

# Composite Pd and Alloy Porous Stainless Steel Membranes for Hydrogen Production and Process Intensification

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**Project ID: PD007**

This presentation does not contain any proprietary, confidential, or otherwise restricted information.



# Overview

## Timeline

- Start : 5/7/2007
- Finish : 5/6/2011
- 100% Complete

## Budget

- Total Project Cost: \$ 2,228,851
  - DOE Share: \$ 1,756,444
  - Recipient Share: \$ 472,407
- Funding Received:
  - FY08: \$ 442,785
  - FY09: \$ 420,638
  - FY10: \$ 392,803
  - FY11: \$ 500,218

- DOE Award #: DE-FC26-07NT43058
- DOE Project Manager:

Dr. Daniel Driscoll

## Subcontractor

- Adsorption Research Inc. (ARI)

## Barriers

### → Barriers Addressed:

- Re-reproducibility of high H<sub>2</sub> flux targets
- Setting of Pd thickness and support characteristics to meet 2015 DOE targets
- Determination optimum temperature WGS
- 2D model for CMR simulations, safety and economical analysis
- Absorbent selection & PSA system build-up, testing completed with syngas+H<sub>2</sub>O, H<sub>2</sub>S and COS.

### → Technical Targets\*\*

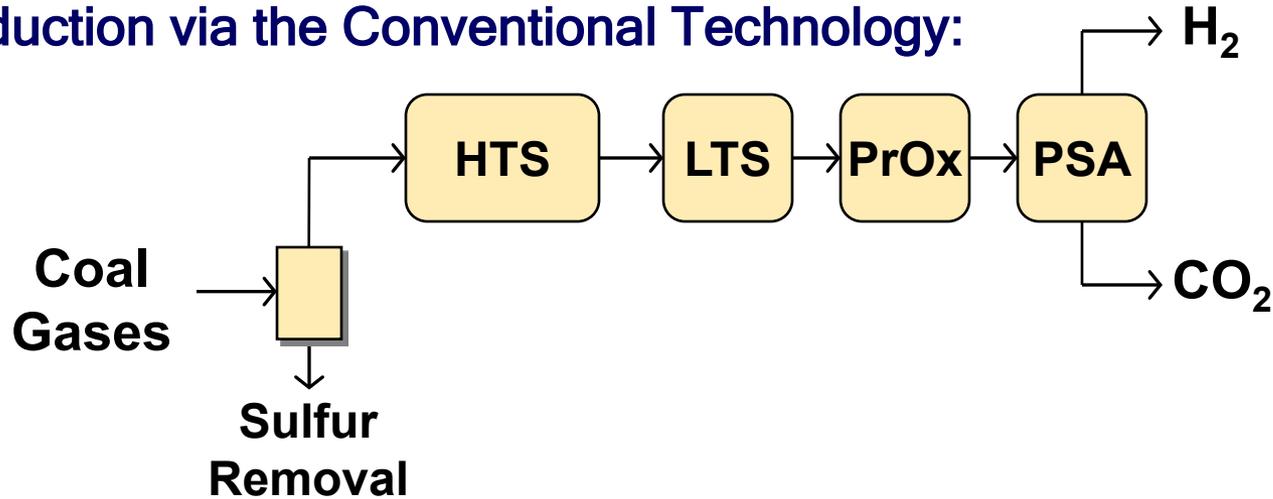
	H <sub>2</sub> Flux <sup>§</sup> [scfh/ft <sup>2</sup> ]	Temp. [°C]	ΔP max. [psi]	H <sub>2</sub> Purity	Sulfur Tolerance
2010	200	300-600	400	99.5%	20 ppm
2015	300	250-500	800-1000	99.99%	>100 ppm
§ @ 100 psi ΔP H <sub>2</sub> partial pressure					
CO Tolerance: Yes; WGS Activity: Yes					

# Project Objectives & Relevance

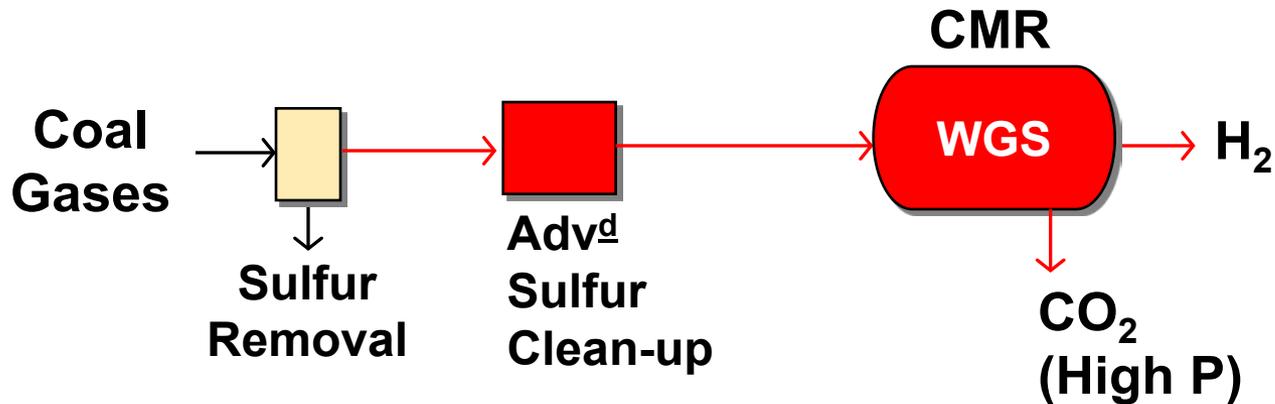
- **Synthesis of composite Pd and Pd/alloy porous Inconel membranes for WGS shift reactors with long-term thermal, chemical and mechanical stability with special emphasis on the stability of hydrogen flux and selectivity**
- **Demonstration of the effectiveness and long-term stability of the WGS membrane shift reactor for the production of fuel-cell quality hydrogen**
- **Research and development of advanced gas clean-up technologies for sulfur removal to reduce the sulfur compounds to <2 ppm**
- **Development of a systematic framework towards process intensification to achieve higher efficiencies and enhanced performance at a lower cost**
- **Rigorous analysis and characterization of the behavior of the resulting overall process system, as well as the design of reliable control and supervision/monitoring systems**
- **Assessment of the economic viability of the proposed intensification strategy through a comprehensive calculation of the cost of energy output and its determinants (capital cost, operation cost, fuel cost, etc.), followed by comparative studies against other existing pertinent energy technologies**

# Approach: Coal Gasification & CMR

H<sub>2</sub> Production via the Conventional Technology:



Novel Catalytic Membrane Reactor (CMR):



# Project Schedule & Milestones

Tasks	Year 1				Year 2				Year 3				Year 4			
	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4
	Months															
	3	6	9	12	15	18	21	24	27	30	33	36	39	42	45	48
Gas Clean-up & Fast PSA using Structured Adsorbent			M1 ✓		G1 ✓											
						M2 ✓										
Membrane Synthesis		M4 ✓								M3 ✓						
				M5 ✓			G2 ✓									
Membrane Characterization & Reactor Performance						M6 ✓										
										M7 ✓						
Membrane Reactor Modeling			M8 ✓													
Process Intensification					M9 ✓											
Process Control System; Design & Implementation								M10 ✓								
Process monitoring System; Design & Implementation										M11 ✓						
Address process safety; risk assessment/management																
Program Management & Reporting																

# Membrane Properties & Permeation Test Set-up

## ➤ Membrane:

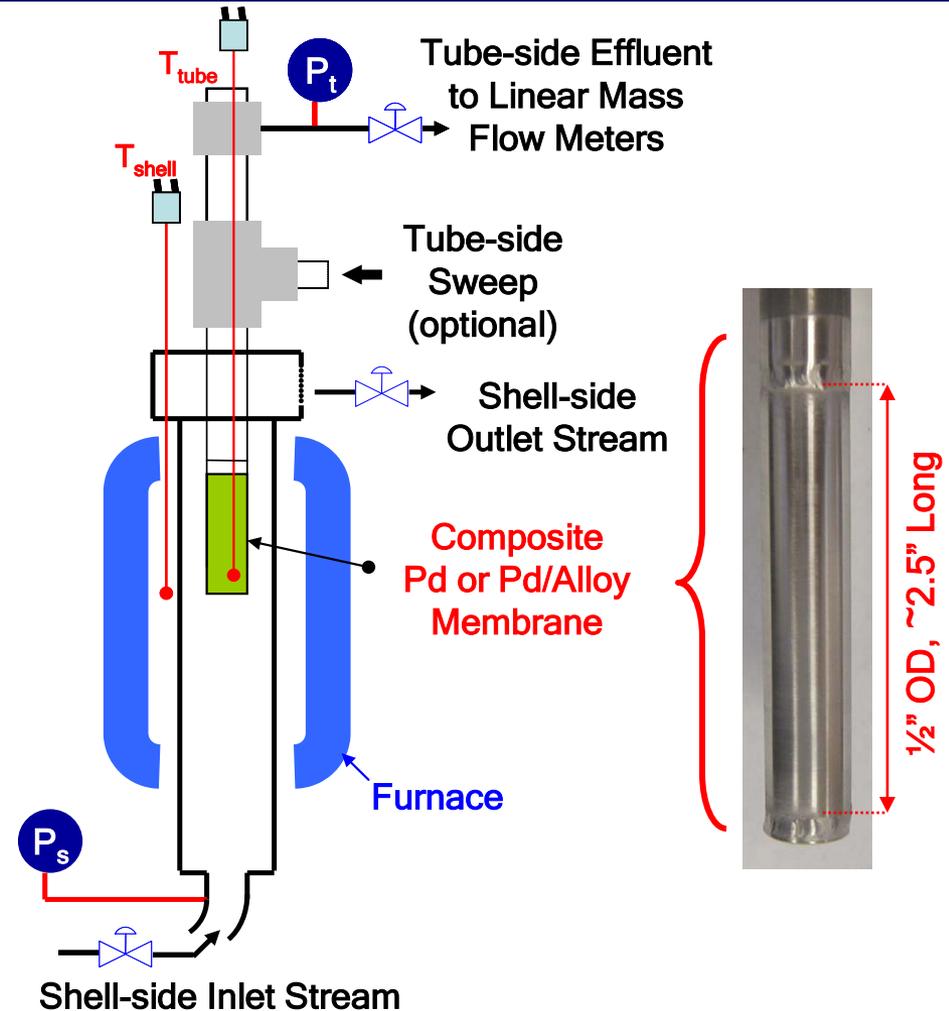
Pd supported on porous Inconel (media grade 0.1  $\mu\text{m}$ )

## ➤ Method of Preparation: Electroless Plating

## ➤ Geometry:

Tubular (Plated on the outside of a tube)

