

*Analysis of Hydrogen and Co-Product Electricity  
Production from Coal with  
Near Zero Pollutant and CO<sub>2</sub> Emissions using an  
Inorganic Hydrogen Separation Membrane Reactor*

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# Large Scale Production of H<sub>2</sub> from Fossil Fuels

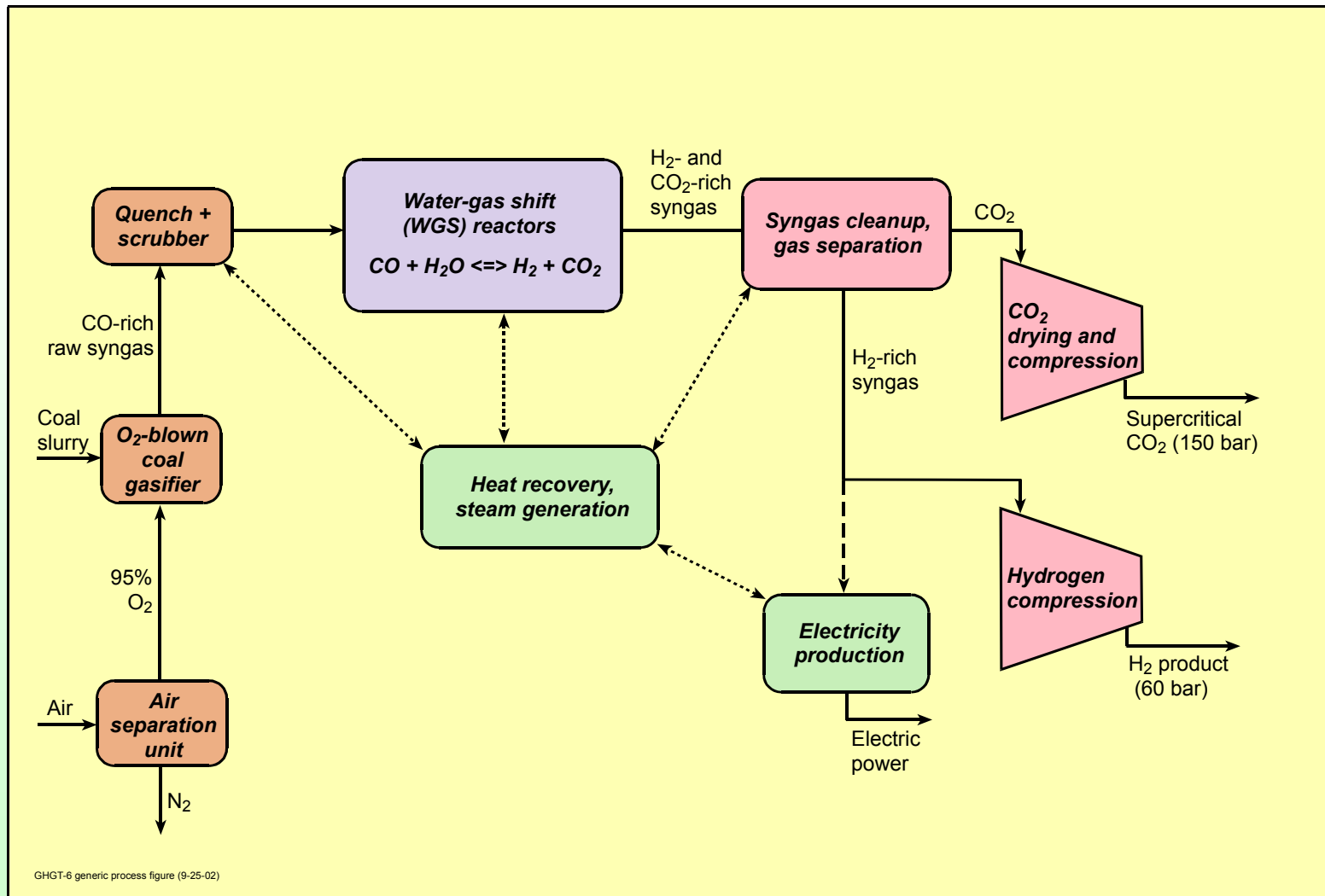
- Four Related Papers Presented Here -

	Natural Gas	Coal & Residuals
CO <sub>2</sub> Venting	Almost all H <sub>2</sub> produced today	Refineries, chemicals, NH <sub>3</sub> production in China 2) “Conventional technology”
CO <sub>2</sub> Capture	1) FTR vs. ATR with CC	2) “Conventional technology” 3) Membrane reactors 4) Overview

# *H<sub>2</sub> from Coal: Motivation*

- Distributed energy use (transportation and heating) responsible for ~2/3 of global CO<sub>2</sub> emissions
- CO<sub>2</sub> capture, compression, dehydration, and pipeline transport from distributed sources is *very* expensive.
- Low carbon energy carriers are needed: electricity...*and hydrogen?*
- If CO<sub>2</sub> sequestration is viable, fossil fuel decarbonization likely to be the cheapest route to electricity and hydrogen for many decades.
- Coal is of great interest because it is:
  - *Plentiful.* Resource ~ 500 years (vs. gas/oil: ~100 years).
  - *Inexpensive.* 1-1.5 \$/GJ HHV (vs. gas at 2.5+ \$/GJ).
  - *Ubiquitous.* Wide geographic distribution (vs. middle east).
  - *Clean?!* Gasification, esp. with sequestration, produces little gaseous emissions and a chemically stable, vitreous ash.
- *Example:* China: extensive coal resources; little oil and gas. Potential for huge emissions of both criteria pollutants and greenhouse gases.

