Reformate Cleanup Technology

Michael A. Inbody, Rodney L. Borup, and José I. Tafoya
Fuel Cell Team
Los Alamos National Laboratory
Los Alamos, NM 87545

Los Alamos NATIONAL LABORATORY

Fuel Cell Program

Technical Objectives

- Research and develop CO removal technology based on Preferential Oxidation (PrOx) for integration into fuel processor systems to meet technical targets for contaminant removal, transient performance, energy efficiency, cost, volume, weight, and durability.
- Examine removal of other impurities identified in fuel cell system durability testing
  - Hydrocarbons, ammonia, hydrogen cyanide, carbon, particulate

Key Technical Targets for Reformate Cleanup

- CO removal - < 10 ppm CO steady-state, < 100 ppm transient
- H_2/CO selectivity (efficiency) - <0.2
- Transients (time from 10% to 90% power) - < 1 second
- Startup (cold start to maximum power) - < 0.5 min from 20 °C, < 1 min from –20 ° C
- Other contaminants – e.g., NH_3 < 0.1 ppb
- Fuel Processor targets -
  - Reformate cleanup is a component of the fuel processor subsystem. Thus, it must meet the overall targets of cost, volume, weight, efficiency, and durability.

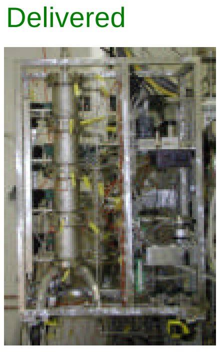
Approach

- Focus on technology transfer and collaboration
  - Transfer PrOx technology developed in the course of the six year OAAT research program at Los Alamos.
  - McDermott Technology
    - Provide Laboratory PrOx subsystem for PrOx and catalyst testing
    - Collaborate on design
  - Argonne National Laboratory
    - Support ANL quick-start fuel processor program with PrOx design and experiments
  - CRADA with H2Fuel, LLC
  - Publications and presentations
  - Other interactions as requested
- Complete ongoing reformate cleanup projects
  - PrOx catalyst investigations
  - PrOx transient response
- Support fuel cell system durability testing
  - Transport of other impurities through PrOx components
  - Removal of other impurities using modified PrOx components

Tech Transfer/Collaborations: *McDermott Technology*

- Goals
  - Collaboration on and support of PrOx testing and catalyst investigations using LANL developed laboratory PrOx subsystem
  - Transfer LANL PrOx experience and knowledge
  - Collaborate on PrOx reactor designs

Laboratory PrOx Subsystem Developed and Delivered



Lab PrOx subsystem shown mounted in LANL PrOx test facility

- PrOx subsystem developed and delivered to McDermott Technology
  - Based on a modular PrOx design developed for laboratory testing at LANL.
- PrOx Subsystem features:
  - 4 stage design for high inlet CO concentrations (up to 2% CO)
  - Air injection flow control and measurement
  - Coolant flow control and measurement
  - Pressure and temperature instrumentation
  - Data acquisition and control interface with computer control system
  - Modular design with replaceable catalyst cartridges for catalyst reconfiguration – lengths and supports can be changed.
- Ongoing Work
  - Collaboration with McDermott personnel on setup and interface of PrOx subsystem in their test facility
  - Collaborate on PrOx testing and PrOx catalyst investigations

Tech Transfer/Collaborations: *Argonne National Laboratory*

- Goals:
  - Support ANL Quick-start fuel processor research program with LANL PrOx technology integration
    - Identify PrOx options enabling rapid fuel processor startup
    - Experimental measurements of PrOx component response to simulated startup conditions and events
    - Identification of PrOx catalysts and component design for rapid start

PrOx Startup Issues

- Maintain fuel processor outlet CO concentration within fuel cell stack tolerances through the startup transient and transition to normal operation.
  - Low catalyst light-off temperature for CO oxidation
  - Wide temperature range for CO selectivity
  - Reduced thermal mass of catalysts and components in contact with the flow
  - Startup heating mechanisms
  - CO absorption during startup transient with regeneration during normal operation
  - High CO startup option
  - Control options with staged reactors

PrOx Startup Simulation Experiments

PrOx single-stage configuration for measurement of catalyst temperature response

- Transient simulation capabilities of PrOx test facility can be used to simulate startup transient operation and measure PrOx component response
- Conduct heat transfer experiments to measure monolith temperature response to a heating gas stream.
- Test catalyst and PrOx configurations for startup CO control
- Status:
  - Preliminary experiments have been conducted on a single-stage catalyst configuration
  - Based on the observations, we plan to modify the experiment to better simulate a startup scenario.