## ➤ Membrane Area $\approx 25 \text{ cm}^2$

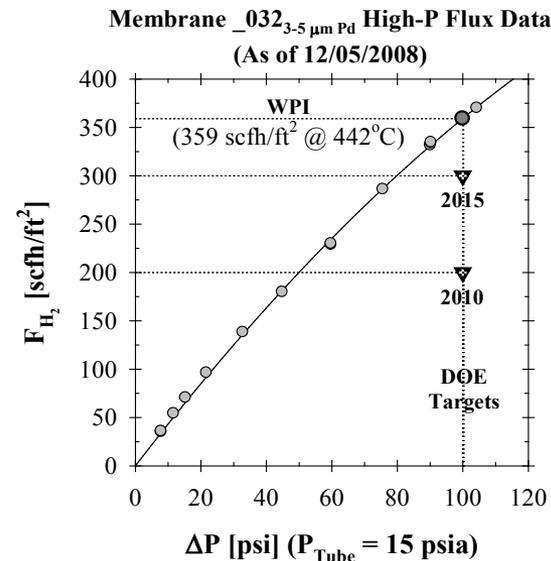


Similar setup equipped w/ pre-heater, mixer, cold trap & GC was utilized for the mixed gas & WGS reaction tests

# Previous achievements

- Excellent **long-term H<sub>2</sub>/He selectivity stability** was achieved over a total testing period of **~3550 hours (>147 days)** (membrane #029)
- Selectivity stability reproduced with membranes #030 / 031 / 033
- The thicknesses of #029/030/031/033 were all higher than 7 μm
- Selectivities in the order of 5000 -10 000 after long test at 450°C
- #032 showed high flux 359 scfh/ft<sup>2</sup> but low selectivity of 450 (3-5 μm thick)

#029 <sub>7.6 μm</sub>	<ul style="list-style-type: none"> <li>• H<sub>2</sub>/He ≅ ∞ at 450°C</li> <li>• H<sub>2</sub> purity: ≥ 99.999%</li> <li>• 3550 hours (&gt;147 days)</li> </ul>
#030 <sub>7.9 μm</sub>	<ul style="list-style-type: none"> <li>• H<sub>2</sub>/He ≅ 10000 at 450°C</li> <li>• H<sub>2</sub> purity: ≥ 99.99%</li> <li>• 1400 hours (&gt;58 days)</li> </ul>
#031 <sub>7.0 μm</sub>	<ul style="list-style-type: none"> <li>• H<sub>2</sub>/He ≅ 4500 at 450°C</li> <li>• H<sub>2</sub> purity: ≥ 99.99%</li> <li>• 2200 hours (&gt;90 days)</li> </ul>
#033 <sub>8.7 μm</sub>	<ul style="list-style-type: none"> <li>• H<sub>2</sub>/He ≅ 9725 at 450°C</li> <li>• H<sub>2</sub> purity: ≥ 99.99%</li> <li>• 800 hours (&gt;32 days)</li> </ul>



# Progress Towards DOE H<sub>2</sub> Flux targets

Mem.	Support	LPd (μm)	Temp. (°C)	Time (hs)	Permeance / Pd Foil (scfh/ft <sup>2</sup> -psi <sup>0.5</sup> )	Selectivity (H <sub>2</sub> /He)	F at ΔP=100 psi Exp. / Pd Foil (scfh/ft <sup>2</sup> )
034	0.1 μm/Inc.	8.7	300	200	12 / <u>24</u> ¶	-	<b>82 / 164</b> ■
035	0.1 μm/Inc.	20	300	150	6 / <u>10</u>	-	<b>82 / 68</b>
036	0.1 μm/Inc.	11.4	450	100	33.4 / <u>35.2</u>	-	<b>229 / 241</b>
036R	idem	12 (3-5 wt%Au)	450		16 / ~ <u>33</u>	200	<b>110 / 226</b>
038	0.1 μm/Inc.	6	450	170	38 / <u>67</u>	330	<b>280 / 459</b>
039	0.1 μm/Inc.	7.3	450	165	33 / <u>55</u>		
040	0.1 μm/ PSS 316L	12.3	450	120	20 / <u>33</u>		
041	0.1 μm PSS 316L	8.6	450	200	20 / <u>47</u>		
042	0.1 μm PSS 316L	13.4	450	200	20 / <u>30</u>		

**The presence of mass transfer from support is significant**

¶ xx / xx = permeance of the composite membrane / permeance of a free-standing Pd foil having the same thickness

■ XX / XXX flux at ΔP=100 psi of composite membrane / flux at ΔP=100 psi of Pd foil with the same thickness

RED = does not meet DOE's 2010 target

GREEN = exceed DOE's 2010 target but does not meet DOE's 2015 target

BLUE = meet DOE's 2015 flux target

Underlined = actually measured (not underlined = extrapolated from measured permeance)

Several membranes have the potential to meet 2015 DOE's target



# Support specifications

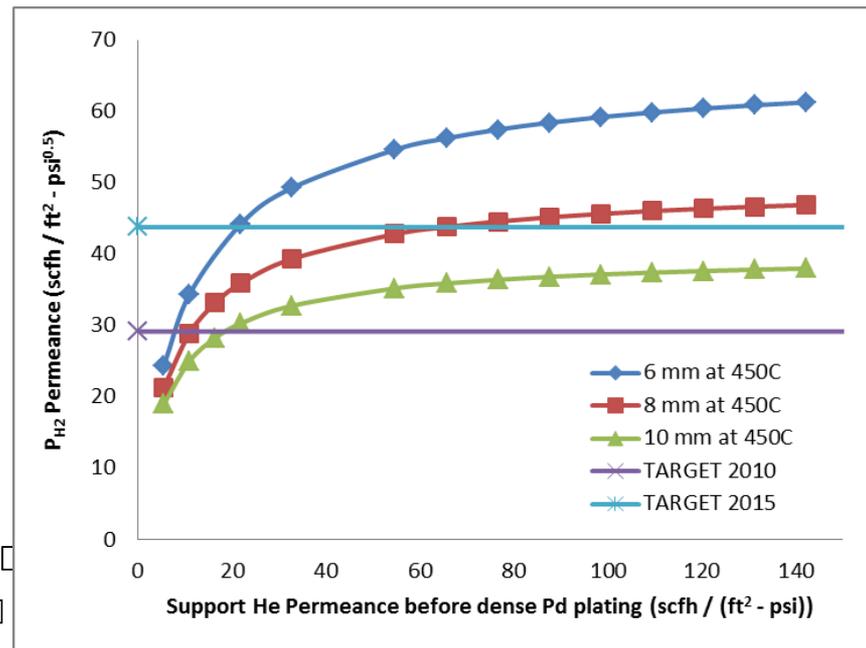
	H <sub>2</sub> Flux <sup>§</sup> [scfh/ft <sup>2</sup> ]	Temp. [°C]	ΔP max. [psi]	H <sub>2</sub> Purity	Sulfur Tolerance
2010	200	300-600	400	99.5%	20 ppm
2015	300	250-500	800-1000	99.9%	>100 ppm
§ @ 100 psi ΔP H <sub>2</sub> partial pressure (or 6.85 psi <sup>0.5</sup> )					
CO Tolerance: Yes; WGS Activity: Yes					



•Hydrogen permeance target  
 2010 → 29.2 scfh/ft<sup>2</sup>-psi<sup>0.5</sup>  
 2015 → 43.8 scfh/ft<sup>2</sup>-psi<sup>0.5</sup>



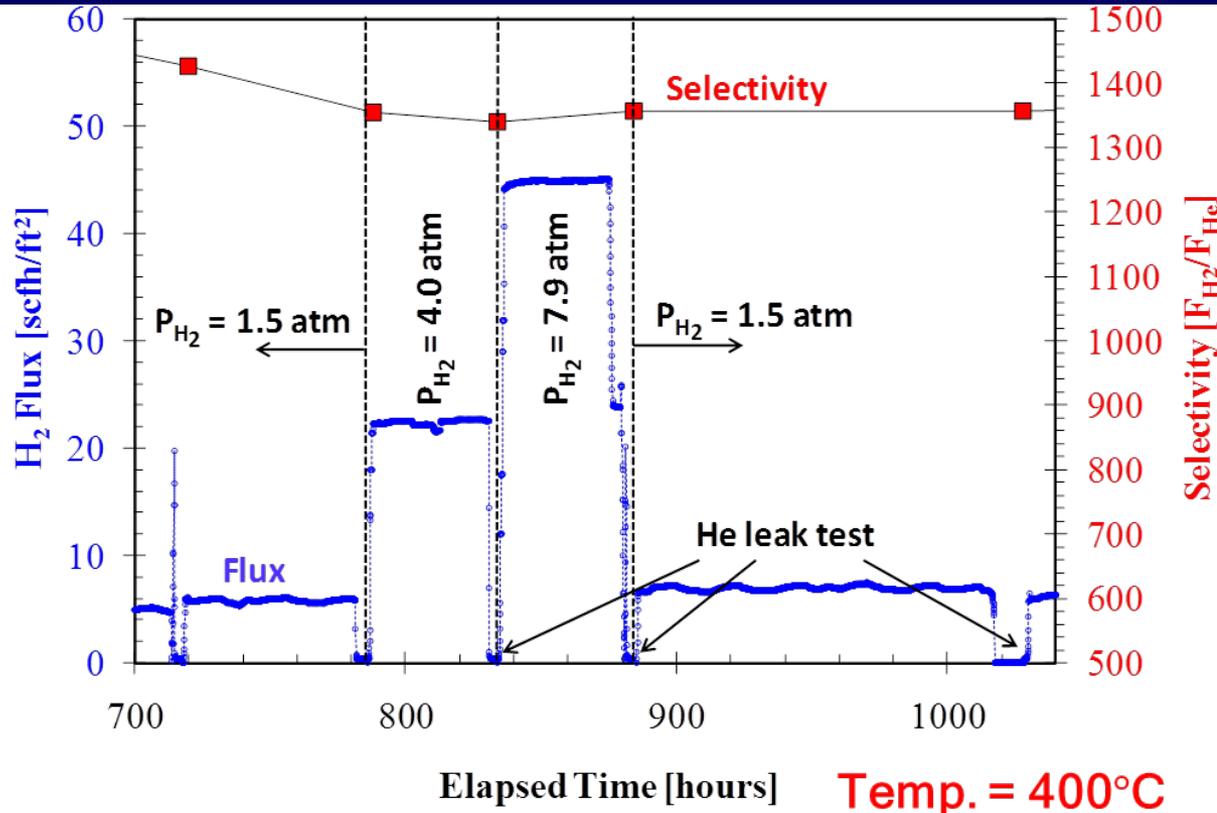
•Pd foil targets  
 → 11.8 μm at 450 C  
 → 9.2 μm at 450 C



▮ 450°C was found to be an optimum temperature for WGS-MR in our group (see later)

- Thickness targeted **7-8 μm** (e.g., 029-031, very selective)
- Support He permeance before Pd deposition **higher than 65 scfh/(ft<sup>2</sup> - psi)**
- Bare support He permeance should be higher than 130 scfh/(ft<sup>2</sup> - psi)**