# Generic Process: Coal to H<sub>2</sub>, Electricity, and CO<sub>2</sub>

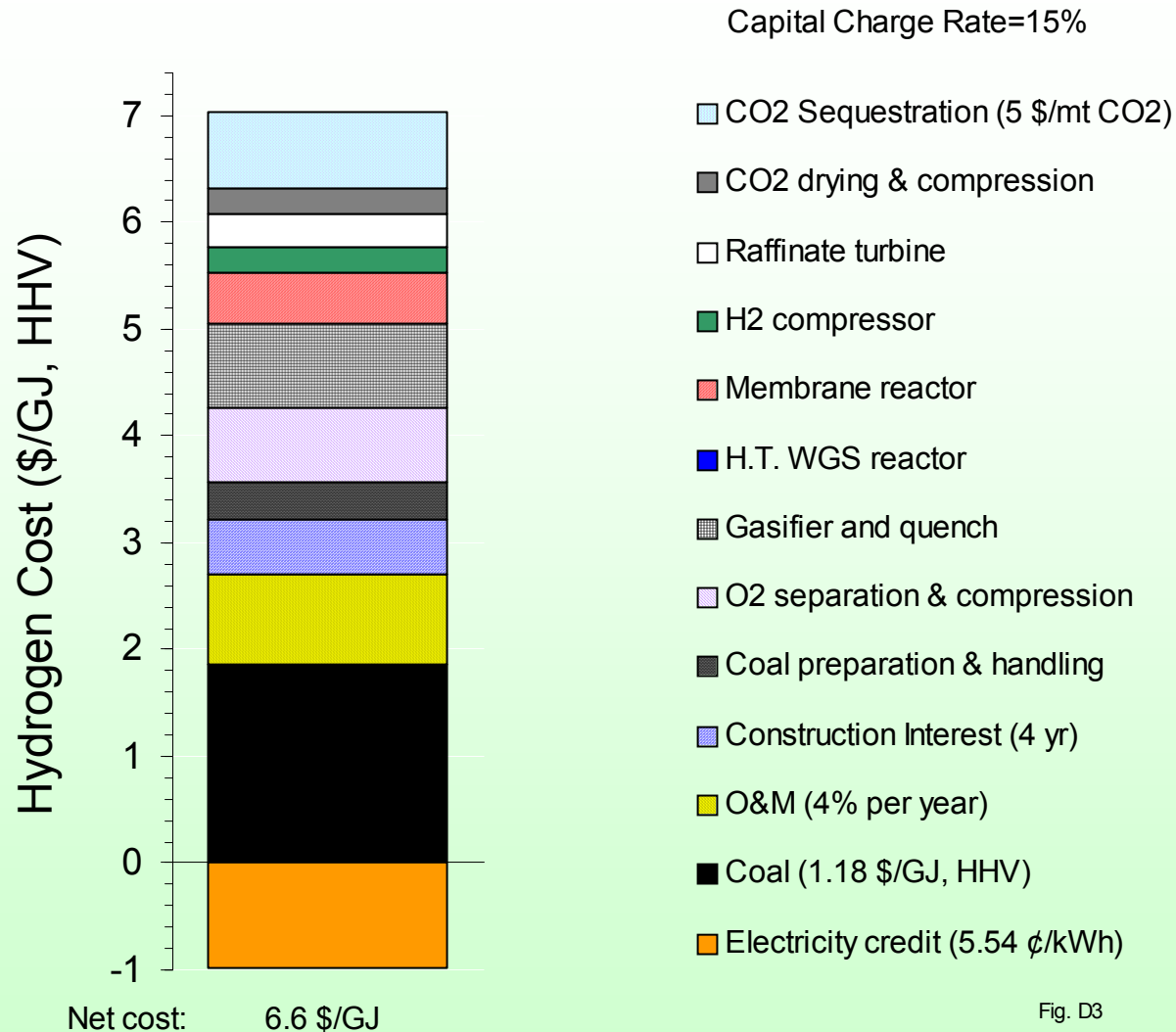


- All work presented here is based on O<sub>2</sub>-blown, entrained flow, coal gasification, primarily Texaco quench (some E-Gas).

# *Process Modeling*

- Heat and mass balances (around each system component) calculated using:
  - Aspen Plus (commercial software), and
  - GS (“Gas-Steam”, Politecnico di Milano)
- Membrane reactor performance calculated via custom Fortran codes
- Component capital cost estimates taken from the literature, esp. Holt, et al. and EPRI reports on IGCC
- Benchmarking/calibration:
  - Economics of IGCC with carbon capture studied by numerous groups
  - Used as a point of reference for performance and economics of our system
  - Many capital-intensive components are common between IGCC electricity and H<sub>2</sub> production systems (both conventional and membrane-based)

# Disaggregated Cost of H<sub>2</sub> Production



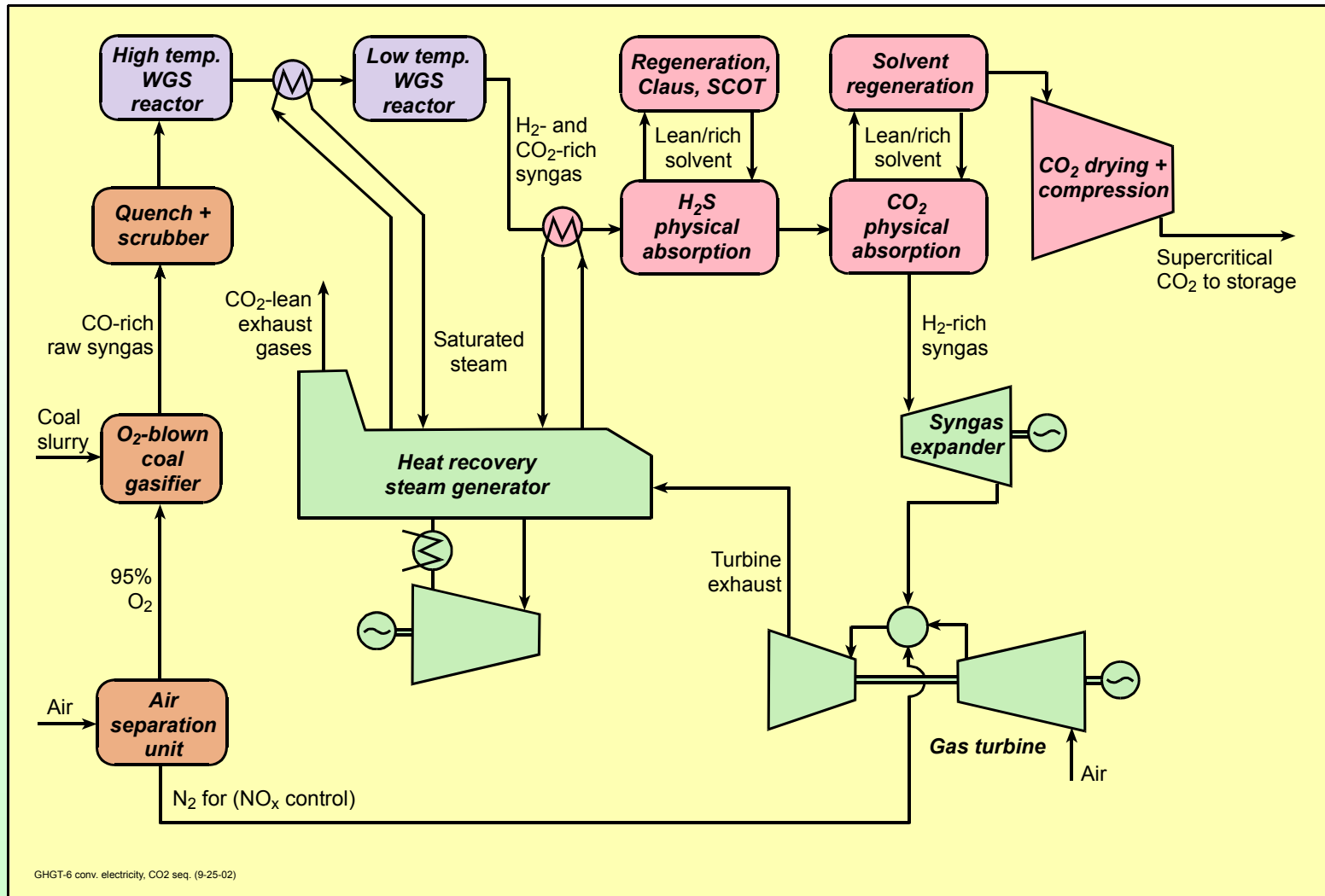
- 70 bar gasifier, 83.2% HRF, uncooled raffinate turbine, scale: 1 GW<sub>th</sub> H<sub>2</sub> (HHV)

# *Economic Assumptions*

Coal price (2000 average cost to electric generators)	1.18 \$/GJ (HHV)
Capacity factor	80%
Capital charge rate	15% per yr
Interest during construction	16.0% of overnight capital
O&M costs	4% of overnight capital per year
CO <sub>2</sub> storage cost *	5 \$/mt CO <sub>2</sub> (~0.7 \$/GJ H <sub>2</sub> HHV)
U.S. dollars valued in year	2002
Plant scale	1 GW <sub>th</sub> H <sub>2</sub>
Coal Type	Illinois #6

