



Pall Corporation



# High-Performance, Durable, Palladium-Alloy Membrane for Hydrogen Separation and Purification



Presented by:

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This presentation does not contain any proprietary or confidential information

# Project Contributors

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# Overview

## Timeline

- July, 2005 start date
- September, 2009 end date
- 30% complete

## 2010 Targets

- Cost: \$2.50 per GGE for H<sub>2</sub> (delivered/untaxed)
- H<sub>2</sub> Quality: 99.95%

## Budget

- \$4 Million Project Total
  - \$2.4M DOE share
  - \$1.6M Contractor share
- \$100K DOE funding in FY05
- \$175K DOE funding in FY06
- \$540K DOE funding in FY07\*

\* Anticipated

## Partners

- Chevron
- Colorado School of Mines
- ORNL – High Temperature Materials Lab

# Project Objectives

- Establish the technical and economic viability for use of a palladium alloy composite membrane in a distributed hydrogen production system
  - Propose a process that leverages the technical capabilities of the membrane for maximum economic benefit (reduced *gallon of gas equivalent* cost)
  - Optimize membrane performance in terms of hydrogen throughput, purity and durability
  - Minimize capital cost for the gas separation module
    - Pressure vessel
    - Internal hardware
    - Membrane
    - Substrate



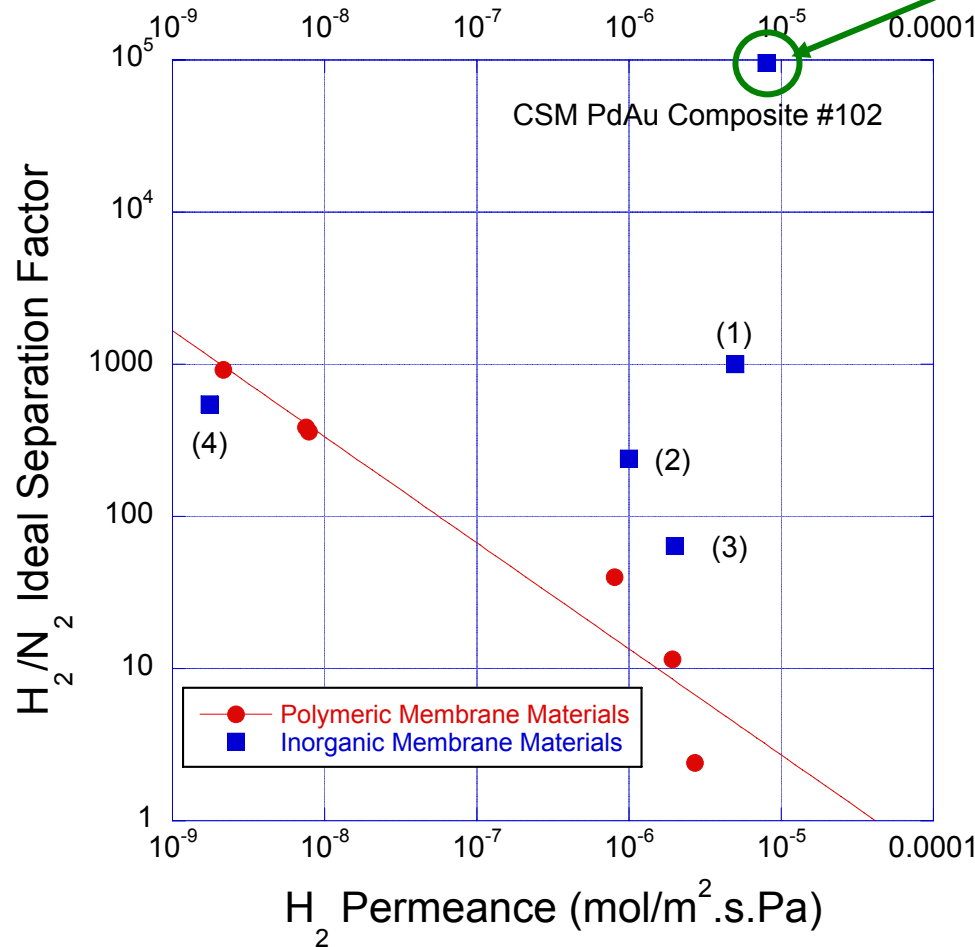
Photo courtesy of Chevron

## Why Membrane ?

- Capital and operating cost for a hydrogen production system can potentially be reduced through process intensification
- Membranes that can be operated at high temperatures allow for integration with high temperature reforming processes
- Simple, compact separation systems can be designed using membranes