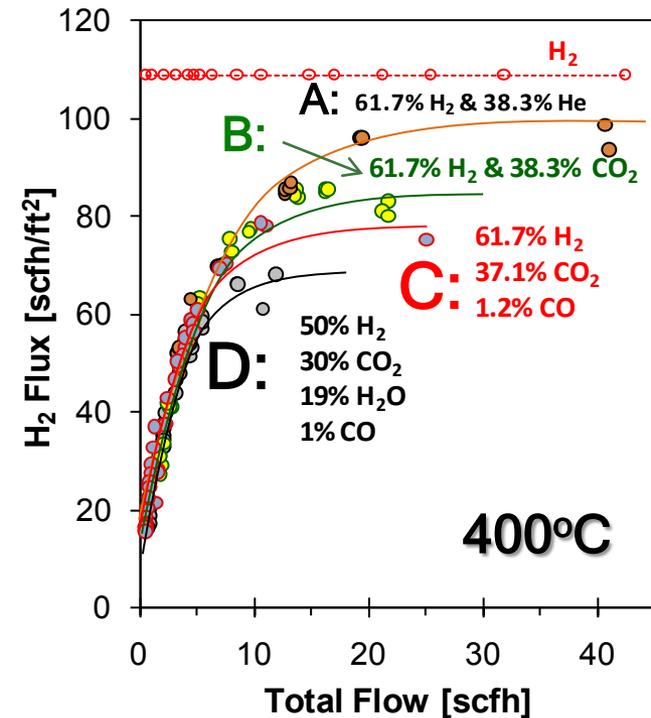
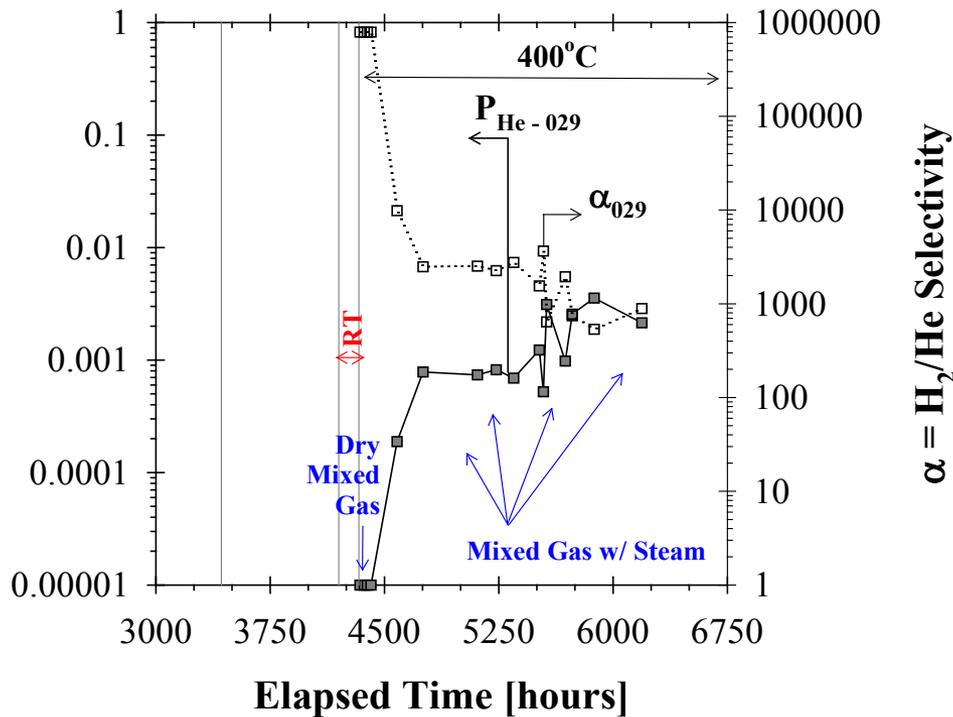
# High Pressure H<sub>2</sub> Testing/Selectivity stability



- Membrane: MA-79
- PSS support,
- 17 μm Pd/Ag layer,
- 10 μm dense Pd layer
- High pressure H<sub>2</sub> fed to the membrane for 100 hours
- Stable H<sub>2</sub> fluxes of 74.5 and 147 scfh/ft<sup>2</sup> at 60 and 120 psi respectively
- H<sub>2</sub>/He selectivity over 1000 and stable over the entire testing period

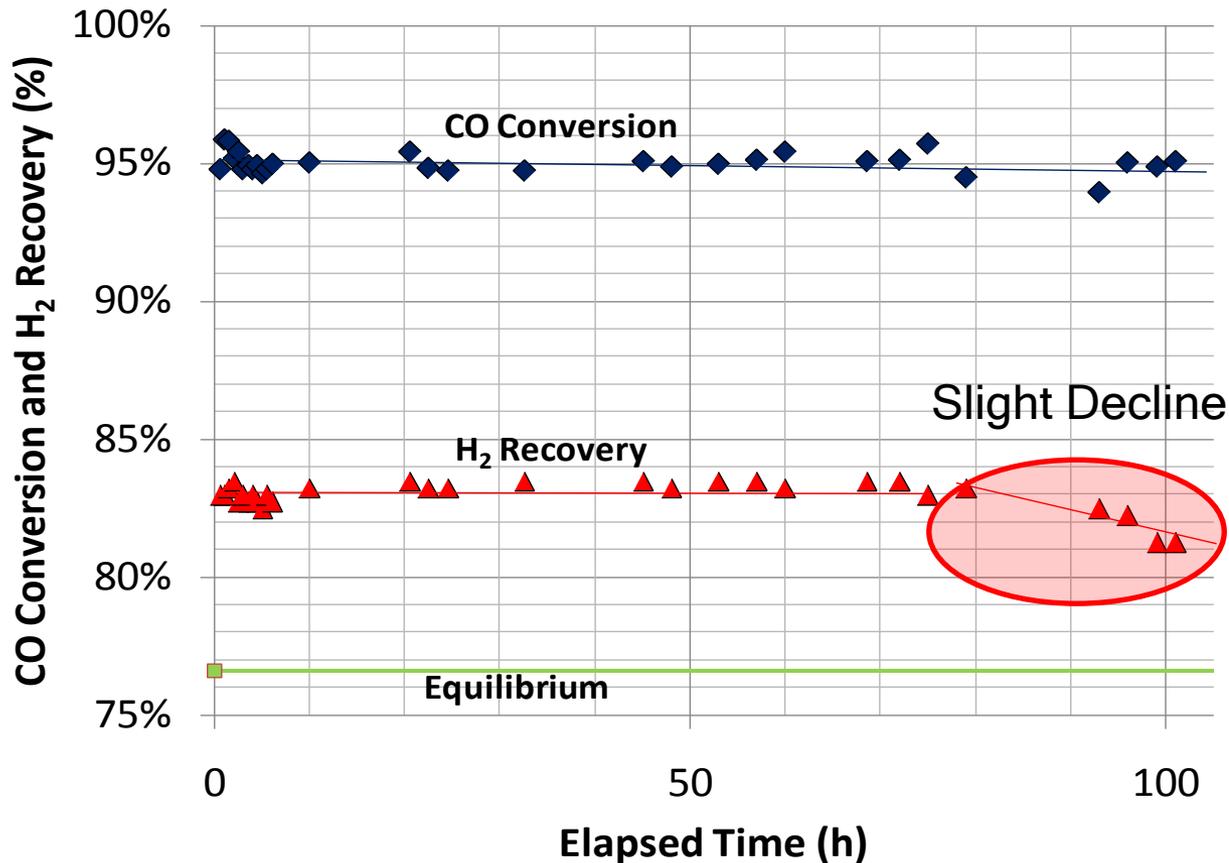
- Pressures of 330 psi have also been tested → H<sub>2</sub>/He Selectivity before and after 330 psi. equaled 250
- n(H/Pd) estimated at 0.05 at 330 psi / 450°C → max compression stress -300-500 Mpa
- → high pressure has a negligible effect on leak formation

# Mixed Gas Testing of Membrane #029<sub>7.6 μm Pd</sub>



- Mixed gas permeation testing for an **additional ~3000 hours** at **~400°C** & at a **ΔP range of 1-14 atm** (P<sub>Low</sub>=1 atm) w/ stable H<sub>2</sub> Flux, H<sub>2</sub>/He Selectivity & no significant increase in He leak after successive testing at **400°C**
- **Below 10 scfh**, high recovery (> 90%) and no significant/additional inhibiting effect of ~19% steam or CO on H<sub>2</sub> flux
- **Permeate: H<sub>2</sub> only, no other gases were detected**
- **Retentate: High-pressure CO<sub>2</sub>**

# Long-term WGS CMR<sub>13.1</sub> $\mu\text{m}$ Pd



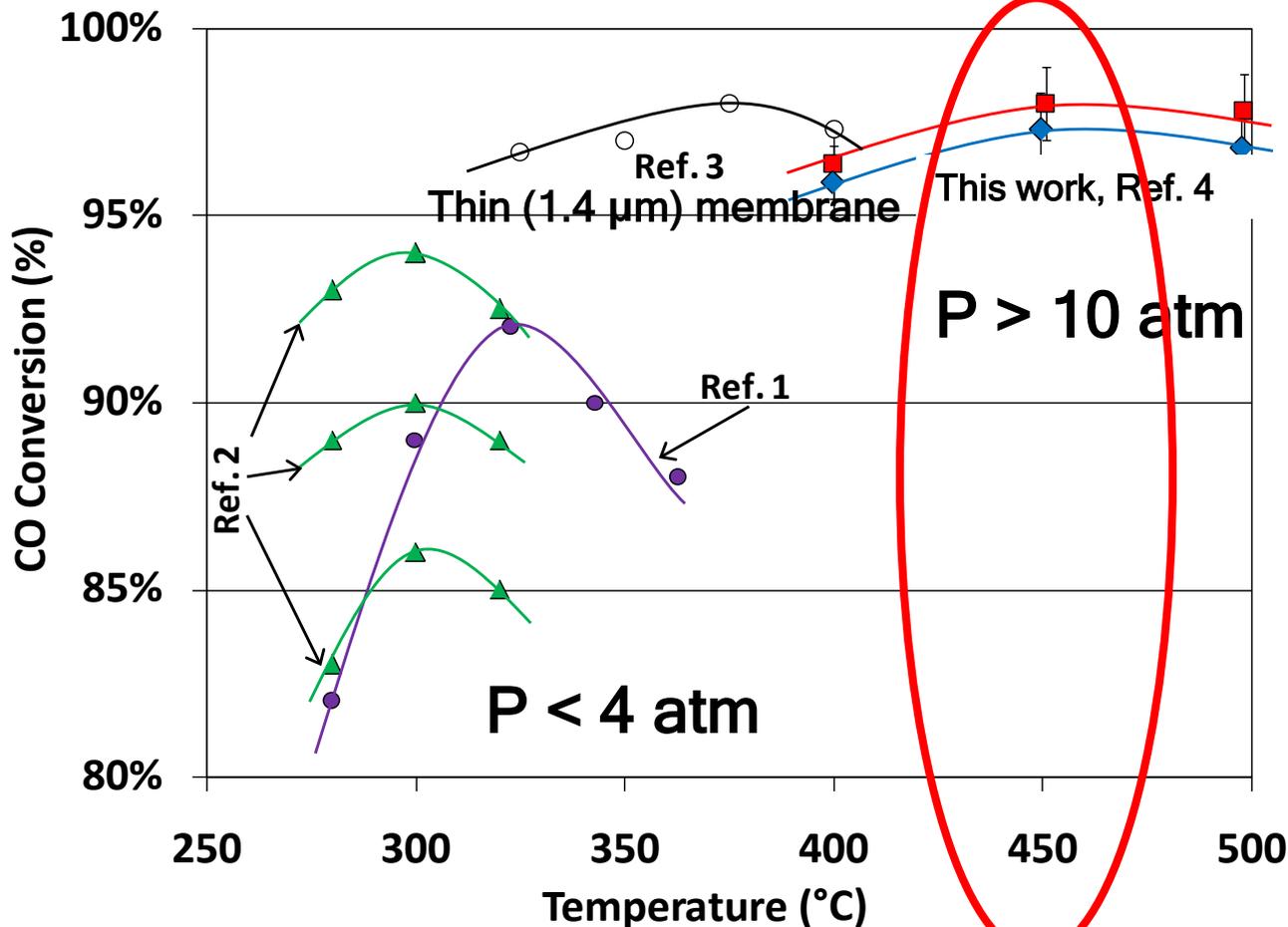
Feed Conditions	
22.7% CO, 22.0% H <sub>2</sub> , 9.9% CO <sub>2</sub> , 45.4% H <sub>2</sub> O	
H <sub>2</sub> O/CO	2.0
GHSV (h <sup>-1</sup> ):	4500
T (°C):	450