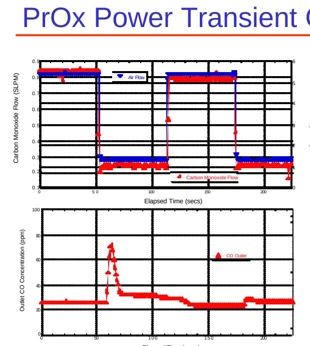
PrOx Catalyst Performance on Low Mass Substrates

- Improve PrOx steady-state and transient performance by incorporating catalysts on low-mass substrates
  - Reduce size and number of stages with improved catalysts
- Compare performance of PrOx catalysts on:
  - High cpsi monoliths
  - Ceramic foams
  - Metal foams
- Measure CO conversion and selectivity as a function of:
  - Temperature range (20 °C to 200 °C)
  - Space velocity
  - Inlet CO concentration (1000 ppm – final stage to 2% - high inlet)
  - Air Injection (oxygen stoichiometry)
- Status:
  - PrOx catalysts have been obtained on monoliths, ceramic and metal foams
  - Catalyst testing has begun with testing of PrOx catalyst on high cpsi monoliths

Measurement of PrOx Transient CO Response

- Rapid transient and startup response requires diagnostic measurements at a time scale faster than the time scale of the desired response.
  - Target transient response is less than a second
  - The experimental resolution of the PrOx test facility was on the order of 1 second; gas concentration measurements ranged from ~1 second to 3 minutes not including the sample line delays.
- To speed up the gas concentration measurements, we have started implementation of a tunable diode laser absorption system for *in situ* measurements of CO concentration
  - Optical access flanges permit an *in situ* absorption measurement at the catalyst outlet, thus eliminating the sample delay time.
  - Examining possibilities to expand *in situ* measurements to include oxygen, ammonia, and hydrogen sulfide by adding additional laser wavelengths.
- Experiments
  - Power transients – change in total flow in response to power demands
    - Investigate required timing for air injection
    - Quantify CO concentration transients
  - Composition transients – change in gas composition
    - Sensitivity to variations in CO concentration
    - Sensitivity to variations in air injection flows

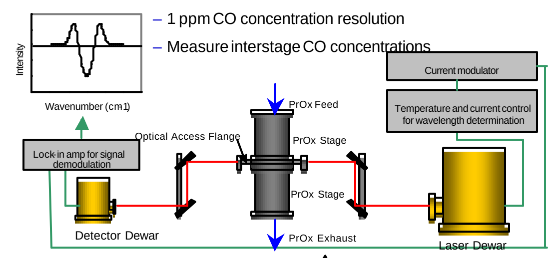
PrOx Power Transient CO Response



- Measurement of Outlet CO in response to a step transient
  - Single-Stage Monolith
  - Nominal 2000 ppm CO Inlet
  - 10 kWth to 30 kWth LHV H_2 total flow, gasoline reformate
- Outlet CO response
  - Air lead 1 sec up-transient
  - Air lag 1 sec down-transient
  - Maintained below 100 ppm peak
- Experiment Resolution - ~ 1 second

Transient *in situ* CO Diagnostics

- Tunable Diode Laser Absorption System
  - 100 msec temporal resolution
  - 1 ppm CO concentration resolution
  - Measure interstage CO concentrations



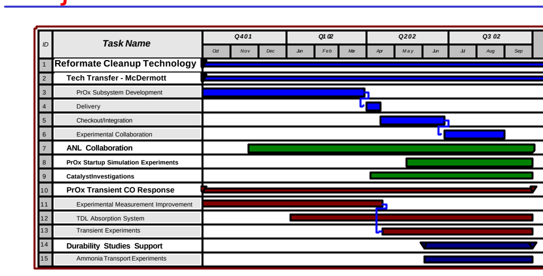
Durability Studies Support

- Measure effects of contaminants identified in the Fuels/Durability testing program on the PrOx performance
- Investigate mechanisms and catalysts for removal of these contaminants
- Contaminants identified
  - Hydrocarbons – incomplete partial oxidation
  - Ammonia – produced from bound-nitrogen fuel components or in off-normal operating conditions
- Experiments
  - Transport of ammonia through PrOx reactor components
    - Effects of ammonia on PrOx catalyst performance and durability
    - Oxidation of ammonia in PrOx reactors

Ongoing Work

- Completion of ongoing tech transfer and collaborations
  - McDermott Technology
  - Argonne National Laboratory Quick-Start FP
  - CRADA with H2fuel, LLC
- Completion of catalyst and transient investigations
- Documentation and publication of non-proprietary knowledge and results developed in PrOx research and development tasks
- Durability studies support

Project Timeline



Future Work

- Trace fuel processor impurity removal
  - Ammonia, nitrogen-containing compounds
  - Other poisons identified in durability testing
- Other methods of hydrogen purification
  - Component separation
- Close out PrOx research and development project
- Industrial collaborations

Summary

- Focus is on technology transfer and collaborations
- Completing work on catalyst investigations and transient CO diagnostics
- Examining effects of other contaminants
- Summing up PrOx research and development program

For more information, contact:

Michael A. Inbody
MS J-576 / ESA-AET
Los Alamos National Laboratory
Los Alamos, NM 87545

Phone: (505)-665-7853
Fax: (505)-665-9507
e-mail: inbody@lanl.gov