# Fast Start Reformer Components

*Greg A. Whyatt*
*Pacific Northwest National Laboratory*
*P.O. Box 999, MS K6-24*
*Richland, WA 99352*
*Phone: (509) 376-0011; Fax: (509) 376-3108; E-mail: greg.whyatt@pnl.gov*

*April D. McMillan*
*Oak Ridge National Laboratory*
*1 Bethel Valley Road*
*Oak Ridge, TN 37831-6087*
*Phone: (865) 241-4554; Fax: (865) 576-8424; E-mail: mcmillanad@ornl.gov*

*Michael A. Inbody*
*Los Alamos National Laboratory*
*P.O. Box 1663, MS J-579*
*Los Alamos, NM 87545*
*Phone: (505) 665-7853; Fax: (505) 665-9507; E-mail: inbody@lanl.gov*

*DOE Technology Development Manager: Nancy Garland*
*Phone: (202) 586-5673; Fax: (202) 586-9811; E-mail: Nancy.Garland@ee.doe.gov*

## Objectives

- Support the development of the Argonne National Laboratory (ANL) FASTER fuel processor project with the expertise developed at PNNL, ORNL, and LANL.
- Design and fabricate a highly effective recuperator with low pressure drop to recuperate incoming steam and air with autothermal reformer (ATR) reformate.
- Design and fabricate a microchannel mixer to uniformly add air to reformate ahead of the water gas shift (WGS) catalyst to accelerate heating of the WGS catalyst.
- Design and fabricate a lightweight heat exchanger that meets system constraints and operational performance requirements.
- Optimize heat exchanger assembly for insertion into the FASTER system.
- Demonstrate a heat duty of at least 1 kW with a pressure drop of <0.1 psi.
- Investigate and develop a preferential oxidation (PrOx) reactor component design for integration into the FASTER fuel processor.
- Deliver these components as part of a compact assembly for installation into the FASTER reactor system.

## Technical Barriers

This project addresses the following technical barriers from the Fuel Cells section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year R,D&D Plan:

- I. Fuel Processor Start-up/Transient Operation
- L. Hydrogen Purification/Carbon Monoxide Cleanup
- M. Fuel Processor System Integration and Efficiency

## Approach

- Design and fabricate a highly effective, low pressure drop recuperator.
- Design and fabricate micro-channel mixer for introducing air into reformate during startup.
- Deliver a compact recuperator/mixer assembly for installation into the FASTER experimental system.
- Design a prototype heat exchanger (HX) meeting original specifications for Tier 1; fabricate a test rig to validate HX design.
- Test prototype HX under operational constraints and measure all inlets and outlets, i.e., pressure, temperature, flow, etc.
- Analyze the data to determine heat duty as a function of water flow rate at a given inlet gas temperature.
- Identify and measure the performance of PrOx catalysts for use in a PrOx reactor component.
- Collaborate on PrOx component design and fabricate PrOx components.
- Measure PrOx component response to simulated startup conditions and events.

## Accomplishments

- Design work on recuperator, mixer, and assembly complete.
- Both microchannel units are currently being diffusion bonded.
- Modeled design concept to estimate pressure drop and heat transfer of the graphite foam heat exchanger at all sizes specified by project lead, ANL.
- Designed a prototype heat exchanger approximately 3-inch inner diameter and 5-inch outer diameter with a machined channel into which a cooling jacket was press fit.
- Tested prototype and met or exceeded all specifications. Improvements in design of the prototype HX were made and a second iteration prototype was fabricated.
- Delivered all of Tier 1 heat exchangers at the end of June, 2003. Fabrication of Tier 2 heat exchangers is on schedule; anticipated delivery date is the end of July, 2003.
- Measured kinetic parameters of PrOx catalysts for reactor design.
- Investigated performance of PrOx reactor under simulated startup conditions.
- Obtained catalyst substrates for unique PrOx component design and started fabrication.