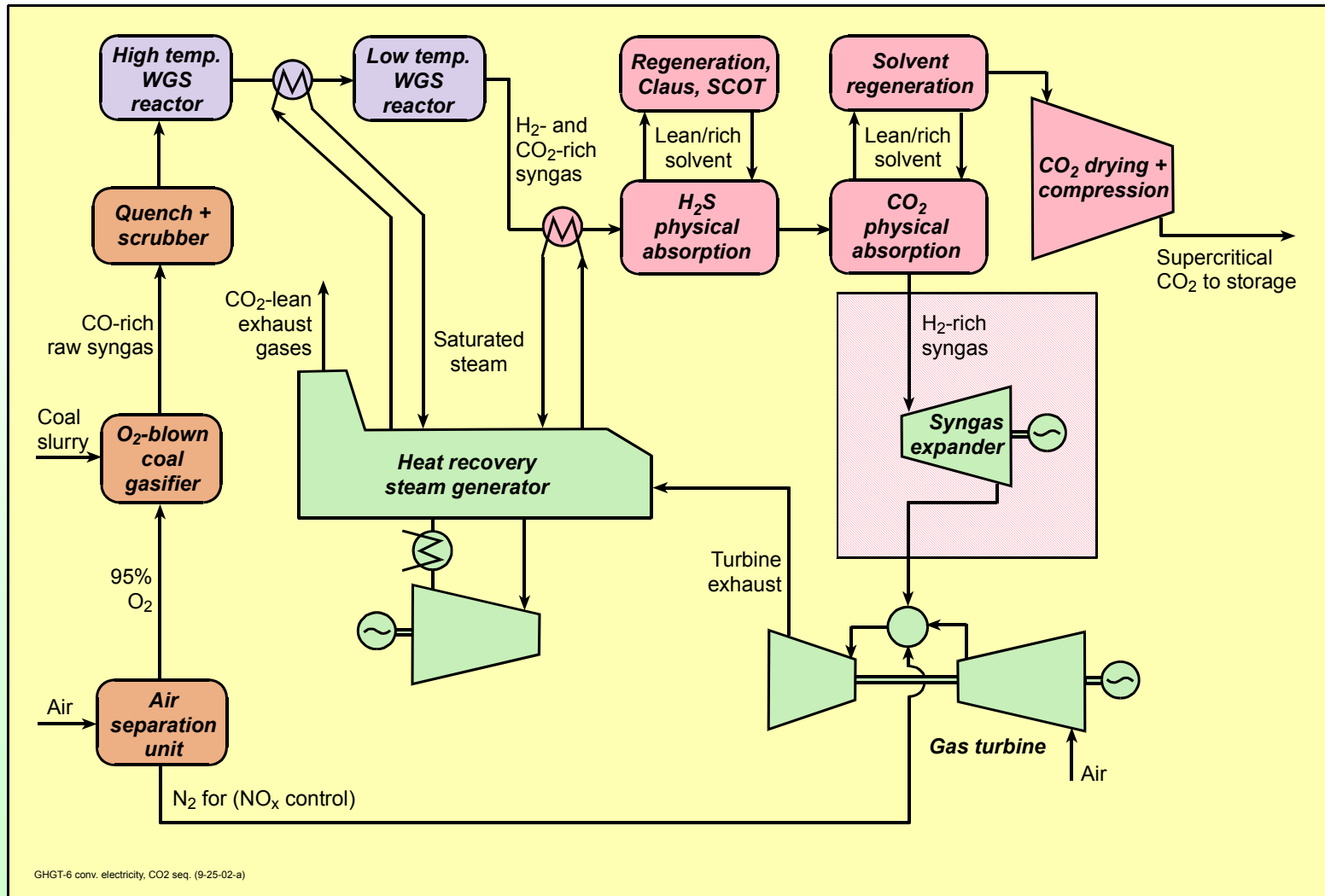
- “Best case” cost estimate for: 16,000 tonne/day CO<sub>2</sub>, 100 km pipeline, 2 km deep injection well (layer thickness > 50 m, permeability > 40 mDa)

# Benchmark: IGCC Electricity with CO<sub>2</sub> Capture



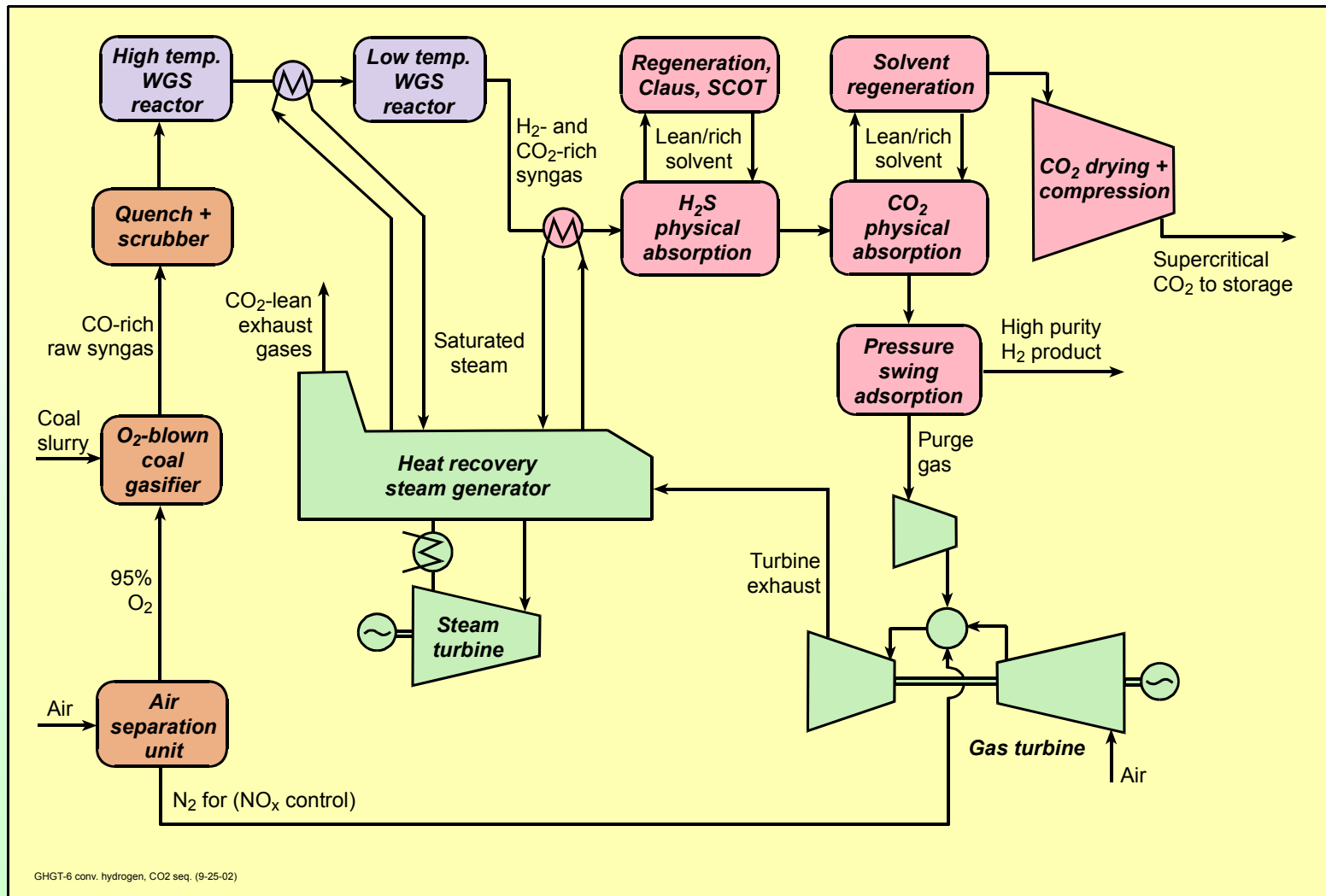
- Cost: 6.4 ¢/kWh, efficiency: 34.9% (HHV). (70 bar gasifier, scale: 362 MW<sub>e</sub>)