# Why Palladium Membrane ?

## Project Accomplishments

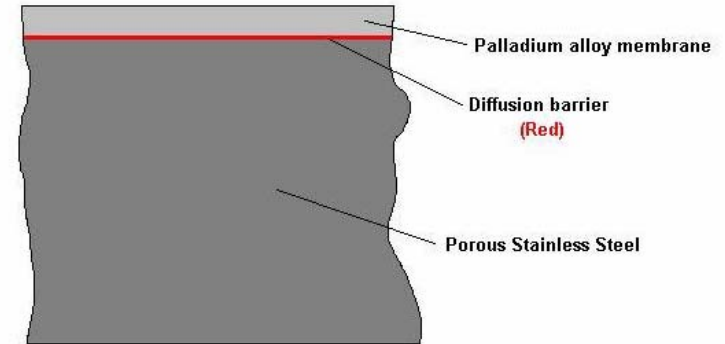


1. Lee, D., Zhang, L., Oyama, S. T., Niu, S., and R. F Saraf, *J. Membr. Sci.*, **231**, 117(2004).
2. Kajiwara, M., Uemiya, S., Kojima, T., and E. Kikuchi, *Catal. Today*, **56**, 65(2000).
3. DeVos, R. M. and H. Verweij, *Science*, **279**, 1710(1998).
4. Hassan, M. H., J. D. Way, P. M. Thoen, and A. C. Dillon, *J. Membr. Sci.*, **104**, 27(1995).
5. Polymer line from : Robeson, L. M., *J. Membr. Sci.*, **62**, 165(1991).

# Components of a Composite Membrane

## **Pd alloy membrane**

- Functional layer provides for gas separation
- Critical features: thickness, alloy composition and number of defects



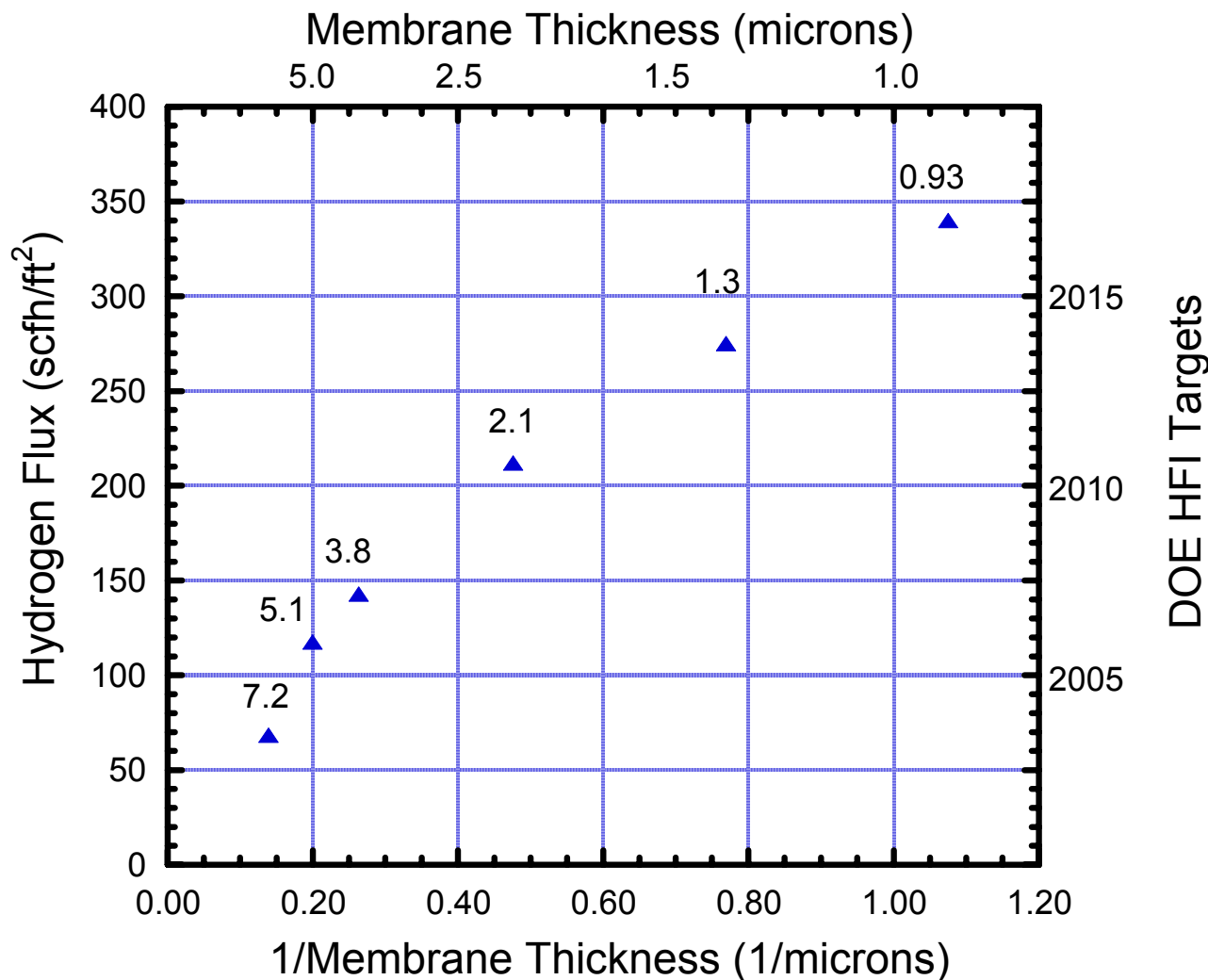
## **Diffusion barrier**

- Enables formation of functional layer
- Critical features: surface properties, material, gas permeability, number of defects