Membrane: AA-8R	
Selectivity (F <sub>H<sub>2</sub></sub> /F <sub>He</sub> )	
Initial	4000
After WGS Experiment	400 (-90%)
Permeance (scfh/ft <sup>2</sup> psi <sup>0.5</sup> )	
Initial	27.9
After WGS Experiment	26.6 (-4.6%)

- Stable CO conversion and H<sub>2</sub> recovery were observed for up to 80 hours
- Stable H<sub>2</sub> permeance after WGS test
- Significant selectivity decline after test

Flux @ 450°C = 44.3 scfh/ft<sup>2</sup> ( $\Delta P$  of 14.7 psi)

# WGS CMR Temperature Dependence (existence of a maximum)



➤ Maximum in CO conversion results from balance between thermodynamic equil. and reaction kinetic

- At low temperatures
  - Low catalytic activity
  - High therm. equil. conv.
- At high temperatures
  - High catalytic activity
  - High permeance
  - Low therm. equil. conv.

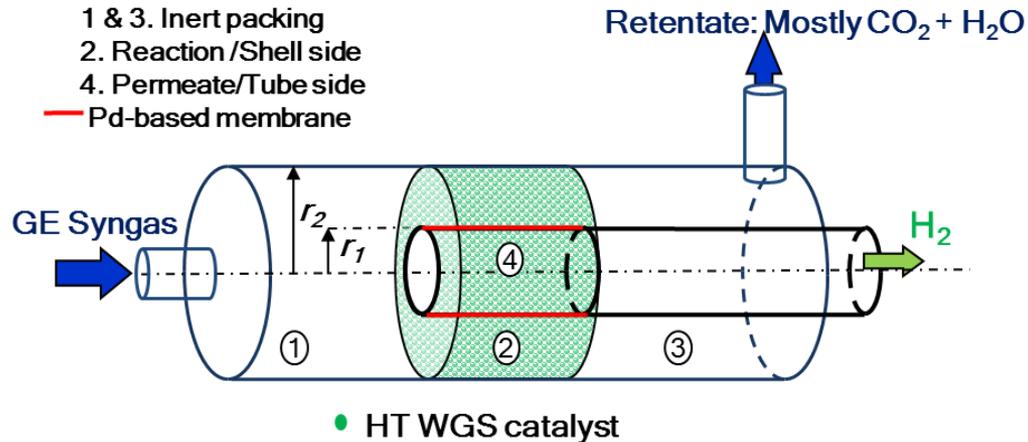
<sup>1</sup>Basile *et al.*, Gas Sep & Pur 1996; 10(4), 243-54.

<sup>2</sup>Barbieri *et al.*, J Power Sources 2008; 182, 160-7.

<sup>3</sup>Bi *et al.*, Int J H Energy 2009; 34, 2965-71.

<sup>4</sup>Augustine et al. Int. J. H. Energy, 2011, accepted for publication. Published online

# 2D Membrane Reactor Model



GE Syngas Composition (Dry):  
45% CO + 38% H<sub>2</sub> + 17% CO<sub>2</sub>

Properties of Pd/Inconel Membrane:  
 $Q_o = 5409 \text{ ft}^3 \cdot \mu\text{m}/(\text{ft}^2 \cdot \text{h} \cdot \text{psi}^{0.5})$   
 Pd Thickness = 10  $\mu\text{m}$   
 $E_p = 15.6 \text{ kJ/mol}$   
 Selectivity (H<sub>2</sub>/He) =  $\infty$

Mass balance equations at the unsteady state conditions:

$$\frac{\partial}{\partial t} \left( \frac{F_{H_2}}{F_{Total}} \right) = \frac{1}{C_{Total}^{Shell}} \left[ -\frac{\partial F_{H_2}}{\partial v^{Shell}} + \frac{1}{\Delta v^{Shell}} \int_{W_y}^{W_{y+\Delta y}} r_{H_2} dw - \frac{Q \cdot A_m}{l \cdot \Delta v^{Shell}} \cdot \Delta P_{H_2}^{0.5} \right] \quad \text{Reaction side (1)}$$

$$\frac{\partial}{\partial t} \left( \frac{F_{H_2}}{F_{Total}} \right) = \frac{1}{C_{Total}^{Tube}} \left[ -\frac{\partial F_{H_2}}{\partial v^{Tube}} + \frac{Q \cdot A_m}{l \cdot \Delta v^{Tube}} \cdot \Delta P_{H_2}^{0.5} \right] \quad \text{Permeate side (2)}$$

Energy balance equation in the reaction side at the unsteady state conditions

$$\left( \sum_{i=1}^n C_i c_{pt} \right) \frac{\partial T}{\partial t} = - \left( \sum_{i=1}^n F_i c_{pt} \right) \frac{\partial T}{\Delta v_{shell}} + (-r_{CO}) [-\Delta H_{Rxn}(T)] \quad (3)$$

Two dimensional mass balance equations at the steady state and isothermal conditions:

$$\frac{\partial(u \cdot C_i)}{\partial y} - \frac{D_{ef,i}}{u} \left[ \frac{\partial^2(u \cdot C_i)}{\partial r^2} + \frac{1}{r} \frac{\partial(u \cdot C_i)}{\partial r} \right] + \rho_{bulk,cat} R_i = 0 \quad (4)$$

# Process Intensification

➤ Performance target levels:  $X_{CO} = 98\%$  &  $R_{H_2} = 95\%$

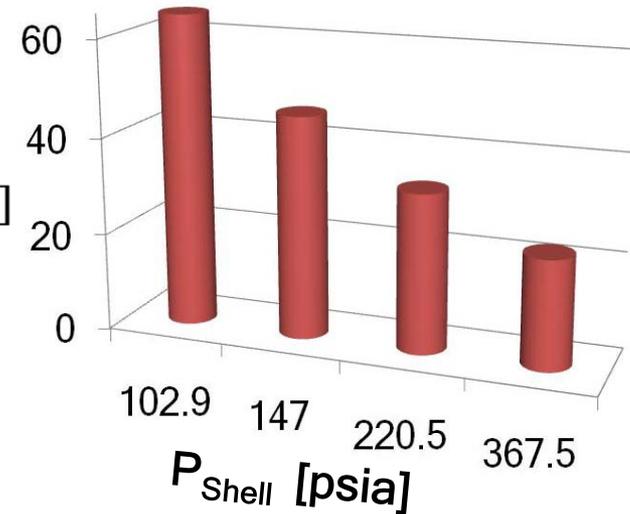
For the non-isothermal MR:  $T_{Rxn,Max} = 500^\circ C$  &  $T_{Rxn,Min} = 300^\circ C$  for pure Pd

## Isothermal Case:

$P_{shell} = 220$  &  $P_{Tube,MR} = 6$  psia  
 $F_{Dry\ feed} = 1.1$  scfh,  $T_{Rxn} = 400^\circ C$

	H <sub>2</sub> O:CO	X <sub>CO</sub> [%]	R <sub>H<sub>2</sub></sub> [%]	F <sub>H<sub>2</sub></sub> Ratio MR / PBR	W <sub>catalyst</sub> Ratio
MR	2	99	96	1.2	1
PBR	9	98	-		

$\frac{V_{MR}}{V_{PBR}}$  [%]

## Non-isothermal Case:

$P_{Shell} = 300$  &  $P_{Tube,MR} = 6$  psia,  $F_{Dry\ feed} = 3.4$  scfh

	H <sub>2</sub> O:CO	X <sub>CO</sub> [%]	R <sub>H<sub>2</sub></sub> [%]	T <sub>Rxn,Max</sub> [°C]	F <sub>H<sub>2</sub></sub> Ratio MR / PBR	Volume Ratio MR/PBR	W <sub>catalyst</sub> Ratio
MR	4	99.5	95	477	1.2	0.6	0.6
PBR	9	98	-	395			

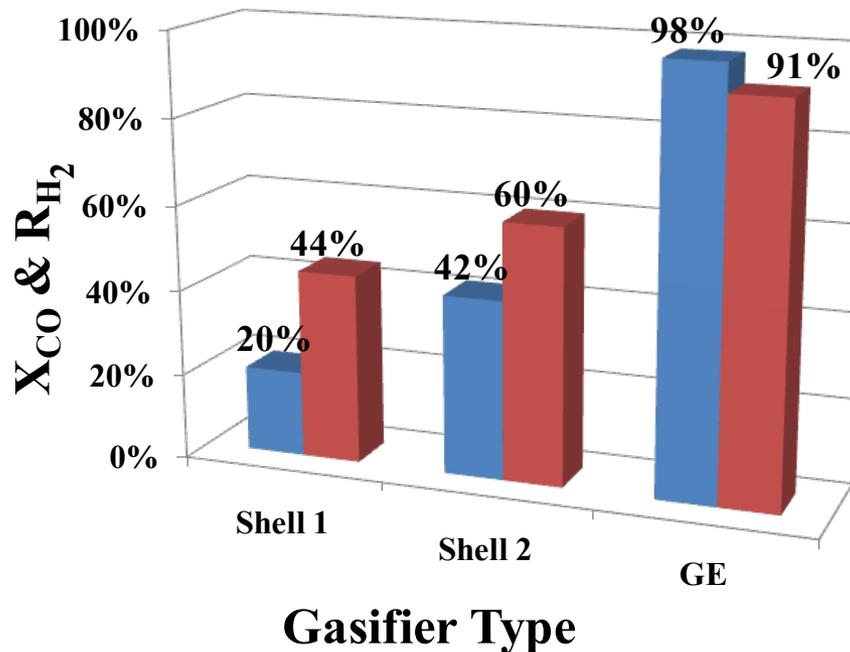
# Pd/Alloy Membrane Reactors in IGCC Plants