# H<sub>2</sub> Production: Add H<sub>2</sub> Purification/Separation



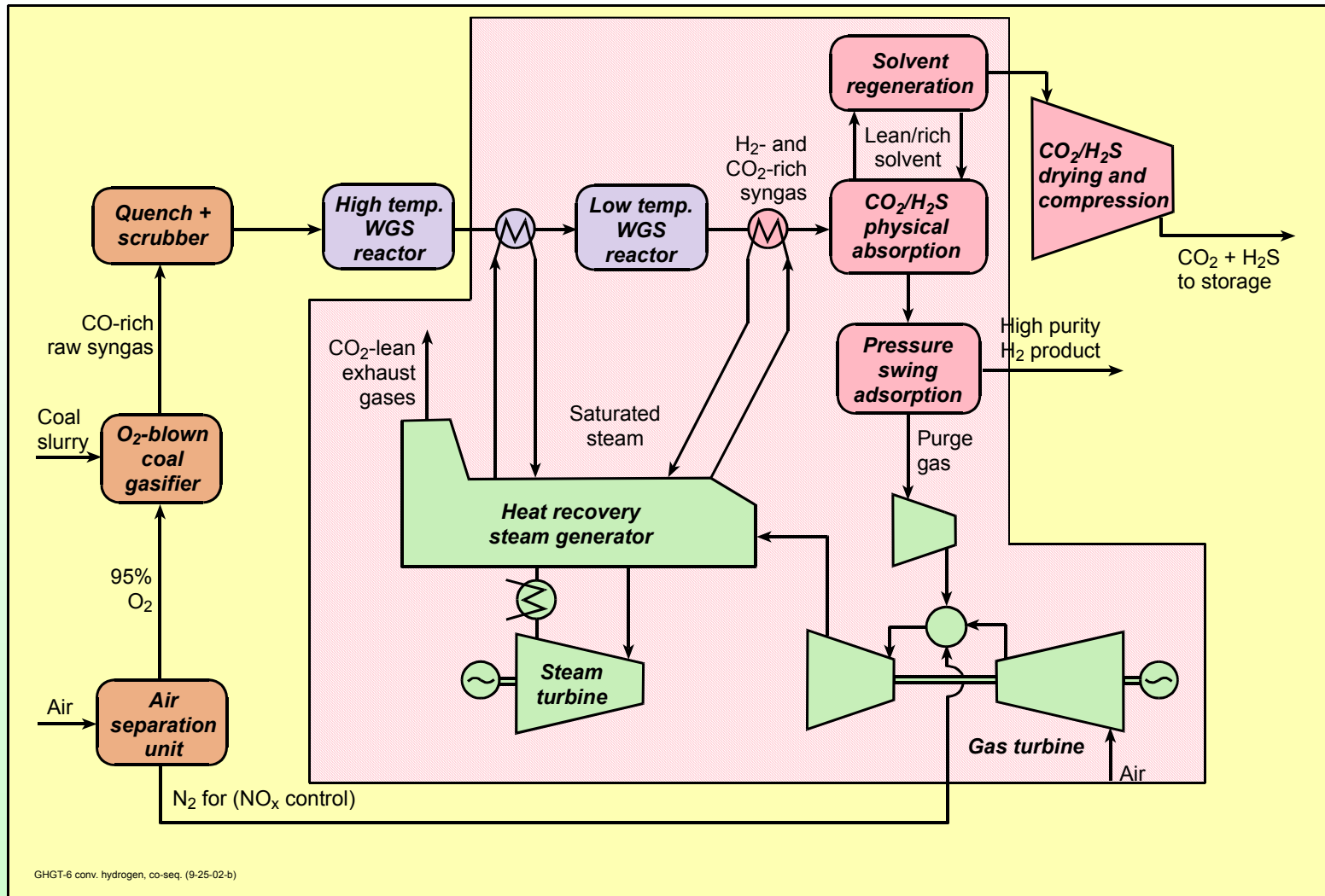
- Replace syngas expander with PSA and purge gas compressor.

# Conventional H<sub>2</sub> Production with CO<sub>2</sub> Capture



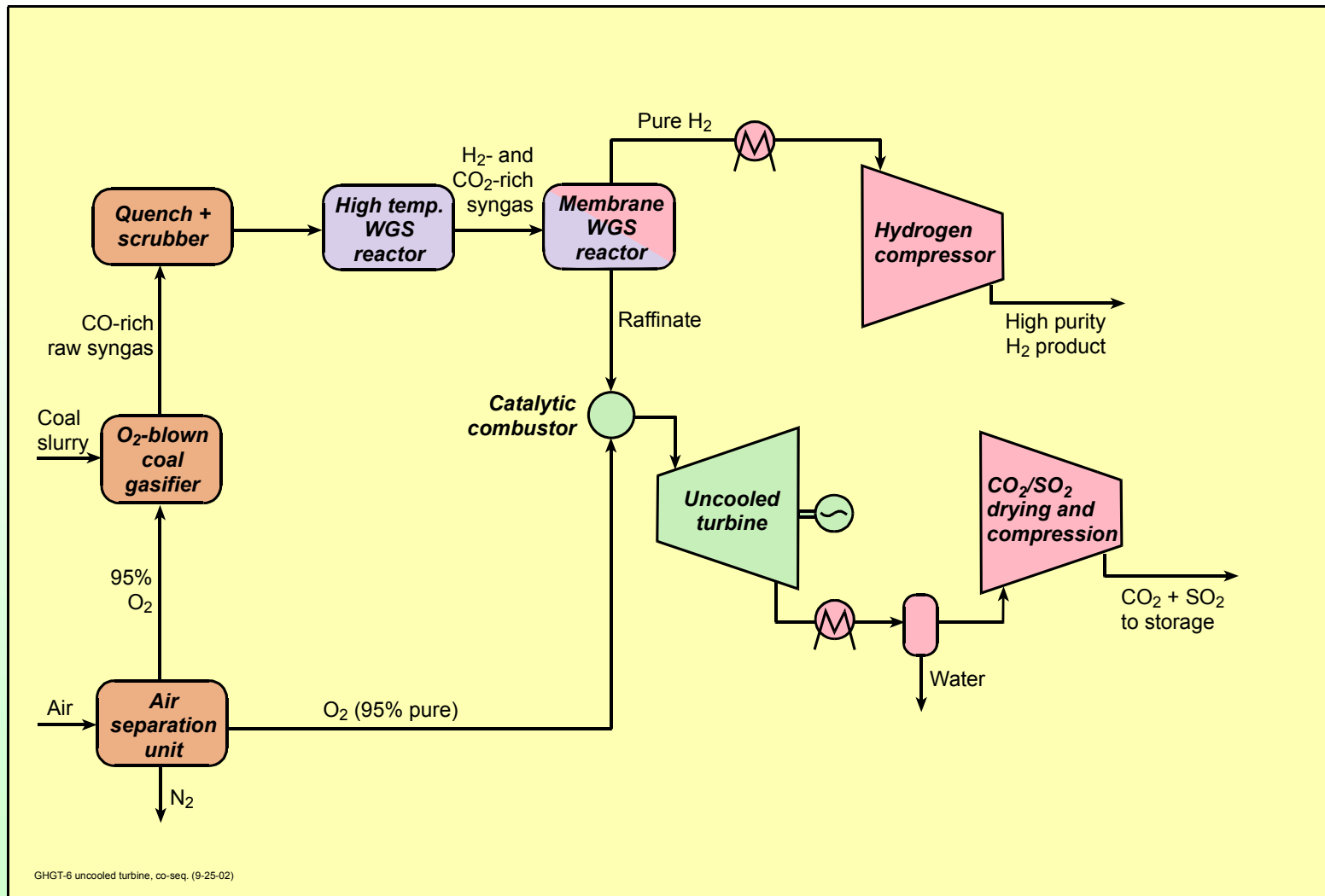
- H<sub>2</sub> cost: 7.3 \$/GJ (HHV). (85% HRF, scale: 1 GW<sub>th</sub> H<sub>2</sub> HHV, @ 6.4 ¢/kWh)

# Change $H_2$ - $CO_2$ Gas Separation Scheme



- This work uses a membrane to separate  $H_2$  from the syngas instead of  $CO_2$ .

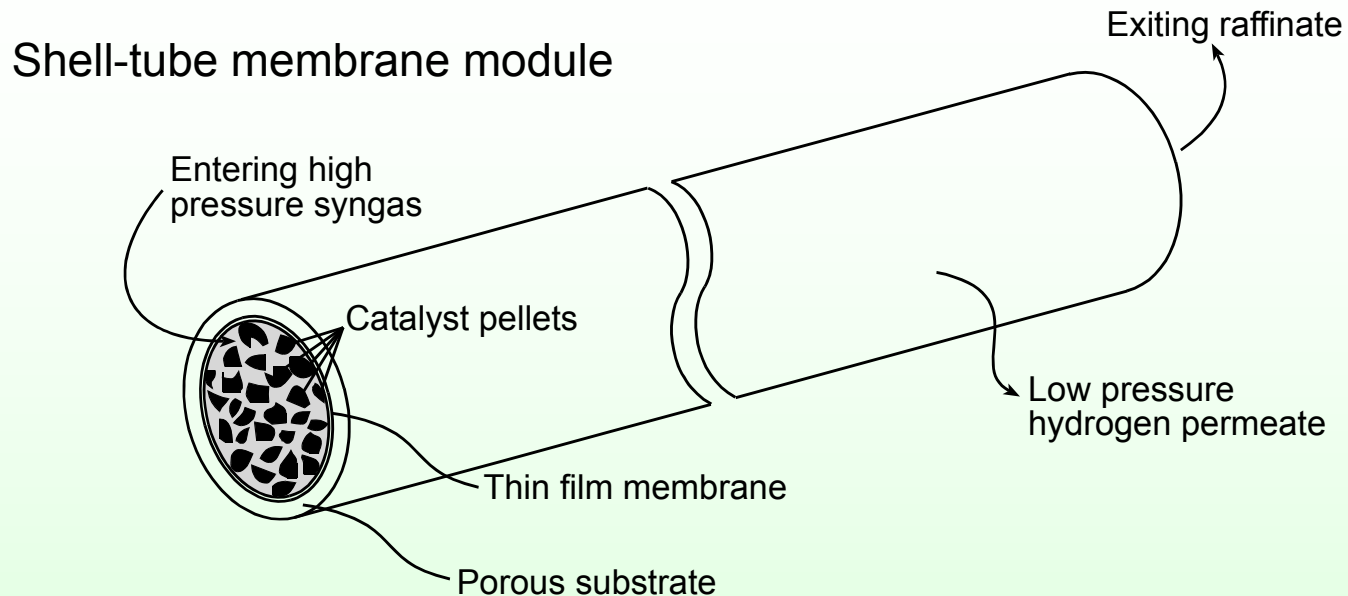
# H<sub>2</sub> Separation Membrane Reactor System