## Future Directions

- Complete fabrication and deliver assembly to ANL for inclusion in FASTER prototype.
- Evaluate performance as part of the FASTER experimental effort.
- Test Tier 1 HX at higher gas inlet temperatures; measure heat duty as a function of water flow rate at higher inlet temperature(s).
- Evaluate different avenues for further reducing the weight of the graphite foam heat exchanger assembly while maintaining heat transfer and pressure drop.
- Verify design performance in multi-stage PrOx experiments.

---

## Introduction

The major technical challenge for the development of a successful onboard fuel processor for fuel cell systems is the 2010 technical target for startup in less than 30 seconds. Overcoming this technical challenge is the objective of the FASTER (Feasibility of Acceptable Start Time Experimental Reactor) project led by Argonne National Laboratory (ANL), whose goal is to design, build, and test an experimental fuel processor to demonstrate concepts for rapid startup. The FASTER project is a collaborative effort which includes the contributions of three additional national laboratories that have developed significant expertise for the DOE Hydrogen, Fuel Cells and Infrastructure Technologies Program. Pacific Northwest National Laboratory (PNNL) has expertise in the development of microchannel heat exchanger technology for reforming components, Oak Ridge National Laboratory (ORNL) has expertise in the development of graphite foam heat exchanger technology, and Los Alamos National Laboratory (LANL) has developed preferential oxidation (PrOx) technology for the removal of carbon monoxide. The contributions of these three national laboratories are reported here.

**Recuperator/Mixer Assembly** - Between the outlet of the autothermal reformer (ATR) and the entrance to the water gas shift (WGS) reactor is an assembly being provided by PNNL. The assembly is designed to fit within the FASTER geometry and contains a recuperator and a mixer. The recuperator will recover heat from the ATR reformate to preheat the air and steam feed to the ATR. This cools the reformate to a temperature suitable for the WGS catalyst. The effective recovery of heat from the hot reformate allows the system to be energy efficient. The micro-channel mixer is positioned at the reformate outlet of the recuperator. During startup, it will mix air into the reformate, causing oxidation of reformate on the WGS catalyst to accelerate heating of the WGS catalyst. A uniform reformate/air mixture is required to prevent local hot or cold spots on the WGS catalyst during startup.

**Graphite Foam Heat Exchangers** – Within the WGS and PrOx reactors, temperature control is paramount to efficient operation. Heat exchangers (HXs) must be lightweight and capable of handling a heat duty of 1 kW at low pressure drops. These heat exchangers must meet the operational and physical constraints of the FASTER system design for effective catalysis downstream. ORNL is providing these HXs to facilitate fast start.

**PrOx** - The preferential oxidation (PrOx) reactor component is typically the last component of the fuel processor, and its function is to control the outlet carbon monoxide to levels below 10 ppm. LANL is using its experience in PrOx research and design to develop a PrOx reactor to enable fast start. This includes reducing the thermal mass of the catalysts and components in contact with the flow, identifying a catalyst with a low light-off temperature for CO oxidation that also maintains CO selectivity over a wide temperature range, and identifying strategies to oxidize high concentrations of CO at startup before the WGS reactors become operational. The goal is to incorporate these features into PrOx reactor components to be fabricated and delivered for assembly into the FASTER fuel processor.

## Approach

**Recuperator/Mixer Assembly** - The micro-channel components are made by etching metal sheets, stacking them in sequence, and then sealing contacting surfaces by diffusion bonding. The recuperator and portions in contact with hot reformate are being fabricated in Inconel 600. Within the recuperator, the fluids exchange heat in a series of counterflow, laminar channels. Lower temperature components including the mixer are being fabricated in 316L stainless steel. The microchannel mixer divides the reformate and air into numerous channels, interleaves them, and then allows them to exit the downstream face of the panel. Actual mixing occurs as the flows exit the closely spaced interleaved channels. Features are included to maintain uniform mixing despite temperature differences between the fluids.