Table 1. Syngas compositions†

	Shell 1	Shell 2	GE
Scrubber T [°C] →	128	160	243
CO	56.4	49.6	15.6
H <sub>2</sub>	29.7	26.3	15.1
CO <sub>2</sub>	1.4	1.3	7.3
H <sub>2</sub> O	7	18.1	61
N <sub>2</sub>	4.53	3.86	0.8
Ar	0.7	0.6	
H <sub>2</sub> S	0.24	0.21	0.12
COS	0.02	0.02	
other	0.01	0.01	0.08

## GE Gasifier

- Highest H<sub>2</sub>O:CO ratio
- Highest gasifier pressure
- Highest X<sub>CO</sub> and R<sub>H<sub>2</sub></sub>
- Lowest H<sub>2</sub>S +COS concentration

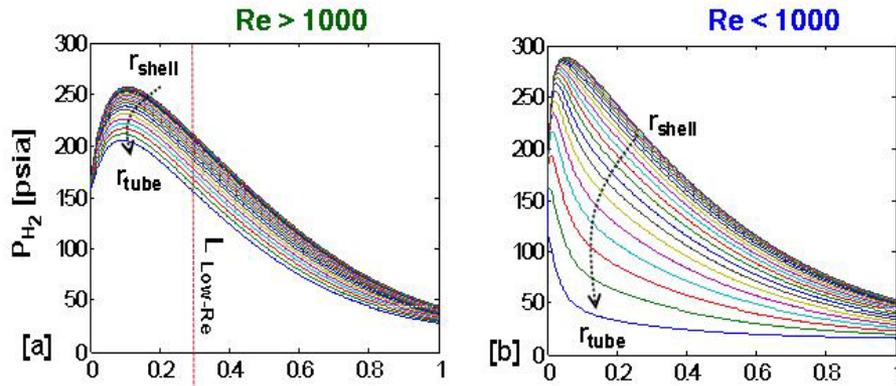


■ CO Conversion    ■ H<sub>2</sub> Recovery

$P_{\text{shell}} = 220$  &  $P_{\text{Tube,MR}} = 15$  psia

$F_{\text{feed}} = 1.1$  scfh ,  $T_{\text{Rxn}} = 450^\circ\text{C}$

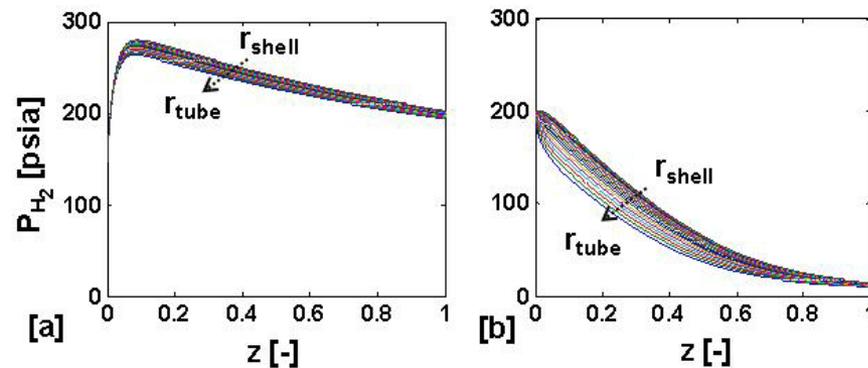
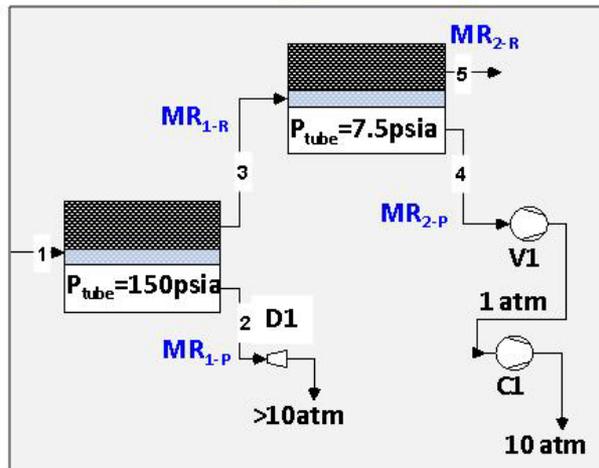
# Two-Dimensional Isothermal Model Results



- Good match between the experimental and modeling results for both  $X_{CO}$  &  $R_{H_2}$
- Operate  $Re > 1000$ , use longer tubes
- $Total Pd Area_{Re > 1000} < Total Pd Area_{Re < 1000}$

$T = 450^\circ C$ ,  $P_{shell} = 735 psia$ ,  $P_{tube} = 15 psia$ ,  $H_2O:CO = 2$

## MR Configuration Options



$T = 400^\circ C$ ,  $P_{shell} = 735 psia$ ,  $H_2O:CO = 2$   
2 in OD  $\times$  31 ft L membrane tubes

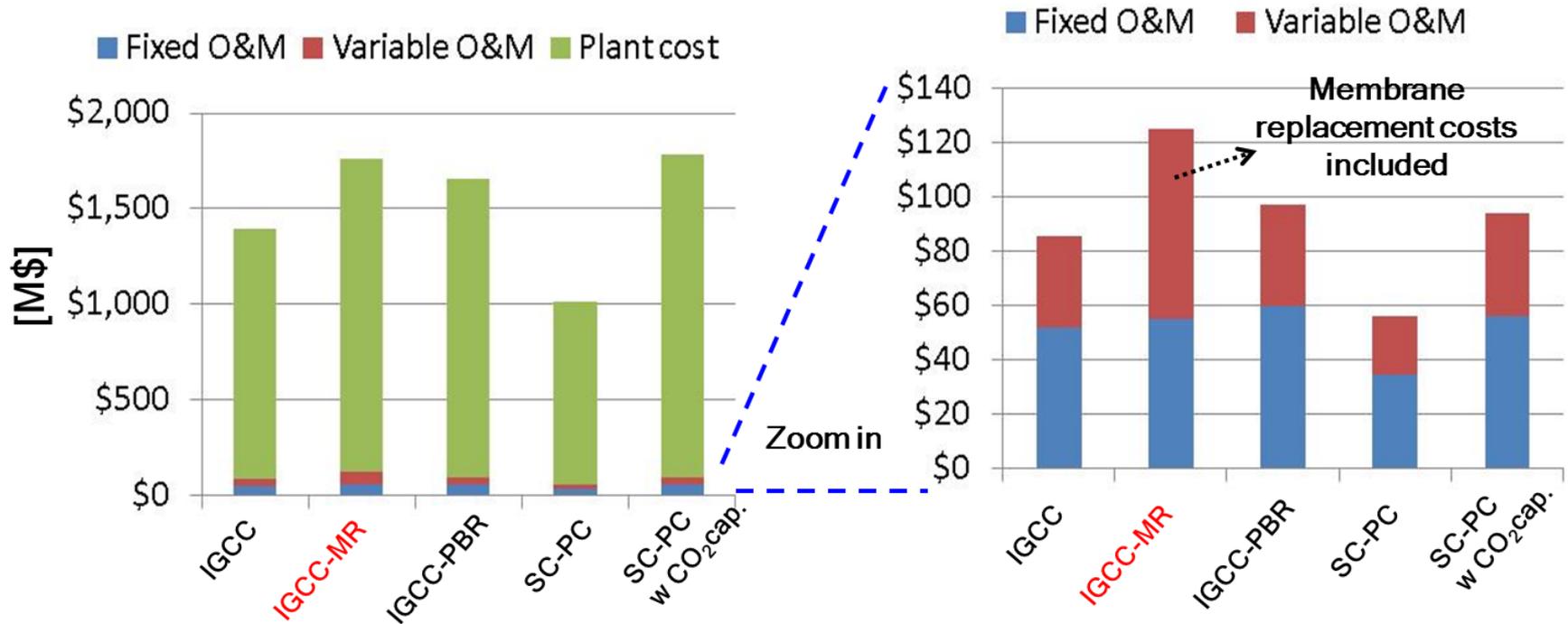
$X_{CO}$ [%]	98.7
$R_{H_2}$ [%]	97.1
$H_2$ tpd	~737

# Capital - O&M Costs

Target: Coproduction of hydrogen and power by using Pd-based membrane reactors ( Mitretek TR 2002-31)

Cost analysis is based on GEE radiant only IGCC power-plant (DOE/NETL 2010/1397) + Pd/Inconel MR

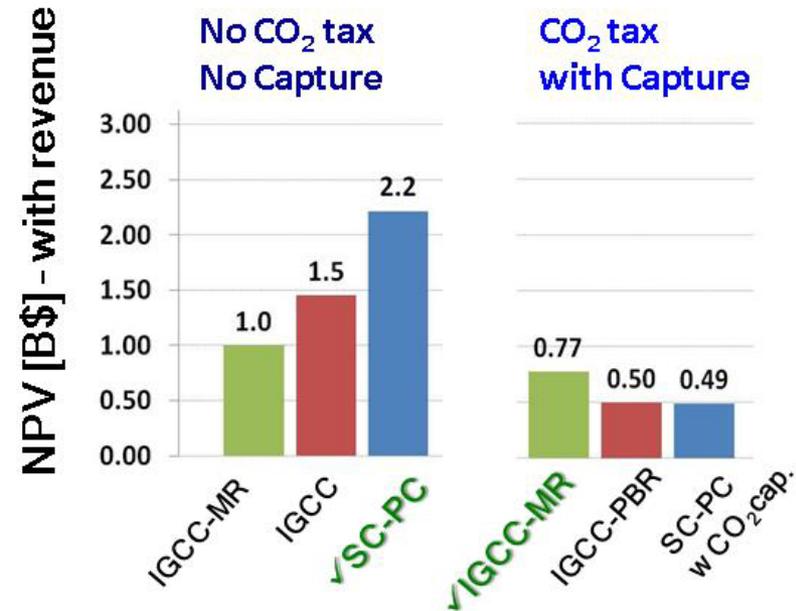
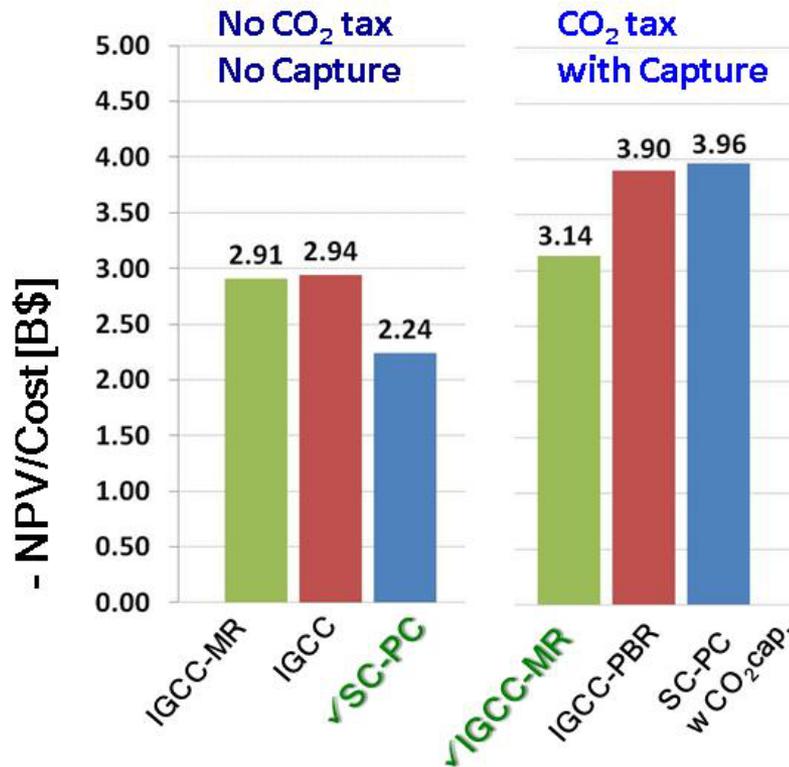
Assumption : 1 year membrane lifetime