## **Porous Stainless Steel**

- Provides mechanical support that can withstand the operating conditions of the process
- Critical features: permeability, weld configuration, mechanical, thermal and chemical compatibility

# The Influence of Pd Membrane Thickness on Pure H<sub>2</sub> Flux @ 400 °C, 20 psig

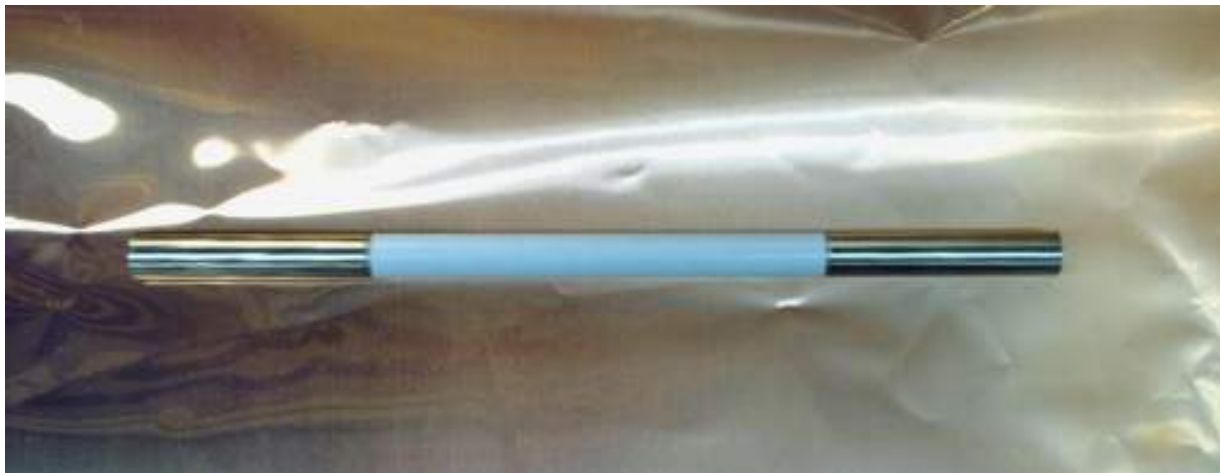




# Technical Accomplishments/ Progress/Results

## Diffusion Barrier Properties

- Surface roughness 2005: Ra = 25 – 35 micro inch
- Surface roughness 2006: Ra = 8 – 12 micro inch
- Membrane over welds 2005: no leaks up to 20 psi
- Membrane over welds 2006: no leaks up to 40 psi