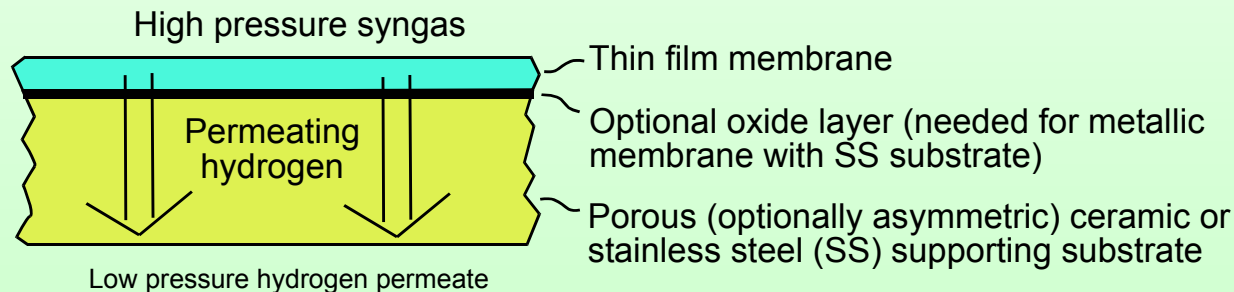
- Employ a H<sub>2</sub> permeable, thin film (10 μm), 60/40% Pd/Cu (sulfur tolerant) dense metallic membrane, configured as a WGS membrane reactor.



# Hydrogen Separation Membrane Reactor (HSMR) Concept



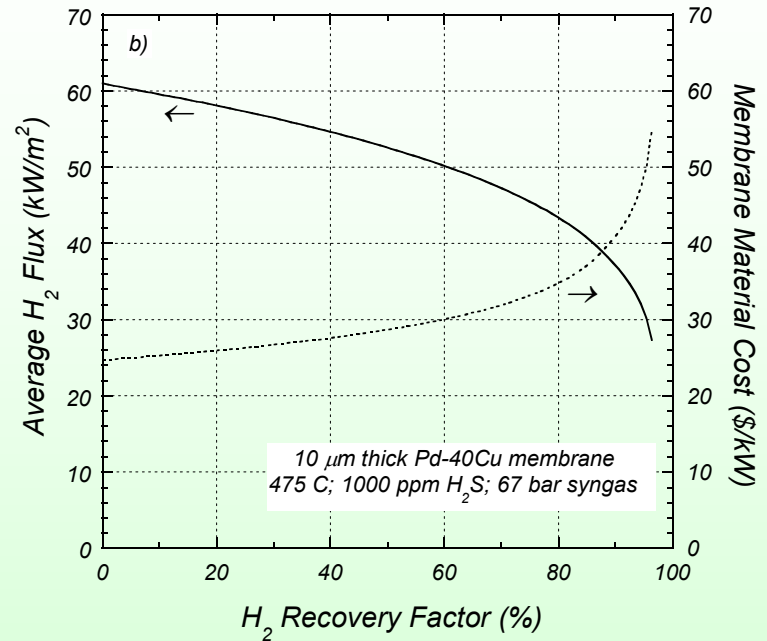
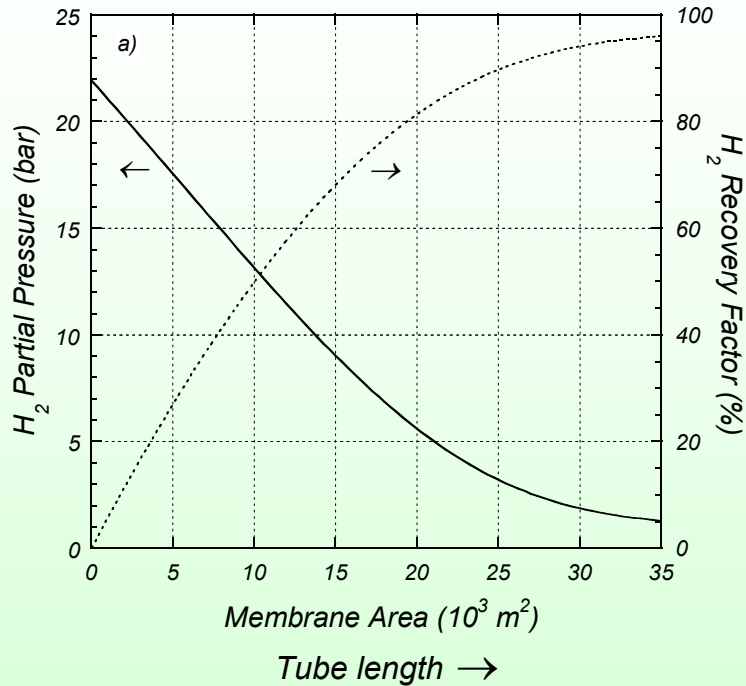
## Membrane Structure:



Membrane Reactor 5 5-3-03

- Alternative HSMR design: high pressure, WGS reaction, and membrane *outside* supporting tube, with  $H_2$  permeating to the interior of the tube

# Typical Membrane Reactor Performance



- $H_2$  Recovery Factor (HRF) =  $H_2$  recovered / ( $H_2$ +CO) in syngas
- HRF increases with membrane area → **diminishing returns**
- Membrane costs rise sharply above HRF~80-90% (no sweep gas)