**Graphite Foam Heat Exchangers** - The approach to designing graphite foam heat exchangers capable of handling the heat duty and pressure drop requirements was to first use computational fluid dynamics (CFD) to model the foam under these constraints. From the model results, a corrugated design was fabricated. The model of the graphite foam capabilities was validated experimentally. Copper was chosen as the coolant jacket material due to its high bulk thermal conductivity and its ability to spread the heat in all directions, aiding in heat transfer. Initially, the inlet and outlet for the cooling water for the jacket were on the same side of the jacket. Infra-red (IR) imaging showed symmetric cooling about the jacket; however, this IR imaging also showed that the coolant two-phase flow could be improved. Thus, in the second iterative prototype, the flow is now bi-furcated, with the inlet and outlet 180° from one another (Figure 1). This should increase the two-phase flow and therefore enhance heat transfer in the assembly.

**PrOx -** The approach to developing a PrOx reactor component has been to first identify PrOx catalysts that have desirable characteristics for startup performance. Laboratory PrOx reactor hardware is used to measure the kinetic parameters of the catalysts under simulated steady-state operating conditions and to measure the PrOx component response to simulated startup conditions and events. Kinetic parameters are supplied to ANL for use in the fuel processor design calculations. Based on the design, catalyst substrates are obtained, catalyzed, and fabricated into the reactor components for the FASTER fuel processor.

## Results

**Recuperator/Mixer Assembly** – The thermal performance requirements for the recuperator are shown in Figure 2. In addition, a number of spatial requirements were placed on the design to allow the assembly to connect to the ATR, match desired connection locations, and fit within the 6-inch diameter core of the FASTER reactor system. The recuperator and mixer are designed and fabrication is under way. A key feature of the recuperator is turndown performance. The recuperator is predicted to maintain >85% effectiveness as the flow is decreased from 100% of design capacity to 10% of capacity. This feature prevents excessively high reformate temperatures (which could damage the WGS catalyst) under the normal range of operating rates. The weight of the components is estimated to be ~800 g for the recuperator, ~176 g for the mixer and ~1100 g for the total assembly. The assembly to be delivered to ANL is shown in Figure 3.

**Figure 1.** Tier 1 Graphite Foam Heat Exchangers for FASTER

**Graphite Foam Heat Exchangers -** Tests indicate that the heat duty is relatively constant for a given inlet gas temperature. Further, tests reveal that heat transfer through the graphite foam may be insensitive to water flow (Figure 4). This will allow for more flexibility in the control of the overall FASTER system; it may be possible to maintain critical