# Porous Stainless Steel

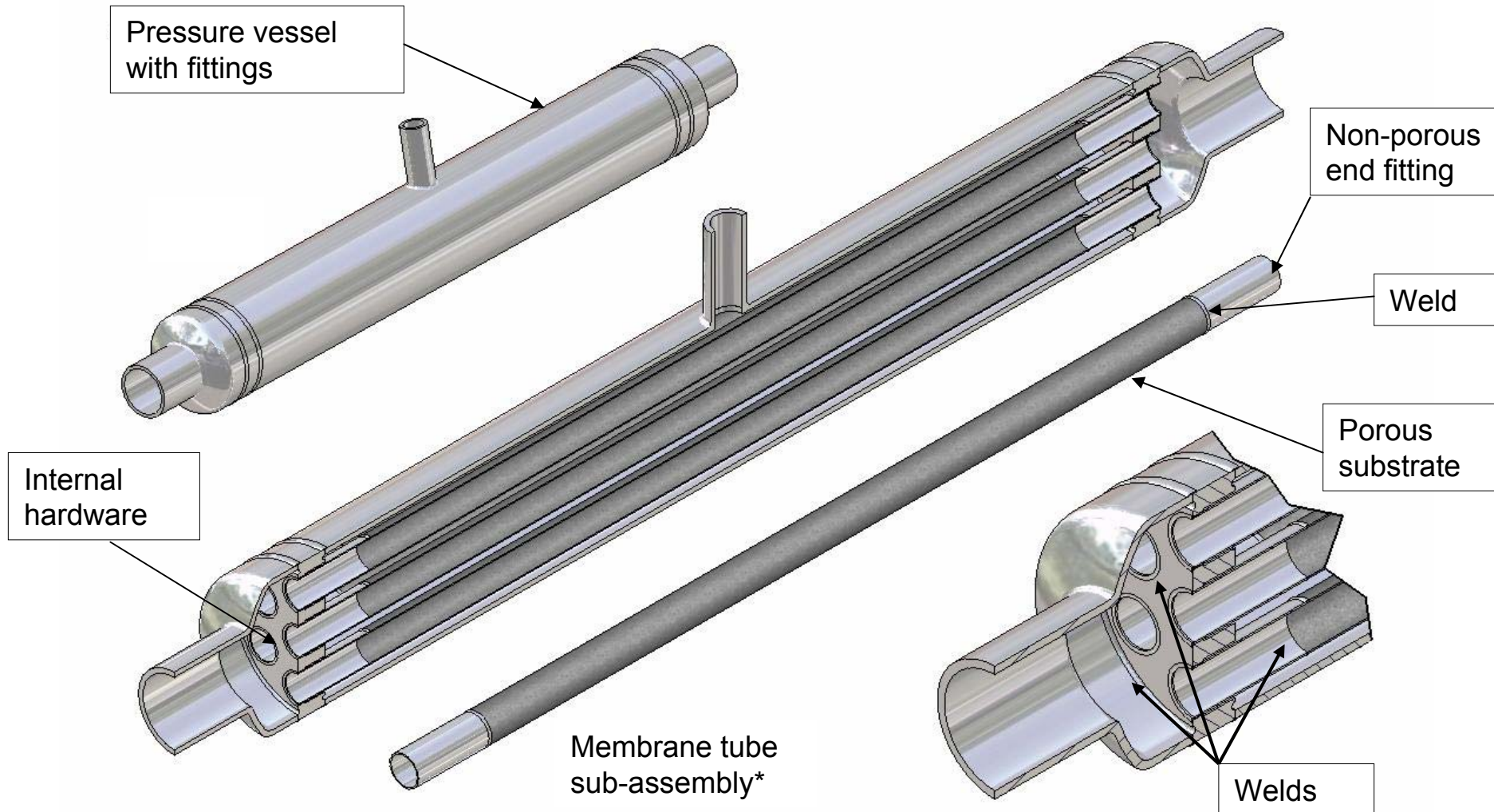


← Support tubes – as welded



Support tubes ZrO<sub>2</sub> coated

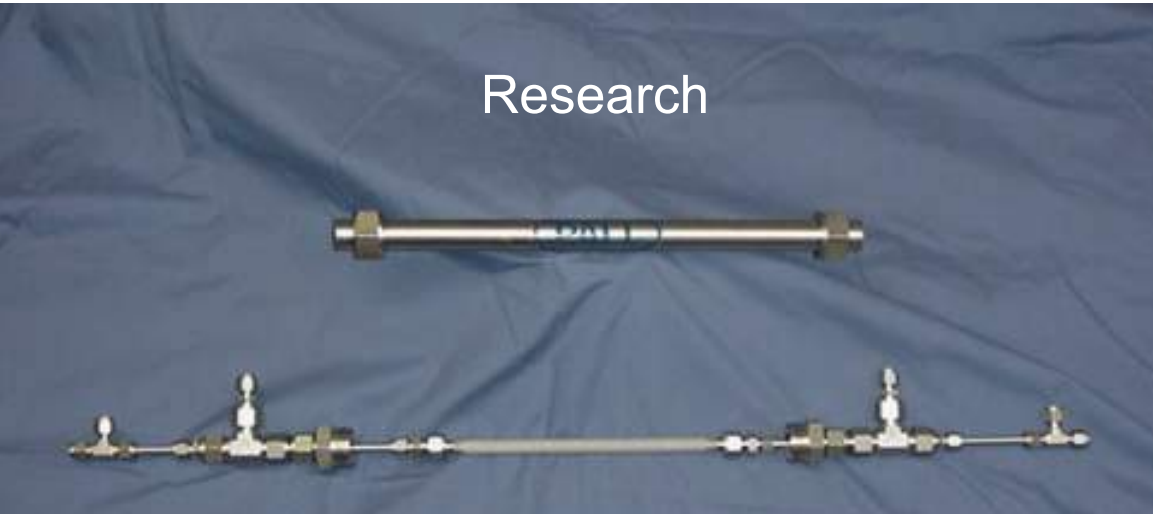
# Components of a Gas/Gas Separation Module