Dimension (each)	2 in OD × 25 ft L
Total Pd Area	~118000 ft <sup>2</sup>
Total H <sub>2</sub> Production	744 tonne/day

Membrane reactor module:  
 Equipment cost : 533 \$/ft<sup>2</sup>  
 Fixed capital investment : 1004 \$/ft<sup>2</sup>  
 Total capital investment : 1254 \$/ft<sup>2</sup>

# Net Present Value (NPV) Analysis

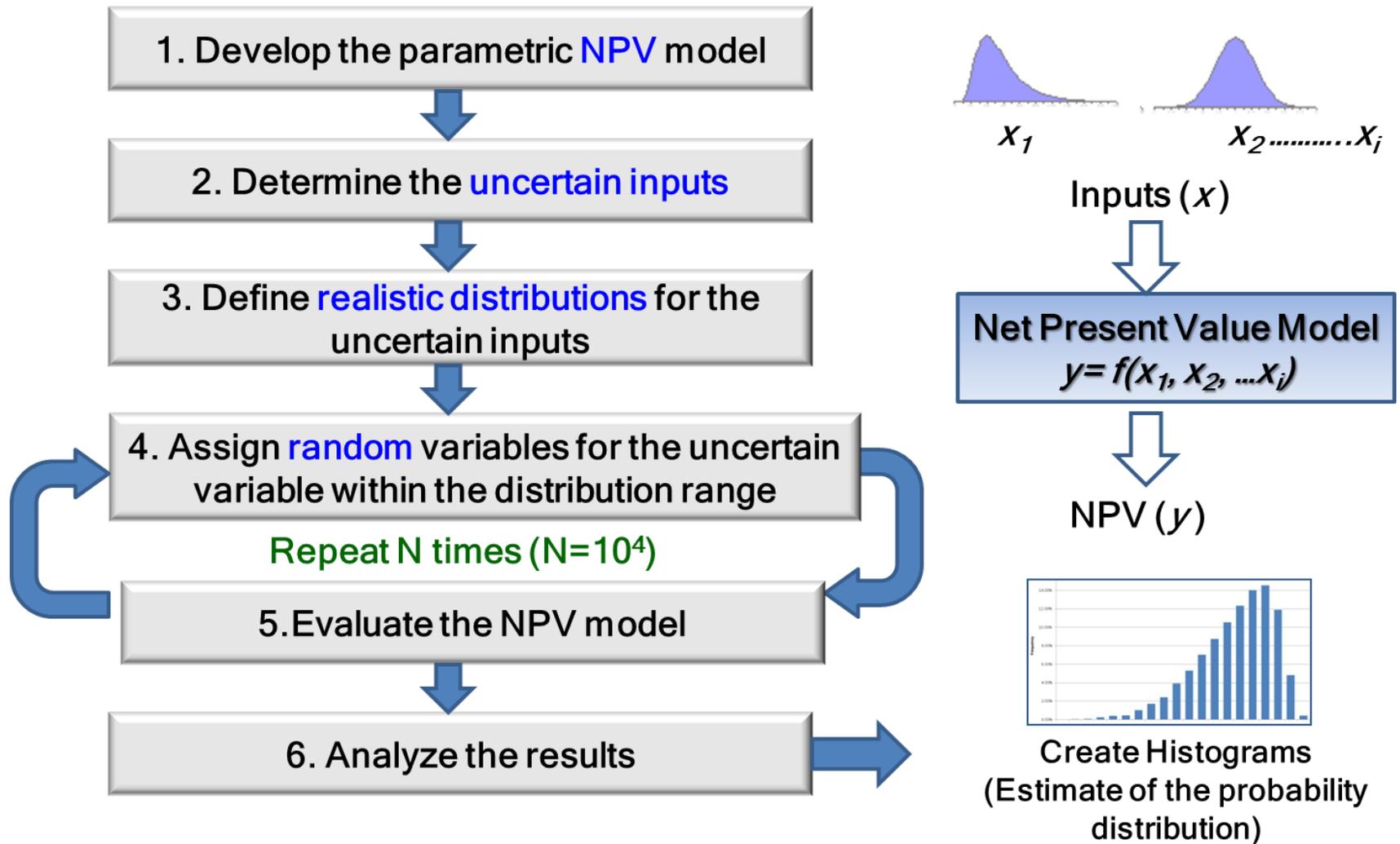


## Assumptions:

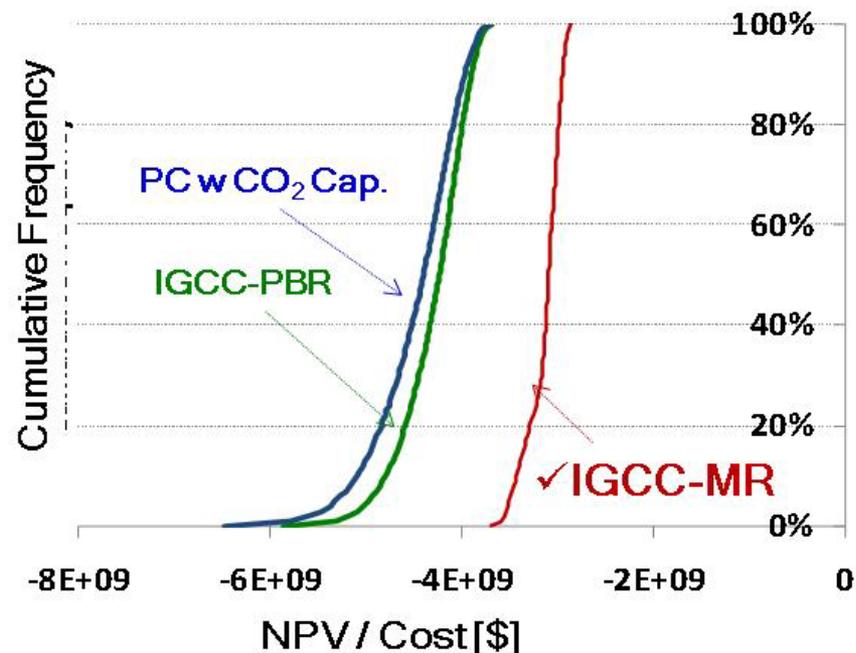
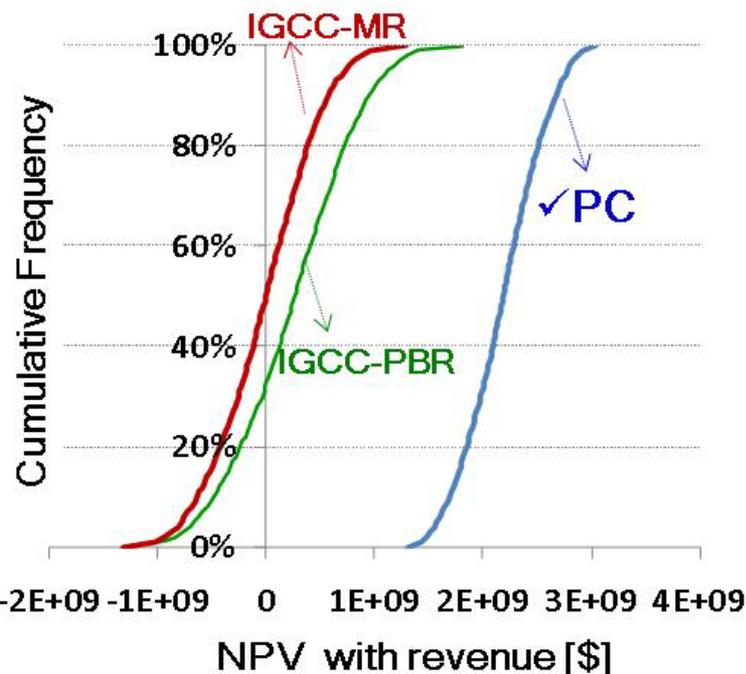
- 40 years plant lifetime
- 1 year membrane lifetime
- Discount rate = 9% for MR 6% for the others
- Inflation rate = 2.5% (U.S. Dep. of Lab. Bureau)
- 25\$/tCO<sub>2</sub> tax, starting in 2015 (MIT, "Future of Coal")
- Electricity selling price = 9 ¢/kWh (U.S. Energy Inf. Adm.)

