Managed by enclosure heater) +40 (Verified)
CO content in product stream Steady-State Transient	ppm ppm	10 100	1 25	Verified via 3 <sup>rd</sup> Party Analysis <1 <6
H <sub>2</sub> S content in product stream	ppbv (dry)	<10	<4	N.D. <sup>1</sup>
NH <sub>3</sub> content in product stream	ppm	<1	< 0.1	N.D. <sup>2</sup>
Select Technical Targets: Distributed Production of Hydrogen from Natural Gas				
Characteristic	Units	2010 Target	2015 Target	Application of CHARM
Production Energy Efficiency	% (LHV)	72.0	75.0	64.4%-68.4% current system Novel high efficiency system concept is in design phase
Production Unit Capital Cost (Uninstalled)	\$	900K	580K	\$2.4M (at 1,550 kg/day capacity), Storage - \$1M
Total Hydrogen Cost	\$/gge H <sub>2</sub>	2.50	2.00	5.94 (automotive), 3.16 (materials handling)

<sup>&</sup>lt;sup>1</sup>Instrumentation detection level of 5.0 ppby; <sup>2</sup>Instrumentation detection level of 1.0 ppm; LHV - lower heating value; N.D. - not detected.

#### Introduction

Nuvera Fuel Cells has been advancing steam reformer technology for over 10 years. Cost reduction and durability improvement is required to evolve the commercialization of distributed hydrogen generation appliances and stationary fuel cell systems. Accordingly, Nuvera is developing a modular reformer with a key emphasis on life-cycle cost reduction via optimization of the burner-reformer assembly.

# **Approach**

The objective of the CHARM project is to develop a modular steam reformer while maintaining balance between cost, efficiency, durability, manufacturability and environmental standards. A low-pressure steam reformer will be produced to address the widest range of applications with the lowest risk. The reformer will be designed for long life and low capital cost in accord with DFMA® principles, high efficiency, and respect for strict emissions regulations. The intent is to develop a steam reformer technology that is scaleable from 500 to 10,000 scfh of hydrogen.

The most significant stigma associated with small-scale reformers is durability. Although industrial-scale reformers have been in use for decades, they rarely see more than a few thermal cycles per year. In order for a

small-scale reformer to survive frequent thermal cycles, the stresses must be reduced to exceptionally low levels and interaction between creep, corrosion and cyclic fatigue must be minimized.

#### Results

The fuel processor design philosophy incorporated the functional requirements, design of the reforming process, inputs from CFD-FEA modeling and DFMA® principles to influence the mechanical design, a stress and failure mode analysis, material selection, and a lifecycle cost analysis to arrive at the final detailed design.

#### Reformer Design

Steam reforming is a strongly endothermic reaction, often requiring temperatures exceeding 900-1,000°C on the reformer walls. Standard practice for design of large industrial systems is a tubular steam reformer surrounded by an "impingement" style burner module. These reformers are designed with thick walls and run continuously with very little or no cycling. Burner fuelair mixture is fired directly onto reformer tubes. That creates localized hot spots on the tube's metal surface leading to formation of local stress points. As a result creep, which is a gradual increase in plastic strain with time at constant load, is a dominant failure mechanism.

Unfortunately, for smaller scale hydrogen generators, such as PowerTap or even automotive refueling stations, a practical reality is that the hydrogen consumption profile can be somewhat irregular, necessitating a need for more frequent cycling. Depending on the application, weekly or even daily start-stop might be dictated by economical or practical considerations.

Nuvera's approach was to design a "hanging" tube reformer to minimize stress. However, a problem of creep, corrosion and cyclic fatigue interaction still remains. When cycled, thermal stress due to local hot spots and resulting plastic deformation at operating temperatures will cause stress in metal when cooled off. Each heat cycle can result in finite amount of plastic strain and consequent fatigue failure in a relatively small number of cycles.

To address the problem, extensive analyses of temperature distribution in metal tubes as well as testing of different materials in actual reformer operation were performed. Based on the life-cycle cost analysis, Inconel 625 caps were adopted as a design feature of the reformer. Inconel has far superior high temperature properties compared to stainless steels and cost impact is minimized by protecting only 6" of tube length in the hottest temperature zone, where direct flame impingement might occur.

#### **Durability Assessment**

Durability prediction requires knowledge of thermal stresses and/or strains in the part that is being designed or analyzed. Coupling of CFD-FEA software packages has proven to be an extremely useful tool, especially for cycling operation. Nuvera worked closely with a manufacturer of FEA software to develop a coupling method. The approach was to transfer reformer tubes boundary conditions: loads, surface temperature, heat flux, pressure, etc., provided by CFD into an FEA model. Results of the combined model for the CHARM reformer with some conservative assumptions are shown in Figure 1.

Expected maximum strain on the reformer tubes due to temperature gradients in the burner is on the order of 0.5%. Therefore cycling capability of the CHARM fuel processor is estimated using some available material properties data for Inconel 625. It is projected the reformer will last for over 1,000 cycles. To complement this model, accelerated fuel processor cycle testing at very high pressures will be performed on the durability test stand in the third quarter of 2008.