# Cost of $H_2$ Compression and HSMR vs. $H_2$ Backpressure

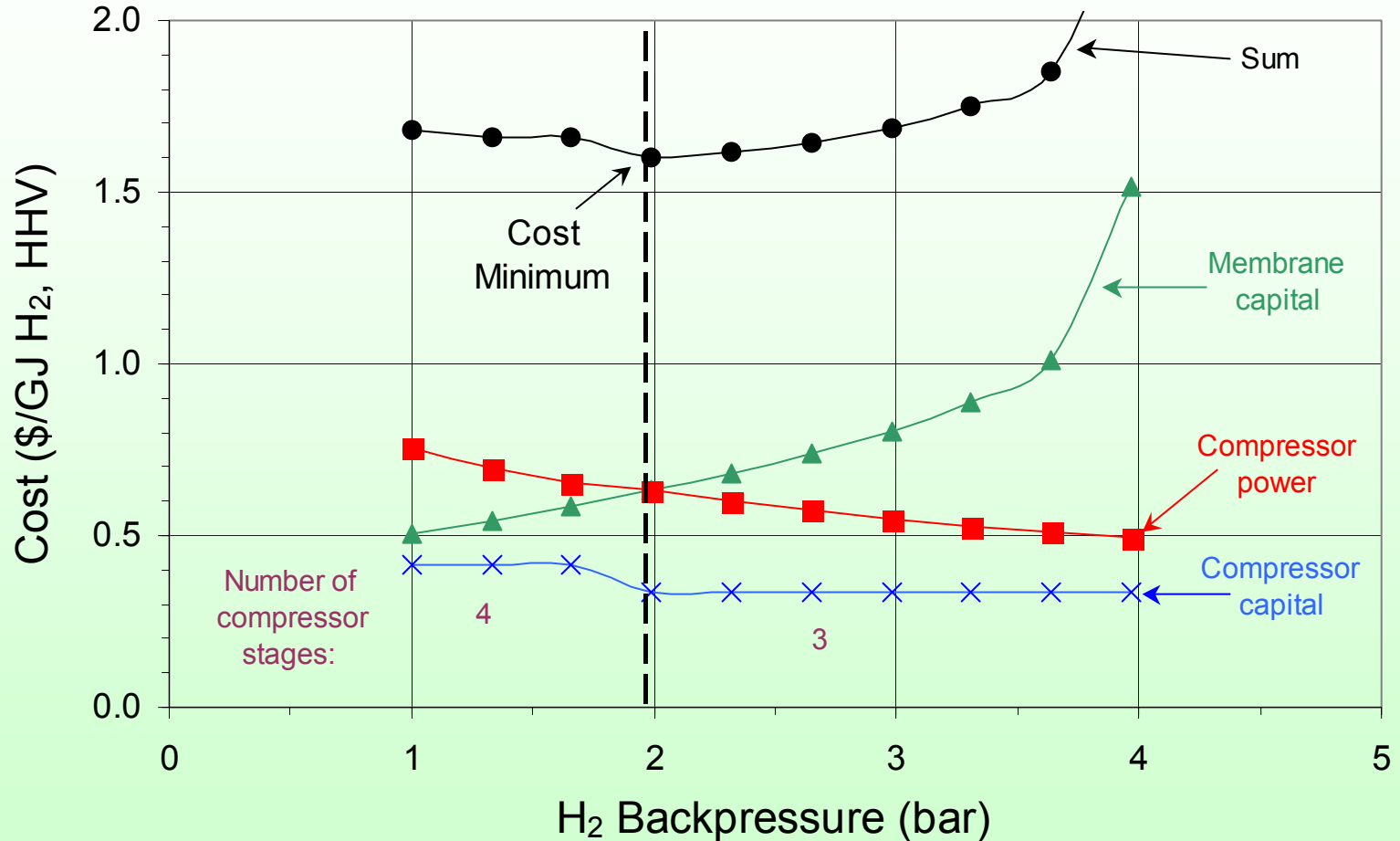


Fig. G4b

- Broad cost minimum seen here (not always) at low  $H_2$  backpressure

# *System Parameter Variations*

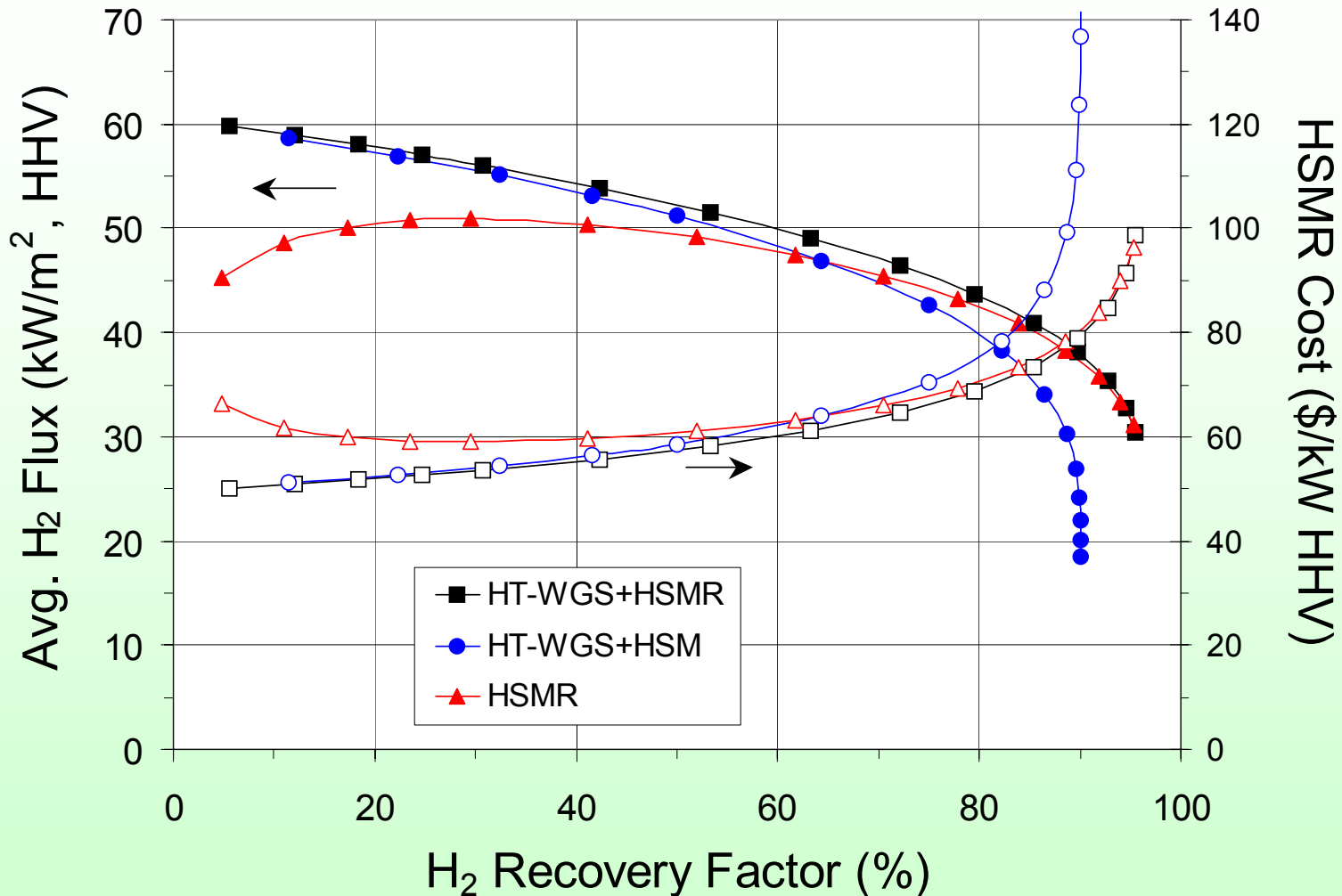
## *System Performance:*

- membrane reactor configuration
- membrane reactor operating temperature
- gasifier/system pressure
- hydrogen backpressure
- hydrogen recovery factor (HRF)
- raffinate turbine technology (blade cooling vs. uncooled)

## *System Economics:*

- membrane reactor cost (and type)
- co-product electricity value
- sulfur capture vs. sulfur + CO<sub>2</sub> co-sequestration

## Effect of HSMR Configuration



- In this system, with an upstream WGS reactor, a membrane *reactor* not obviously necessary for good system performance.