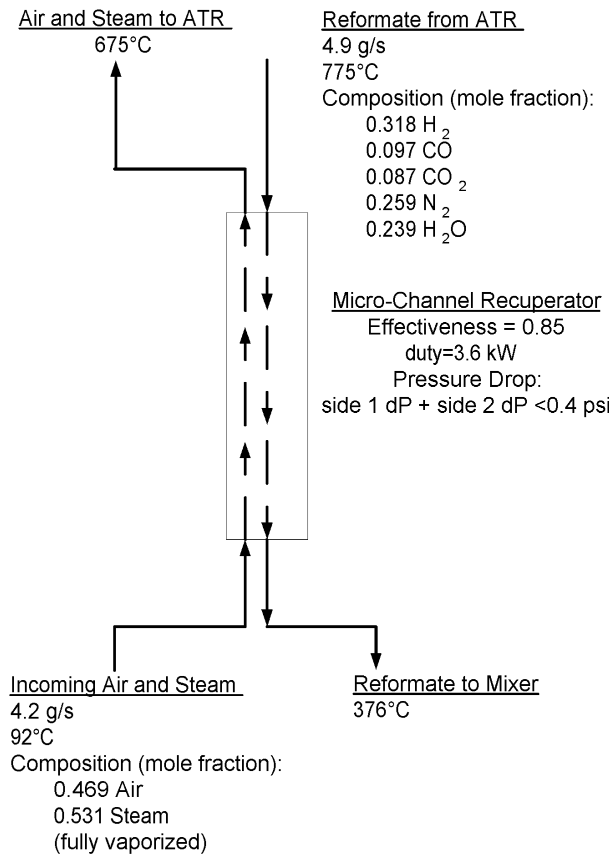


**Figure 2.** Design Requirements for Recuperator Thermal Performance

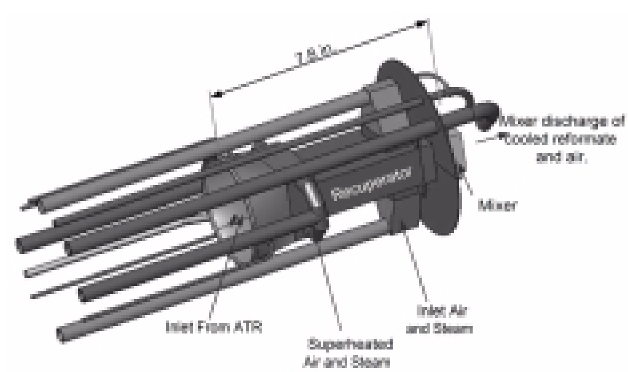


**Figure 3.** Recuperator/Mixer Assembly for Incorporation into FASTER ATR

temperature regimes for each catalyst while using supply water at low flow rates. Feedback may be used to more efficiently control the water flow rates to increase the overall system efficiency.

**PrOx -** PrOx catalysts were obtained from various manufacturers on monolith, ceramic foam, and metal foam substrates. Based on preliminary testing, a catalyst was identified with low light-off temperature and good selectivity. The performance of the catalyst was measured over the range of expected operating conditions, and the data was provided to ANL for their fuel processor model and evaluation of kinetic parameters.

A three-stage PrOx design was chosen to permit greater flexibility in removing higher CO concentrations (>1%). The first and second stages will operate to bring the outlet CO to 2000 to 1000 ppm. The third stage will operate to bring the outlet CO to <10 ppm with a small temperature rise to avoid the reverse WGS reaction. Foam substrates were obtained for the unique geometry of the PrOx component dictated in the fuel processor design.

Single-stage startup experiments were conducted to investigate the startup response of a PrOx catalyst monolith from room temperature. For these experiments, a primary gas flow consisting of 46.4% $H_2$, 32.9% $N_2$, 19.0% $CO_2$, and 1.2% CO at a temperature of 20ºC was established at a gas hourly space velocity (GHSV) ≈31,000 $hr^{-1}$ through a 600cell/$in^2$, 3-inch diameter, 5-inch long PrOx catalyst monolith. Air injection was started, and the resulting outlet CO concentration and monolith temperature profile were measured as a function of time. Figures 5 and 6 show the outlet CO concentration response and monolith temperature response as a function of time, respectively. The outlet CO decreases rapidly to its steady-state values in less than 30 seconds, while the temperature response takes longer to reach steady-state. The low