#### Steam Reformer Catalyst Development

A combination of reformer cycling and high temperatures also impose harsh conditions on steam reforming catalysts. Due to cyclic expansions and

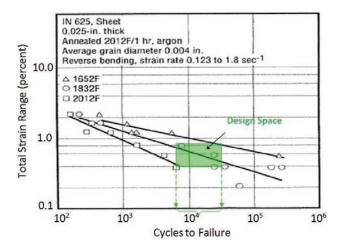


FIGURE 1. Estimate of cycling limitations based on the strain in reformer tubes. Inconel 625 properties are from Aerospace Structural Metals Handbook [3]

contractions pellets can be mechanically crushed resulting in substantial catalyst attrition. In addition, activity loss can result from catalyst sintering, oxidation, poisoning, coking, etc. In an effort to reduce the life-cycle cost of the CHARM reformer, Nuvera in collaboration with a strategic catalyst partner worked on the development of a higher strength and improved hydrothermal stability steam reforming catalyst. Nuvera developed a detailed test plan designed to mimic worst case cycling conditions. Despite slightly lower fresh activity, the new catalyst demonstrated superior activity and hydrothermal stability after just one steaming cycle.

In addition, to improve catalyst mechanical strength a tablet shape was adopted instead of spheres resulting in average crush strength increase from 20 to 35 lbs dead weight load. Further thermal shock testing, which entailed heating the catalyst to 1,000°C and then quenching it in cold water, resulted in no breakage for new catalyst compared to 44% breakage for the original catalyst.

# New Superheater Implementation

Although the original superheater performed reasonably well, it had several drawbacks. Specifically, it was too expensive and very large, hence long start-up and rather complicated and bulky plumbing and packaging. Thus the team identified a new supplier, which agreed to work with Nuvera on design of the new heat exchanger. The result is the new superheater satisfied performance specifications with much smaller size and lower cost.

A new superheater was incorporated into the CHARM system near the 1,100 hours mark and has been running successfully since then. Currently the fuel processor test stand is being run for a durability target of 5,000 hours and 250 cycles, the remainder of which will

happen in counter flow. It is expected that by the end of this deliverable the superheater will have accumulated 3,900 hours and close to 200 thermal cycles.

#### **Desulfurization Material**

NG desulfurization is currently one of the most expensive maintenance items in a hydrogen generation plant. Historically, activated carbon and a wide variety of zeolites have been used for fuel desulfurization. However, the main drawbacks associated with mechanical adsorption (or physisorption) are low overall sulfur capacity and ability to adsorb some but not all types of sulfuric species. Other undesirable effects of mechanical adsorbents include desorption at elevated temperature and easy adsorption of higher hydrocarbons and aromatics.

Desulfurization materials developed by our partner and recently adopted by Nuvera alleviate all of the above mentioned limitations. New technology is based on a selective chemisorption principle, where sulfur species adhere to a surface of adsorbent through the formation of a chemical bond. To cover the entire range of sulfur containing species two metal oxide based desulfurization materials for NG were developed and are used in a mixed desulfurizer bed. The first material is for separation of H<sub>2</sub>S, COS and mercaptans, and second layer is for tetrahydrothiophene, dimethylsulfide and disulfides. Unlike conventional materials, the sulfur removing capacity of the first material increases with increasing temperature.

Nuvera is continuously assessing options for desulfurization. The life-cycle cost of the hydrogen desulfurization method will also be screened before the project completion.

## Hours and Cycles Testing

One deliverable of the current project is to achieve 5,000 hrs and 250 cycles with the CHARM system. Progress towards durability milestones is shown in Figure 2. Until now the system ran for 4,472 hours and 169 cycles. The CHARM reformer will be cycled more frequently during the last month of the project, according to the specially developed cycling protocol.

### **Conclusions and Future Directions**

In the forthcoming months Nuvera will complete the following activities:

 Complete validation of the co-flow and counterflow reformer configurations, including assessment of trade-offs between durability and efficiency for minimization of the hydrogen generator life-cycle cost.

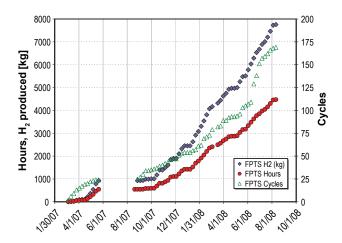


FIGURE 2. Fuel Processor Test Stand Running Summary

- Conclude accelerated fuel processor testing for investigation of creep-fatigue interaction and investigate impact of cycling on expected fuel processor life.
- Assist Argonne National Laboratory in validation of performance of the 1,000 scfh system.
- Demonstrate fuel processor durability with 5,000 hours and 250 cycles.
- Complete conceptual design of scalable and cycleable 10,000 scfh fuel processor.
- Explore novel high efficiency system concept potentially affording cost of hydrogen reduction to below \$3.00/gasoline gallon equivalent delivered at the pump (based on DOE assumptions in 2007 Multi-Year Research, Development and Demonstration Plan).
- Develop novel steam reformer concept with optimized heat flux profile, potentially resulting in fuel processor lifetime exceeding the already aggressive CHARM target of 40,000 hours with cycling.

It is also worth mentioning, that in addition to plans for commercialization of the CHARM technology in the material handling market, Nuvera is now considering developing a system for use in vehicular refueling applications. In concert with our majority owner Hess, Nuvera is in discussion of actualizing such projects in the 2010 time frame.

# **FY 2008 Publications/Presentations**

1. A. Sharma, D. Pollica, B. Rogers, L. Clawson, T. Holmes, "Low Life Cycle Cost Natural Gas Reforming for Fuel Cell Forklift Market Application", American Chemical Society National Meeting, August 2007, Boston, MA.

#### References

- 1. Hydrogen, Fuel Cells & Infrastructure Technologies Program Multi-Year Research, Development, and Demonstration Plan: Planned Program Activities for 2005-2015, http://www.eere.energy.gov/hydrogenandfuelcells/ mypp/.
- **2.** H2A Forecourt Hydrogen Production Model Users Guide, http://www.hydrogen.energy.gov/h2a\_production.html.
- **3.** Aerospace Structural Metals Handbook, Volume 5, 1999 Edition, CINDAS/USAF CRDA Handbook Operations, Purdue University.