- Flaw of averages
- Risk in the uncertain inputs
- Generate a more realistic **distribution** of the plant's economic value rather than a **single-point/estimate**

# Monte Carlo simulations for uncertainties effect on NPV



# Economic Evaluation in the Presence of Uncertainty with Monte Carlo Simulation



## Current Situation

- No CO<sub>2</sub> tax
- PC capacity factor = 85%

## Uncertainty

- IGCC-MR & IGCC-PBR capacity factor = 40-80%
- Electricity selling price:  
min = 6.6, most likely = 8.4, max = 9.9 ¢/kWh

## Regulatory action on carbon emissions after 2015

### Uncertainty

- Initial CO<sub>2</sub> tax:  
min = 0, most likely = 25, max = 100 \$/tCO<sub>2</sub>
- CO<sub>2</sub> tax growth rate:  
min = 2%, most likely = 5%, max = 8%
- Capacity factor = 60-85% for IGCC-PBR&MR  
70-85% for PCw CO<sub>2</sub> cap.
- Pd price: Historical data

# Collaborations

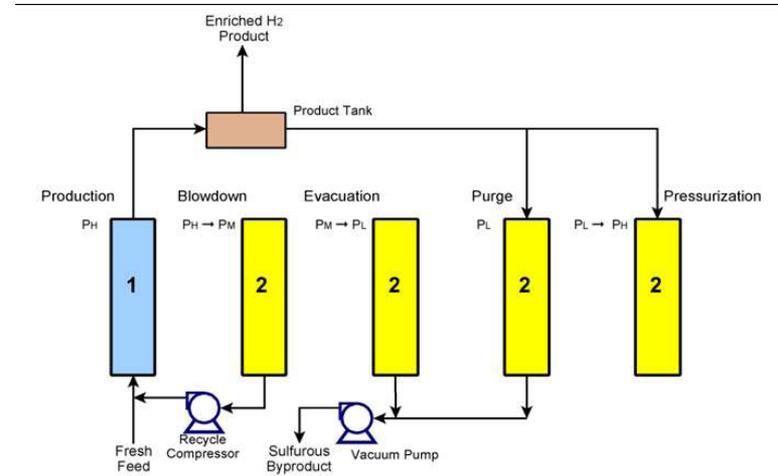
## Adsorption Research Inc. (ARI); sub

- Zeolite 5A and Hisiv 3000 were chosen for all tests
- Feed H<sub>2</sub>S Concentration: 500 to 1,000 ppm(v)
  - Light Product = 98+% Helium (H<sub>2</sub>) Recovery.
  - Light Product = Either <1 or <20 ppm H<sub>2</sub>S.
- Feed: 500 ppm H<sub>2</sub>S + 500 ppm COS ≈ Same Performance as 1,000 ppm H<sub>2</sub>S.
- Feed H<sub>2</sub>S Concentration: 10,000 ppm(v)
  - Apparent Working Capacity Was Reduced.
  - Light Product = 97% Helium (H<sub>2</sub>) Recovery.
  - Light Product = 1.4 ppm H<sub>2</sub>S.
- CO<sub>2</sub> Recovery in Light Product Varied According To The Allowable H<sub>2</sub>S Concentration in Light Product.

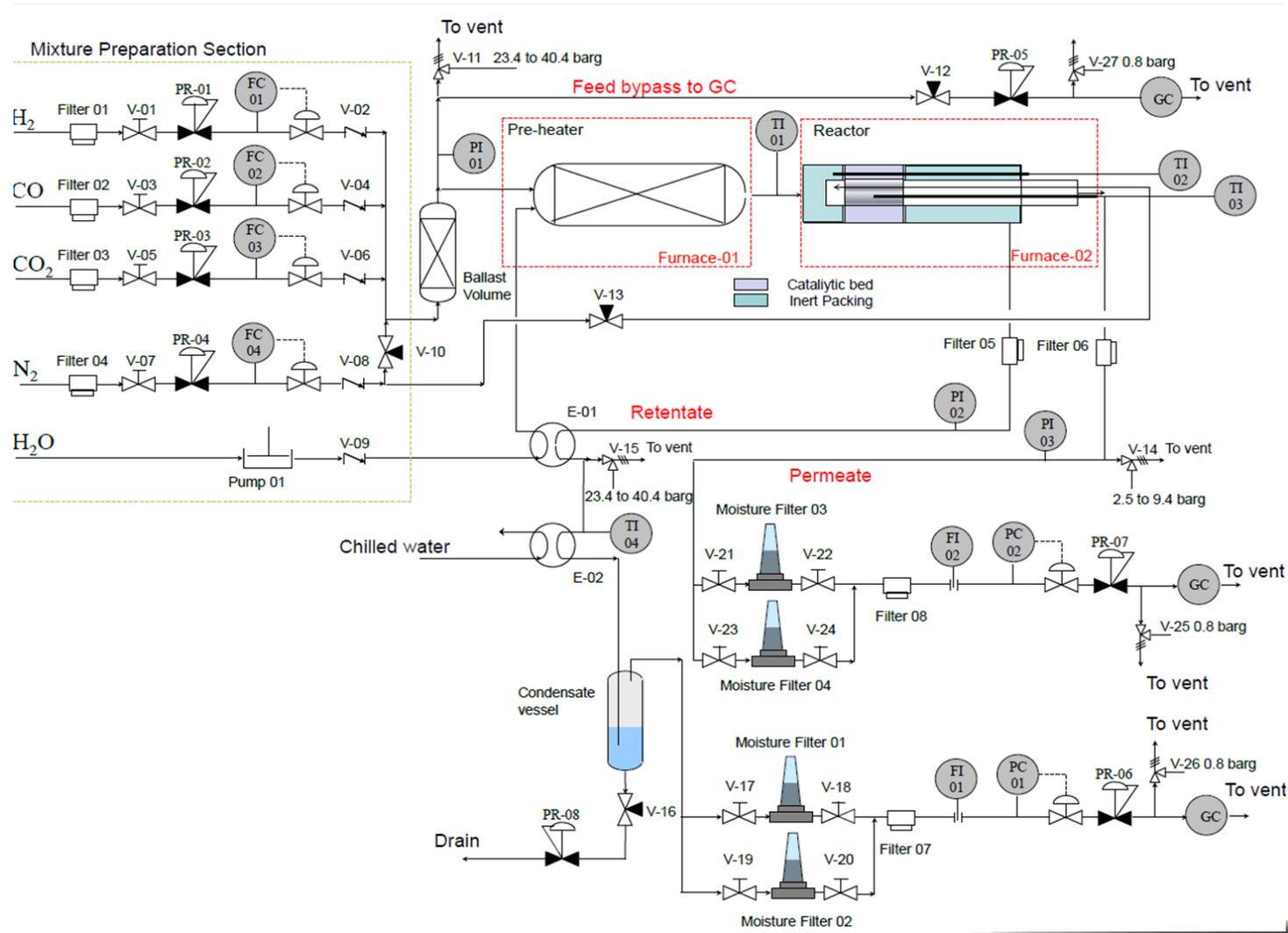
### •General conclusion:

PSA system and the cycle developed are promising however the loss of CO<sub>2</sub> for carbon capture is a prohibiting factor

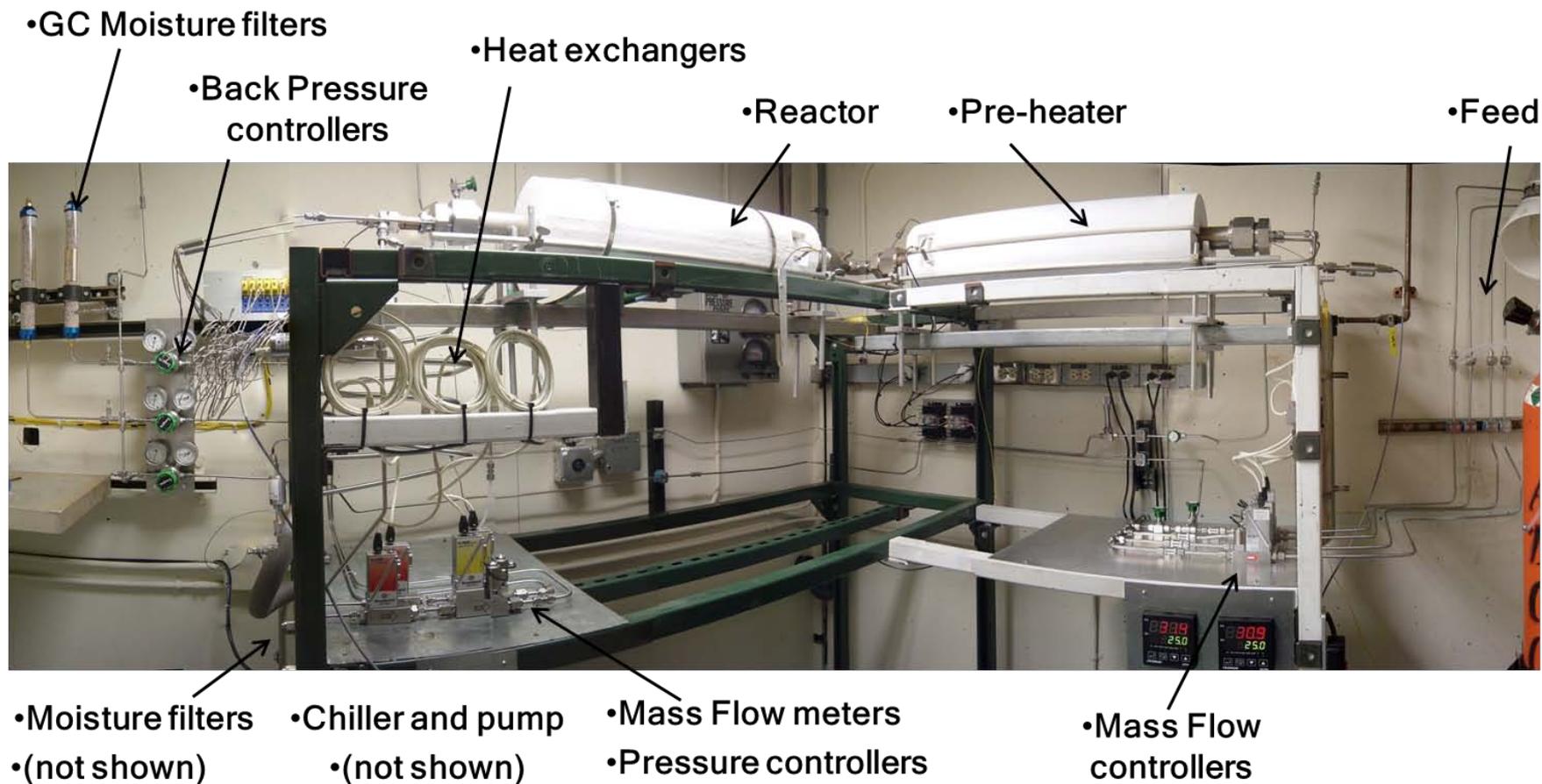
### Current PSA Cycle



# PID of a new WGS CMR testing Rig at WPI



# new WGS CMR testing Rig at WPI



## Proposed Future Work (under grant DE-FE0004895)

- Test new grading techniques on bare support surfaces to achieve 8  $\mu\text{m}$  Pd on any PSS support (alumina deposition)
- Test new Pd deposition methods (e. g. electrodeposition) for selectivity stability
- Continue long term WGS reaction study (an 1000 h run is planned) for both 1/2" OD and 1" OD membranes
- Assess the Syngas effect on selectivity stability
- Continue Pd/Au alloying studies to improve H<sub>2</sub> flux
- Study new coatings for Hydrogen sulfide poisoning inhibition
- Continue long-term sulfur poisoning & recovery experiments

# Project Summary

- Membranes prepared by sequential annealing/polishing (pAP method) -with thickness higher than 7-8  $\mu\text{m}$  have a stable selectivity over thousands of hours at 400-450°C (029/030/31/033) with permeances limited by mass transfer resistance in the support
- Flux of  $\sim 359$  scfh/ft<sup>2</sup>, which exceeded the DOE's 2015 H<sub>2</sub> flux targets [T=442°C &  $\Delta P$  of 100 psi (with  $P_{\text{Low}}=15$  psia)] was achieved but low selectivity H<sub>2</sub>/He ( $\sim 500$ ) due to very low thickness (3-5  $\mu\text{m}$ )
- Calculation made to establish that the minimum initial He permeance of 130 scfh /((ft<sup>2</sup>-psi) for the porous metal supports in order to achieve DOE's 2015 target with a 7-8  $\mu\text{m}$  membrane at 450°C
- Completed the first WGS testing rig for 1"OD membranes at WPI
- Demonstrated that:
  - WGS reaction with high temperature catalyst (HIFUEL) occurs optimally at a temperature of  $\approx 450^\circ\text{C}$
  - Effect of pressure on leak stability negligible
  - 97% CO conversion and 80% H<sub>2</sub> recovery achieved in an 18.1  $\mu\text{m}$  thick Pd-based CMR operated at 450°C,  $\Delta P=200$  psi ( $P_{\text{Low}}=15$  psia) and  $\text{GHSV}_{\text{stp}} = 2900$  h<sup>-1</sup>, **exceeding the equilibrium conversion of 93%**.