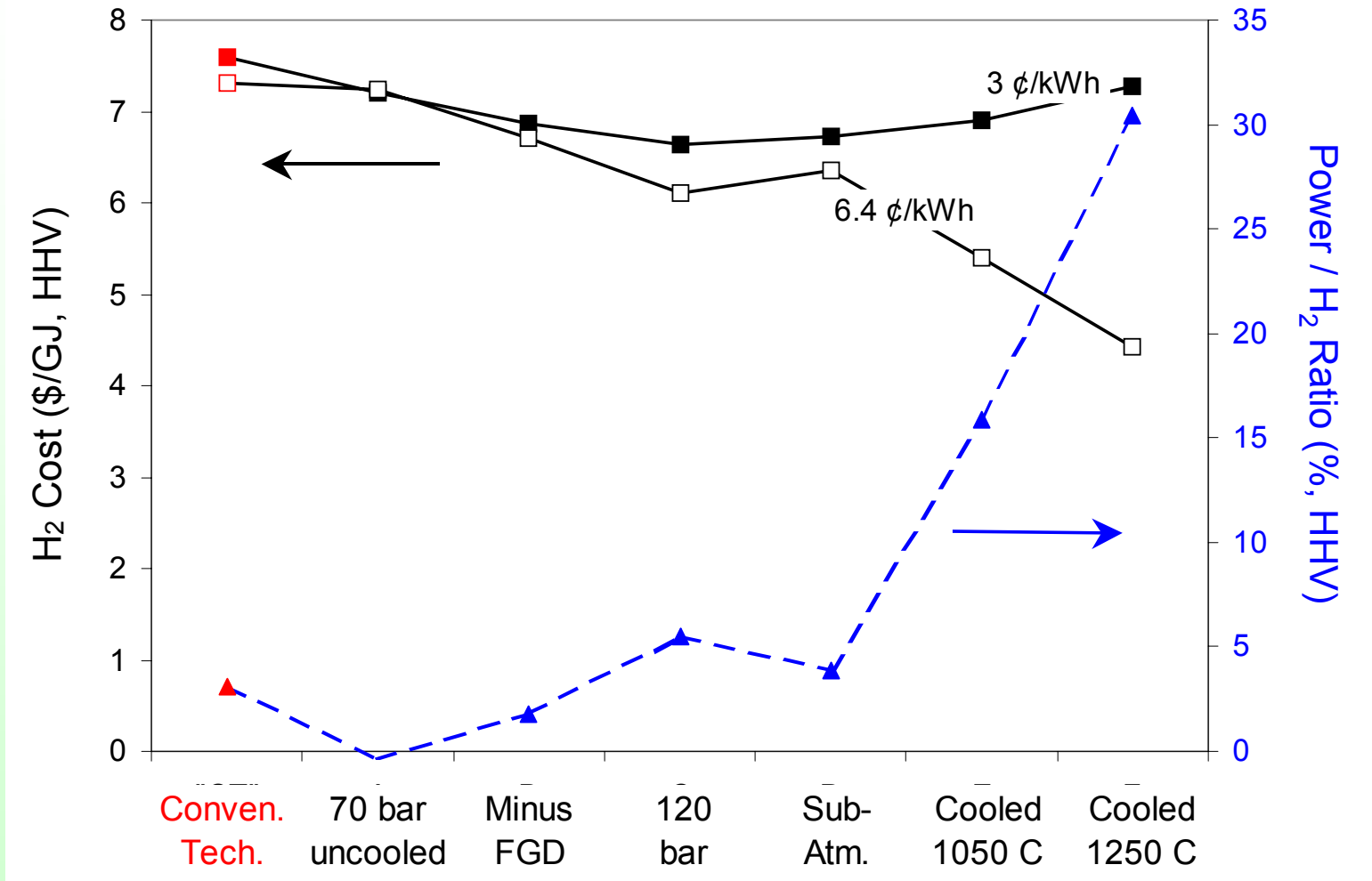
## Modeling Results: Parametric Variations

	Description of system and/or change from one system to the next (in sequence). (Scale=1 GW <sub>th</sub> HHV coal input)	Hydrogen Recovery Factor (%)	“Effective” system efficiency (% <sub>HHV</sub> )*	CO <sub>2</sub> -free H <sub>2</sub> Cost (\$/GJ HHV)	
				@ 6.4 ¢/kWh	@ 3 ¢/kWh
	Conventional technology	85.0	67.0	7.31	7.60
A	Uncooled turbine, 70 bar, FGD	89.0	67.5	7.24	7.20
B	“A” without FGD (co-storage)	87.0	69.1	6.71	6.87
C	“B” with 120 bar gasifier	83.2	70.7	6.11	6.63
D	“C” with sub-atmospheric (0.2 bar) turbine exhaust pressure	86.5	71.5	6.36	6.73
E	“C” with cooled raffinate turbine (TIT=1050 C)	69.1	69.6	5.40	6.90
F	“E” with TIT=1250 C	55.3	66.9	4.42	7.28

\* Effective system efficiency = HHV H<sub>2</sub> output / HHV (coal input – coal saved\*\*)

\*\* Coal saved based on IGCC with CO<sub>2</sub> capture, 34.9% HHV efficiency

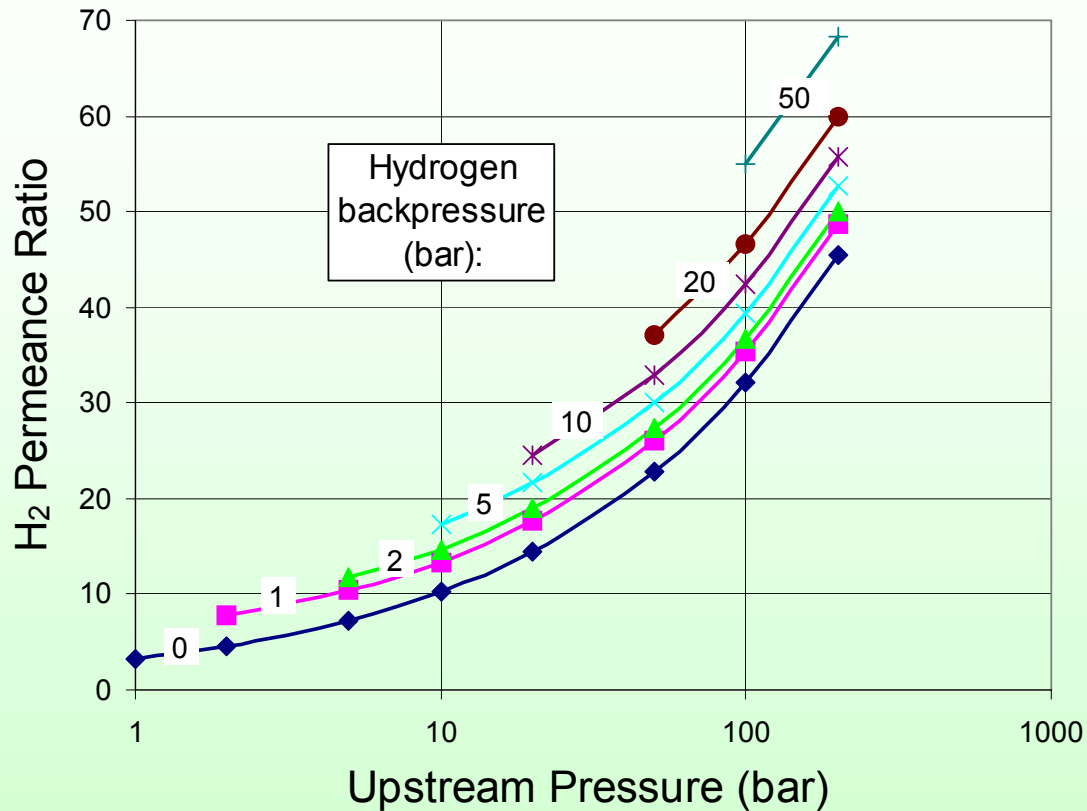
# Modeling Results: Parametric Variations



- H<sub>2</sub> costs via membranes comparable to costs via “conventional technology”
- Power / H<sub>2</sub> ratio and electricity price are key to H<sub>2</sub> costs