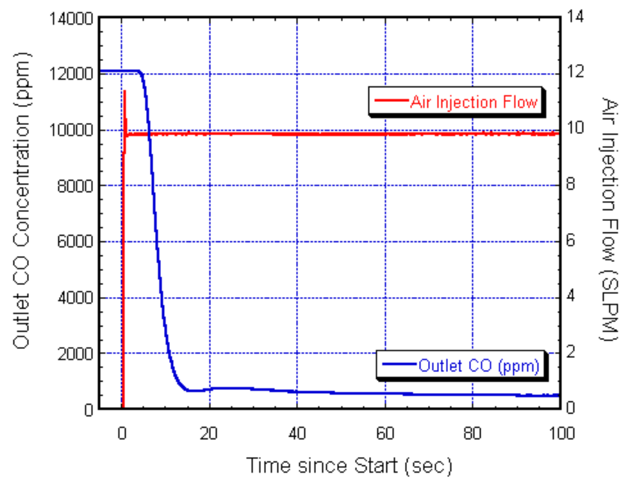


**Figure 5.** PrOx Outlet CO Response as a Function of Time

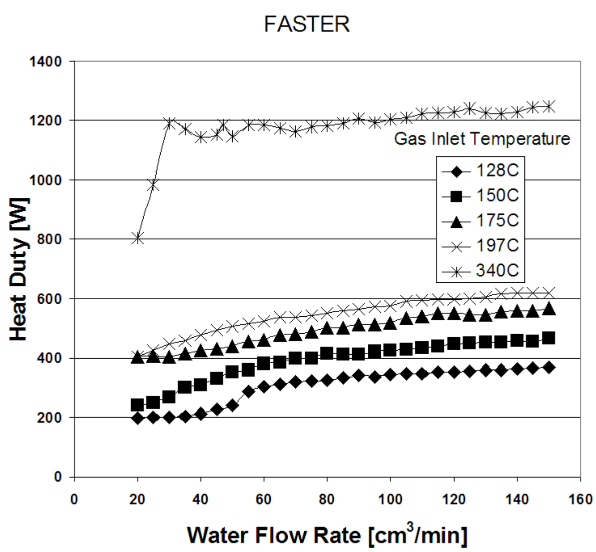


**Figure 4.** Performance of Tier 1 Graphite Foam Heat Exchanger

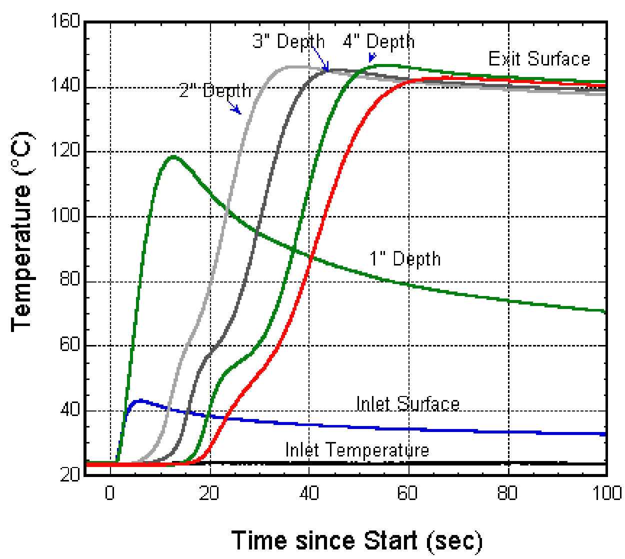


**Figure 6.** PrOx Monolith Temperature Response as a Function of Time

light-off temperature of the catalyst is evident from the response. Similar responses were obtained for both higher (2.2%) and lower (2350 ppm) inlet CO concentration. Based on these experiments, fast startup of the PrOx reactor component appears feasible.

## Conclusions

- A compact, lightweight and efficient microchannel recuperator meeting the desired thermal performance, pressure drop and spatial requirements is possible.
- A microchannel mixer provides a method to uniformly mix air into reformate during startup to accelerate heating of the WGS catalyst.
- Graphite foam has met or exceeded specifications for performance in the FASTER system.
- Graphite foam heat exchangers appear to be relatively insensitive to water flow, especially at higher gas inlet temperatures.
- Further decreases in weight of the heat exchanger assembly may be possible.
- PrOx catalysts have been identified and characterized, and a three-stage PrOx design is being fabricated.
- Outlet CO concentrations could be reduced to steady-state values in less than 30 seconds in PrOx single-stage startup experiments.

## FY 2003 Publications/Presentations

1. Whyatt, G. A., Poster: Micro-Channel Recuperator and Mixer for FASTER (Feasibility of Acceptable Start Time Experimental Reactor) Autothermal Reformer. Presented at the Hydrogen, Fuel Cells and Infrastructure Technologies Program 2003 Merit Review and Peer Evaluation Meeting, May 19-22, 2003, Berkeley CA.

2. Inbody, M. A., R. L. Borup, et al. (2002). Transient Control of Carbon Monoxide with Staged PrOx Reactors. 224th National Meeting of the American Chemical Society, Boston, MA, American Chemical Society.

3. Inbody, M. A., R. L. Borup, et al. (2002). Transient PrOx Carbon Monoxide Measurement, Control and Optimization. 2002 Fuel Cell Seminar.

4. McMillan, A.D., Romanonski, G.R., Klett, J.W., Armstrong, T., and Stinton, D., Annular Graphite Foam Heat Exchangers Were Developed for FASTER. Presented at the Hydrogen, Fuel Cells and Infrastructure Technologies Program 2003 Merit Review and Peer Evaluation Meeting, May 19-22, 2003, Berkeley, CA.

## Special Recognitions & Awards/Patents Issued

1. Patent Application: Greg A. Whyatt, #60-471,130, Microchannel Mixing Device, 5/16/03