# Project Summary

- **Successfully completed steady-state MSR & WGS reaction 2D modeling studies & process intensification analysis:**
  - A 2D model was developed to take into account radial gradients. Several configurations were modeled to achieve  $X_{CO} > 98\%$  and  $R_{H_2} > 95\%$  for the isothermal case.
- **Successfully completed process intensification, safety and economic analysis:**
  - Safety boundaries for WGS feed gas composition, feed temperature, catalyst density have been established for the adiabatic case (in case cooling fails) to achieve  $X_{CO} > 98\%$  and  $R_{H_2} > 95\%$  keeping membrane temperature lower than  $500^\circ\text{C}$ .
  - Capital -O&M costs was calculated to be around  $1254 \text{ \$/ft}^2$  very close to DOE target ( $1000 \text{ \$/ft}^2$  )
  - NPV analysis showed that if  $\text{CO}_2$  taxes were implemented, IGCC-MR with carbon capture would be the preferred configuration.
  - Monte-Carlo analysis was used to study the variability of 11 uncertainties and proved that with taxes on  $\text{CO}_2$  emission, IGCC-MR configuration would be the preferred choice.
- **Completed all simulations, tests of PSA cycles with commercial adsorbents and feeds including all reactants plus  $\text{H}_2\text{S}$  and  $\text{COS}$  (Sub, ARI).**
- **PSA for hydrogen sulfide removal is however not included in the process intensification due to the high removal loss of  $\text{CO}_2$**

# Project Summary Table: Permeation Results

	DOE Targets		Current WPI Membranes (1/2" OD, 2.5" Length, ~24 cm <sup>2</sup> )					
	2010	2015	#025R	#027	#029	#030	#031	#032
Flux [scfh/ft <sup>2</sup> ]	200	300	65.9	36.1	166	178	26.6	359
$\Delta P$ (psi) H <sub>2</sub> partial pressure (P <sub>Low</sub> =15 psia)	100*	100*	15	15	100	102	15	100
Temperature [°C]	300-600	250-500	400	400	450	442	450	442
H <sub>2</sub> /He Selectivity	n/a	n/a	~220	~120	$\infty$	10000	~4500	~450
Total Test Duration [hours]	n/a	n/a	1015	~1250	~4500	~1400	~2200	~523
Thickness [ $\mu$ m]	n/a	n/a	4.2 Pd	6.2 Pd/Au <sub>5 wt%</sub>	7.6 Pd	7.9 Pd	7.0 Pd	3-5 Pd
WGS Activity	Yes	Yes	Not tested	Not tested	Not tested	Not tested	Not tested	Not tested
CO Tolerance	Yes	Yes	Not tested	Not tested	Yes	Not tested	Not tested	Not tested
S Tolerance [ppm]	20	>100	Not tested	Not tested	Not tested	Not tested	Not tested	Not tested
H <sub>2</sub> Purity	99.5%	99.99%	99.0%	99.5%	$\geq 99.999\%$	$\geq 99.99\%$	99.98%	99.8%
$\Delta P$ Operating Capability (Max. Sys. Pressure, psi)	400	800-1000	15**	15**	225**	102**	15**	100**

§ DOE-NETL Test Protocol v7 - 05/10/2008, \* Standard conditions are 150 psia hydrogen feed pressure and 50 psia hydrogen sweep pressure

# Project Summary Table: Mixed Gas & WGS Reaction Results

	DOE Targets		Current WPI Membranes (1/2" OD, 2.5" Length, ~24 cm <sup>2</sup> )				
	2010	2015	AA-4R*	AA-5R*	AA-6R*	AA-7R*	AA-8R*
Flux [scfh/ft <sup>2</sup> ]	200	300	262.3	108.6	427.0	98.1	96.4
$\Delta P$ (psi) H <sub>2</sub> partial pressure (P <sub>Low</sub> =15 psia)	100**	100**	245.1	71.0	222.7	45.4	37.1
Temperature [°C]	300-600	250-500	400	450	450	450	450
H <sub>2</sub> /He Selectivity	n/a	n/a	71,000	2,800	1,100	25	670
Total Test Duration [hours]	n/a	n/a	1,030	1,080	860	350	970
Thickness [ $\mu$ m]	n/a	n/a	14.4	18.1	18.1	14.3	13.4
WGS Activity	Yes	Yes	Not tested	w/ packed catalyst	Not tested	w/ packed catalyst	w/ packed catalyst
CO Tolerance	Yes	Yes	Yes	Yes	Yes	Yes	Yes
S Tolerance [ppm]	20	>100	Not tested	Not tested	Not tested	Not tested	Not tested
H <sub>2</sub> Purity	99.5%	99.99%	99.99%	99.96%	99.91%	96.2%	99.85%
$\Delta P$ Operating Capability (Max. System Pressure, psi)	400	800-1000	250	250	250	250	250

§ DOE-NETL Test Protocol v7 - 05/10/2008

\* R - repaired by mechanical treatment and Pd plating

\*\* Standard conditions are 150 psia hydrogen feed pressure and 50 psia hydrogen sweep pressure

# Project Summary Table: Mixed Gas & WGS Reaction Results

	DOE Targets		Current WPI Membranes (1/2" OD, 2.5" Length, ~24 cm <sup>2</sup> )				
	2010	2015	AA-12R	AA-14	AA-18	AA-21R	AA-22
Flux [scfh/ft <sup>2</sup> ]	200	300	550.5	76.4	76.0	79.6	86.3
$\Delta P$ (psi) H <sub>2</sub> partial pressure (P <sub>Low</sub> =15 psia)	100**	100**	300	29	57	48	28
Temperature [°C]	300-600	250-500	450	450	450	400	450
H <sub>2</sub> /He Selectivity	n/a	n/a	240	190	998	176	80
Total Test Duration [hours]	n/a	n/a	150	360	400	864	144
Thickness [ $\mu$ m]	n/a	n/a	12.7	11.4	19.6	22.0	13.5
WGS Activity	Yes	Yes	Not tested	Not tested	Not tested	Not tested	Not tested
CO Tolerance	Yes	Yes	Not tested	Not tested	Not tested	Not tested	Not tested
S Tolerance [ppm]	20	>100	Not tested	Not tested	Not tested	Not tested	Not tested
H <sub>2</sub> Purity	99.5%	99.99%	99.6%	99.5%	99.9%	99.4%	98.8%
$\Delta P$ Operating Capability (Max. System Pressure, psi)	400	800-1000	400	250	250	400	250

§ DOE-NETL Test Protocol v7 - 05/10/2008

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# Project Summary Table: Mixed Gas & WGS Reaction Results

	DOE Targets		Current WPI Membranes (1/2" OD, 2.5" Length, ~24 cm <sup>2</sup> )				
	2010	2015	AA-24	AA-25	AA-26		
Flux [scfh/ft <sup>2</sup> ]	200	300	84.6	59	51.2		
$\Delta P$ (psi) H <sub>2</sub> partial pressure (P <sub>Low</sub> =15 psia)	100**	100**	37.8	16	23		
Temperature [°C]	300-600	250-500	400	450	500		
H <sub>2</sub> /He Selectivity	n/a	n/a	360	78	233		
Total Test Duration [hours]	n/a	n/a	1220	170	288		
Thickness [ $\mu$ m]	n/a	n/a	16.2	20.6	12.8		
WGS Activity	Yes	Yes	Yes	Not tested	Not tested		
CO Tolerance	Yes	Yes	Yes	Not tested	Not tested		
S Tolerance [ppm]	20	>100	Not tested	Not tested	Not tested		
H <sub>2</sub> Purity	99.5%	99.99%	99.7%	98.7%	99.6%		
$\Delta P$ Operating Capability (Max. System Pressure, psi)	400	800-1000	250	250	250		

§ DOE-NETL Test Protocol v7 - 05/10/2008

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\*\* Standard conditions are 150 psia hydrogen feed pressure and 50 psia hydrogen sweep pressure

# Project Summary Table: Mixed Gas & WGS Reaction Results

	DOE Targets		Current WPI Membranes (1/2" OD, 2.5" Length, ~24 cm <sup>2</sup> )				
	2010	2015	RK01	RK02	RK03	RK04	
Flux [scfh/ft <sup>2</sup> ]	200	300	41.3	82	60.7	83.8	
$\Delta P$ (psi) H <sub>2</sub> partial pressure (P <sub>Low</sub> =15 psia)	100**	100**	15	15	15	15	
Temperature [°C]	300-600	250-500	450	500	450	450	
H <sub>2</sub> /He Selectivity	n/a	n/a	$\infty$	112	2189	1000	
Total Test Duration [hours]	n/a	n/a	1023	373	1017	1250	
Thickness [ $\mu$ m]	n/a	n/a	14	30.8	22	13	
WGS Activity	Yes	Yes	Not tested	Not tested	Not tested	Not tested	
CO Tolerance	Yes	Yes	Not tested	Not tested	Not tested	Not tested	
S Tolerance [ppm]	20	>100	Not tested	Not tested	Not tested	Not tested	
H <sub>2</sub> Purity	99.5%	99.99%	99.9999%	99.11%	99.95%	99.90%	
$\Delta P$ Operating Capability (Max. System Pressure, psi)	400	800-1000	Not tested	Not tested	Not tested	Not tested	

§ DOE-NETL Test Protocol v7 - 05/10/2008

\* R - repaired by mechanical treatment and Pd plating

\*\* Standard conditions are 150 psia hydrogen feed pressure and 50 psia hydrogen sweep pressure