\* Pd alloy membrane not shown, typically on the OD of the tube

# Scalability of Metal Tube Technology

Research



Development



Commercialization



# Guidelines for Membrane Performance

## 2010 DOE / EERE Targets for Dense Metallic Membrane\*

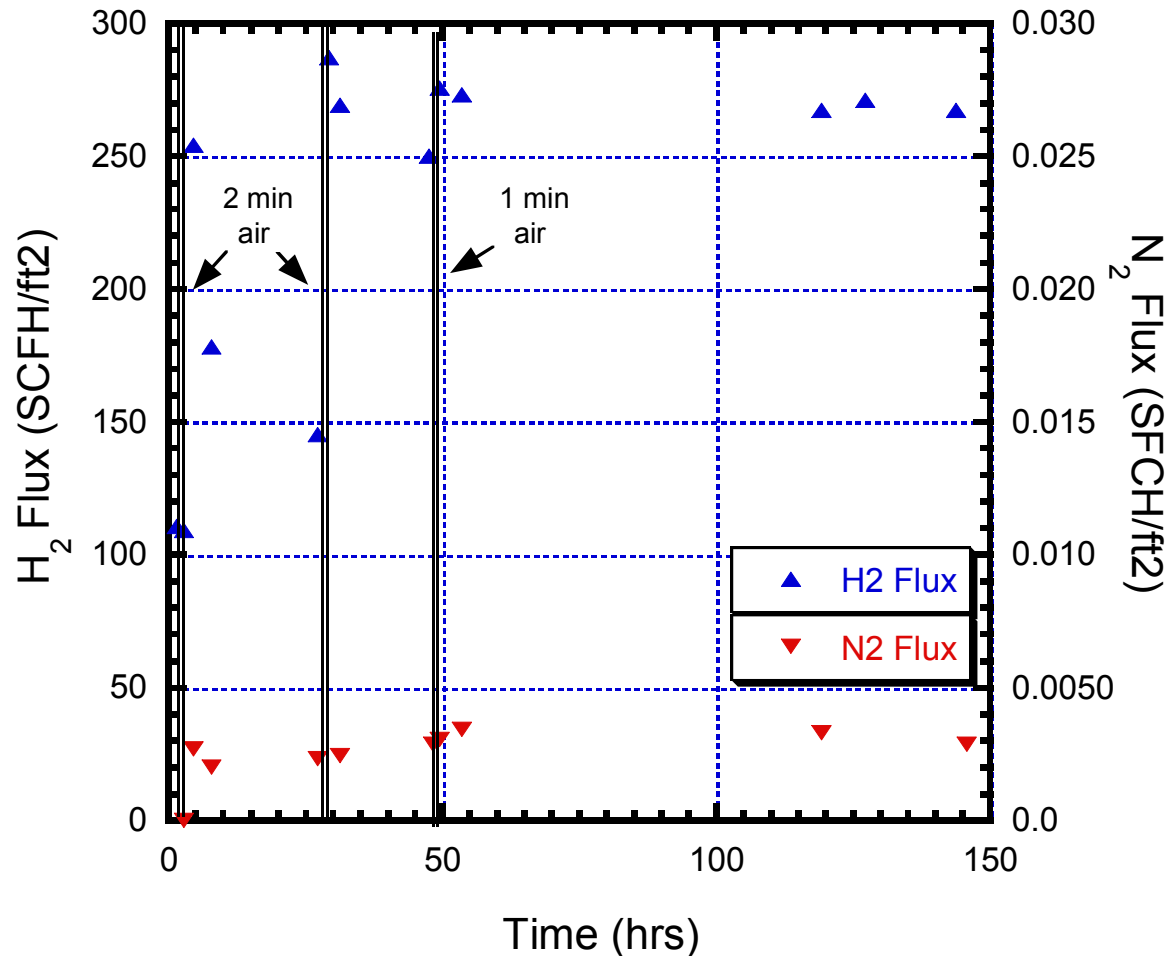
- Flux: 250scfh/ft<sup>2</sup> @20 psi & 400°C
- Module Cost: \$1,000/ft<sup>2</sup>
- Durability: 3 years
- Operating pressure: 400 PSI
- H<sub>2</sub> Recovery: > 80%
- H<sub>2</sub> quality: 99.99 %