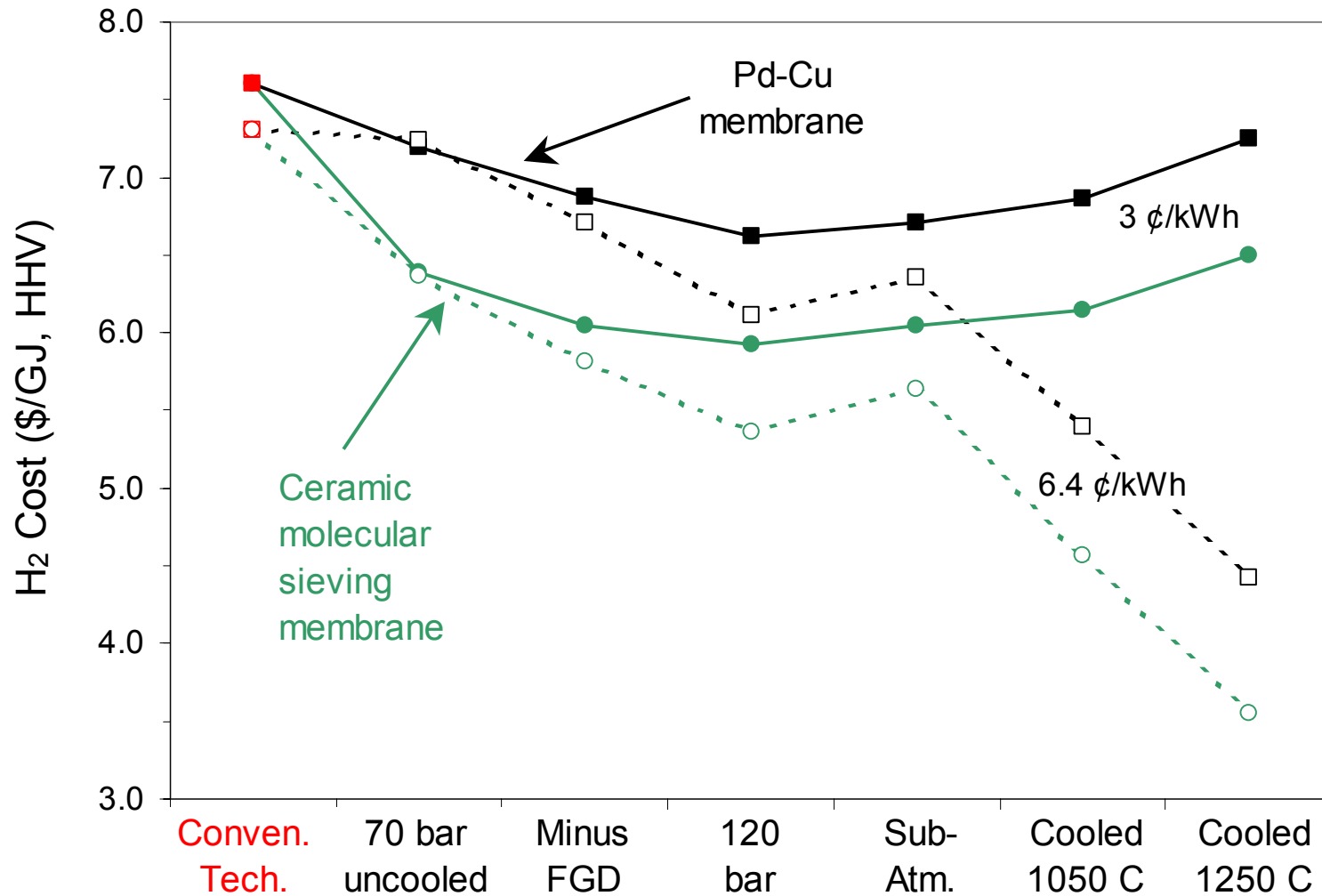
# Oak Ridge Molecular Sieving Membrane

-  $H_2$  Permeance Relative to 60/40 Pd/Cu -



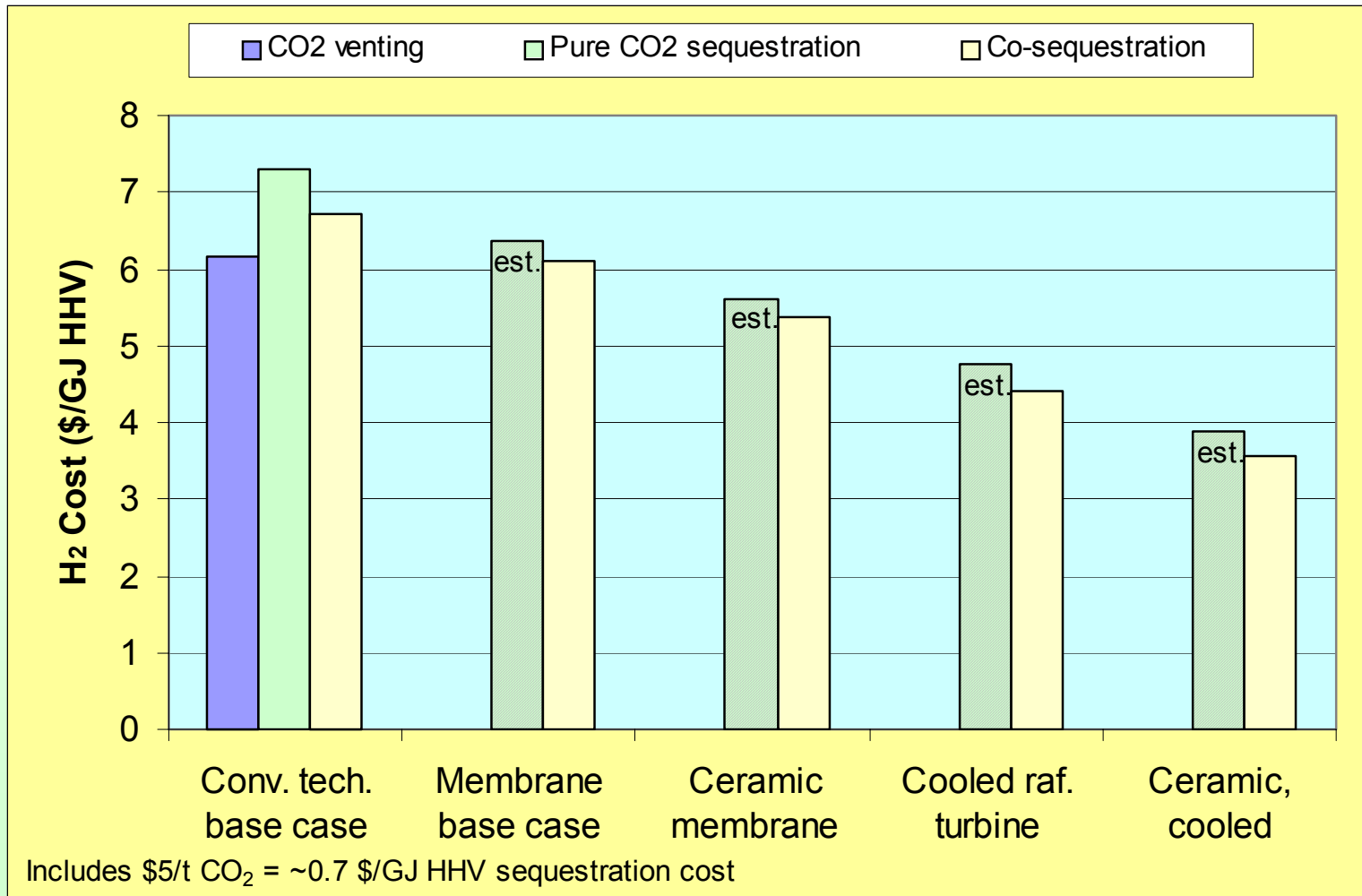
- Membrane permeance pressure dependence:
  - ceramic:  $(P_{\text{high}} - P_{\text{low}})$  vs. metallic:  $(\sqrt{P_{\text{high}}} - \sqrt{P_{\text{low}}})$
- Permeance increase up to factor of ~50 possible. Reduced purity.

# Metallic vs. Ceramic Membrane reactors



- The increased permeance of ceramic membranes allows for potentially lower H<sub>2</sub> costs. (Same membrane unit price assumed, \$3,021/m<sup>2</sup>.)

# Optimistic\* Results Summary



\* Assumes electricity = 6.4 c/kWh

# Observations

- In quench gasifiers, equilibrium shifting does not appear to be very important (membrane *reactor* vs. membrane *permeator*).
- With efficient raffinate turbine and costly membrane reactor, high  $\text{CO} \rightarrow \text{H}_2$  conversion efficiency may not be a design goal.
- High *HRF* yields high system efficiency and low  $\text{H}_2$  costs that are relatively insensitive to the electricity prices; very high *HRF* typically is not required.
- Raising the gasifier pressure from 70 to 120 bar raises system efficiency and has the potential to lower the cost of  $\text{H}_2$ .
- Sub-atmospheric raffinate turbine exhaust pressure might improve efficiency; effect on costs is uncertain.
- Raffinate turbine blade cooling yields low *HRF*, i.e. increased power production, and more complex economic analyses.

## *Further Observations, Future Plans*

- SO<sub>2</sub>-CO<sub>2</sub> co-sequestration raises the system efficiency, may lower the cost of H<sub>2</sub>, and may provide environmental benefits (Hg, etc.).
- Good system design can yield H<sub>2</sub> costs that are relatively insensitive to HSMR cost and H<sub>2</sub> recovery.
- Relative to “conventional technology” membrane reactors might lower the cost of H<sub>2</sub> by ~10% (Pd-Cu) to ~20% (ceramic). **Gas separation is not a large fraction of capital cost.**
- Co-product electricity cost is very important to system design; entwined with H<sub>2</sub> recovery; co-product analysis is complex
- We plan to examine:
  - 1) low steam-to-carbon ratio + syngas cooling,
  - 2) alternative membrane types, configurations, and temperatures.