\* As per the Multi-Year RD&D Plan Updated 11/14/06

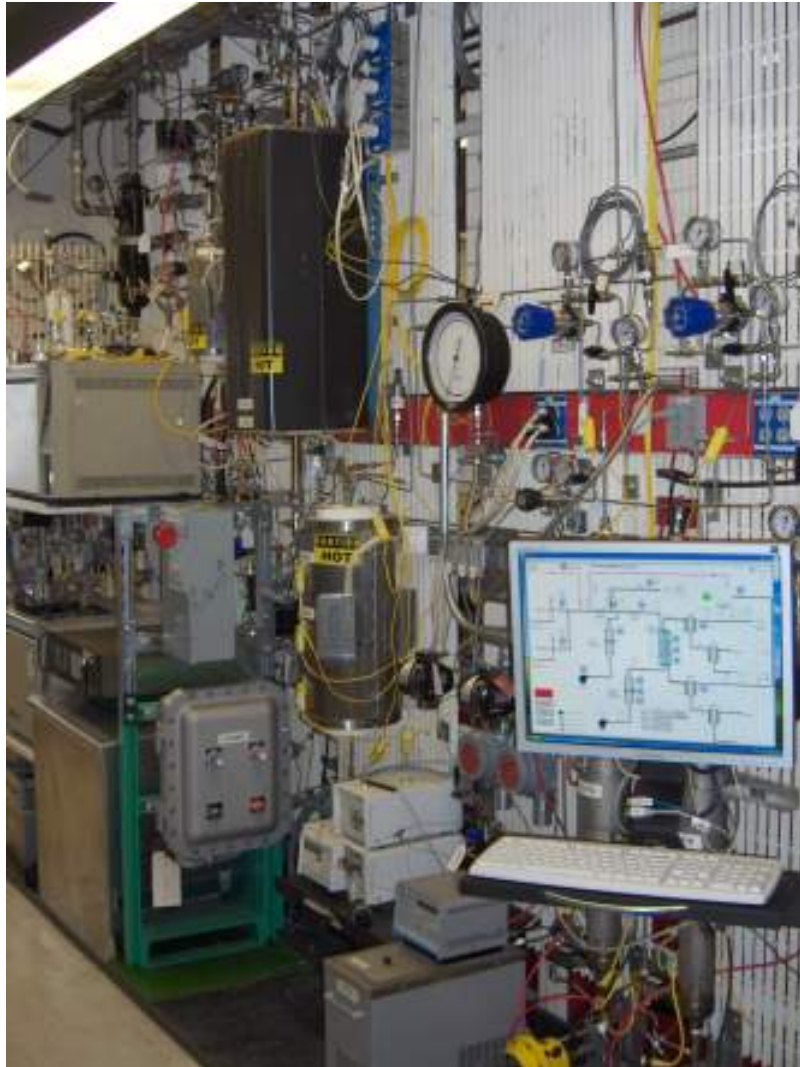
<http://www1.eere.energy.gov/hydrogenandfuelcells/mypp/pdfs/production.pdf>

# Technical Accomplishments/ Progress/Results

**H<sub>2</sub> and N<sub>2</sub> Flux for PdAu Membrane, CSM-Pall-102,  
at 20 psi Transmembrane Pressure and 400 °C.**

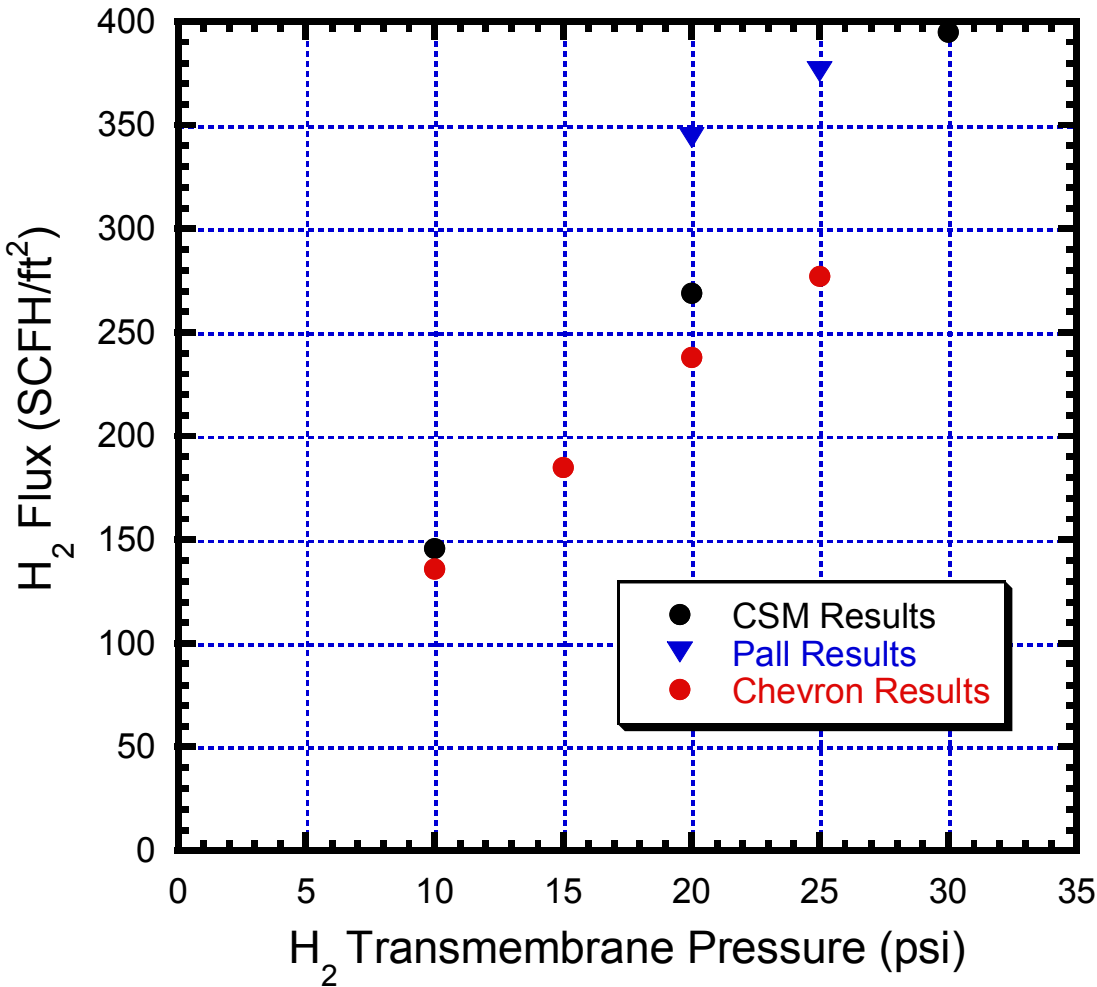


# Automated Gas/Gas Separation Test Stand



*Photos courtesy of Chevron*

# CSM Test Data Confirmed at Pall Corp and Chevron



**Note: Same membrane sample tested at all three locations**

Test Location	Ideal H <sub>2</sub> /N <sub>2</sub> *
CSM	90,000
Pall	∞
Chevron	∞

\* Ideal H<sub>2</sub>/N<sub>2</sub> selectivity at 20 psi

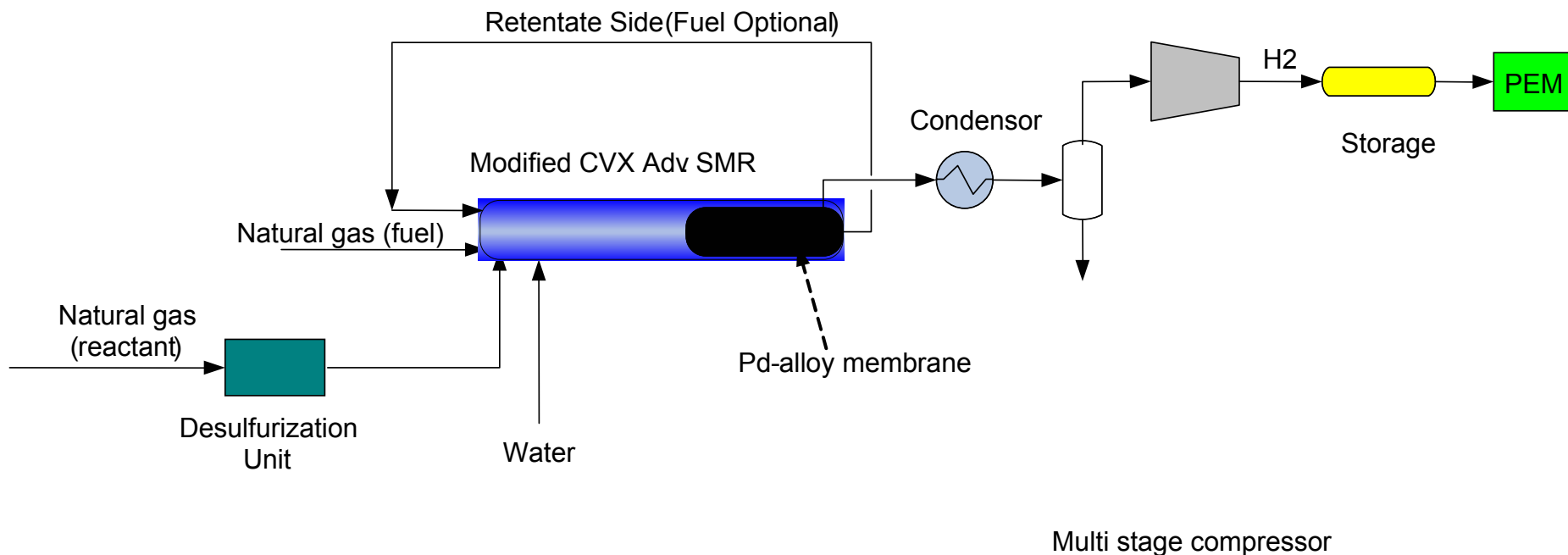


## Future Work

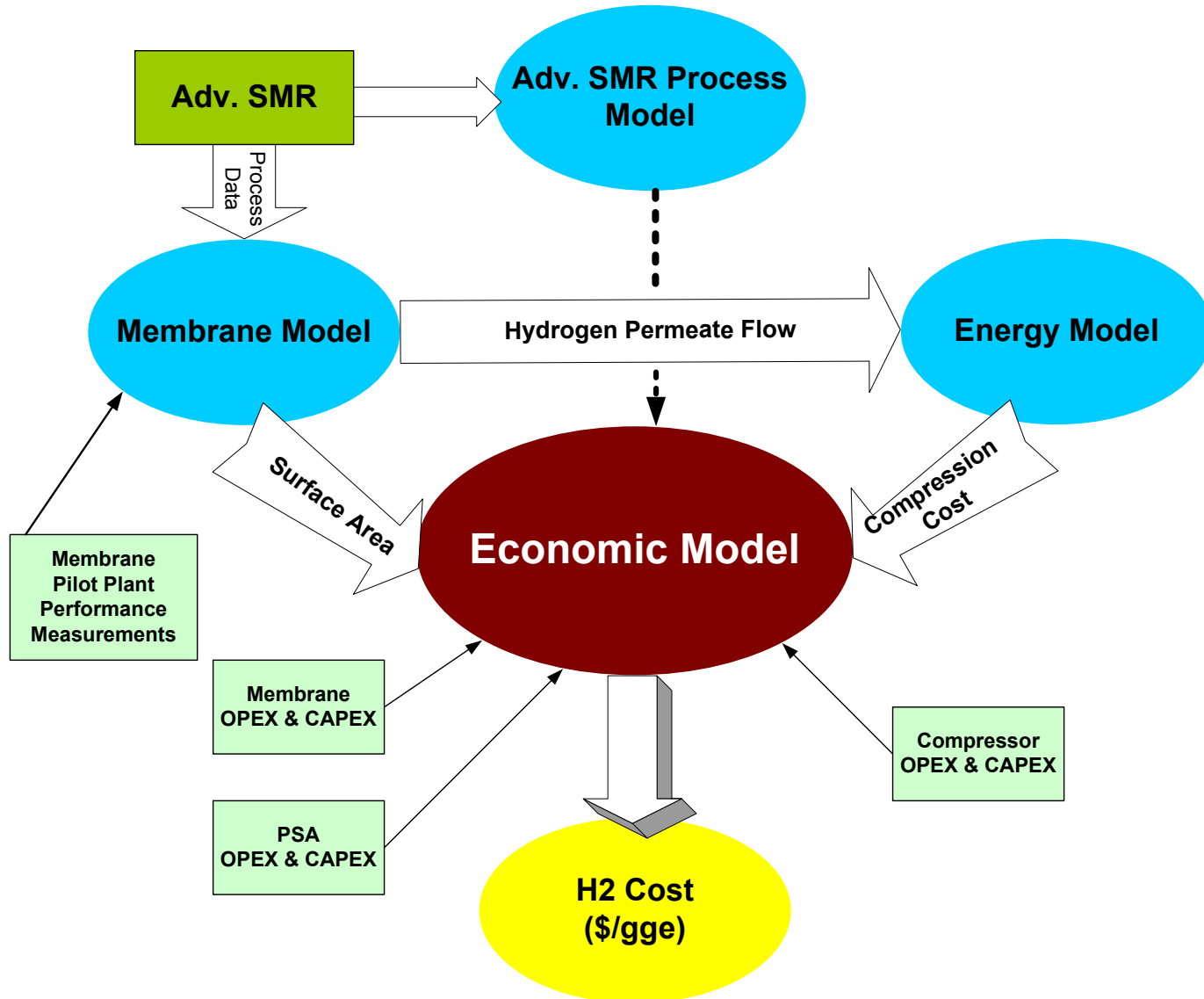
- Optimize substrate and alloy properties to meet or exceed membrane targets
- Test membrane in synthetic reformat streams to establish conditions for >80% hydrogen recovery
- Establish long term durability testing at temperature
- Use membrane properties for economic analysis of advanced process design to achieve targeted H<sub>2</sub> production costs

# Proposed Use of Pd Alloy Membrane in a Hydrogen Production System

## Modified Chevron Adv. SMR Integrated with Pd -alloy Membrane



# Economic Analysis Strategy



# Summary

- Developed porous stainless steel substrate and diffusion barrier that enables formation of high flux Pd alloy membranes
- Produced Pd alloy membranes that exceeded the 2010 target for H<sub>2</sub> flux and purity
- Analyzed membrane and module manufacturing costs to confirm the current cost basis (\$1,500 ft<sup>2</sup>)
- Completed fabrication of an automated test stand for long term testing with synthetic reformat
- Established membrane, process and energy models that will be used to determine overall economics for H<sub>2</sub> production

# Hydrogen Safety

The most significant hydrogen hazards associated with this project are:

- A test system that leaks hydrogen while at temperature. Plumbing is tested for leaks prior to installation in the furnace
- Mixing hydrogen with air while system is at temperature. An argon or nitrogen purge is used to evacuate the system of residual air after an air purge cycle, prior to introducing hydrogen to the system.