Analysis of Hydrogen and Co-Product Electricity Production from Coal with Near Zero Pollutant and CO_2 Emissions using an Inorganic Hydrogen Separation Membrane Reactor

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Large Scale Production of H_2 from Fossil Fuels

- Four Related Papers Presented Here -

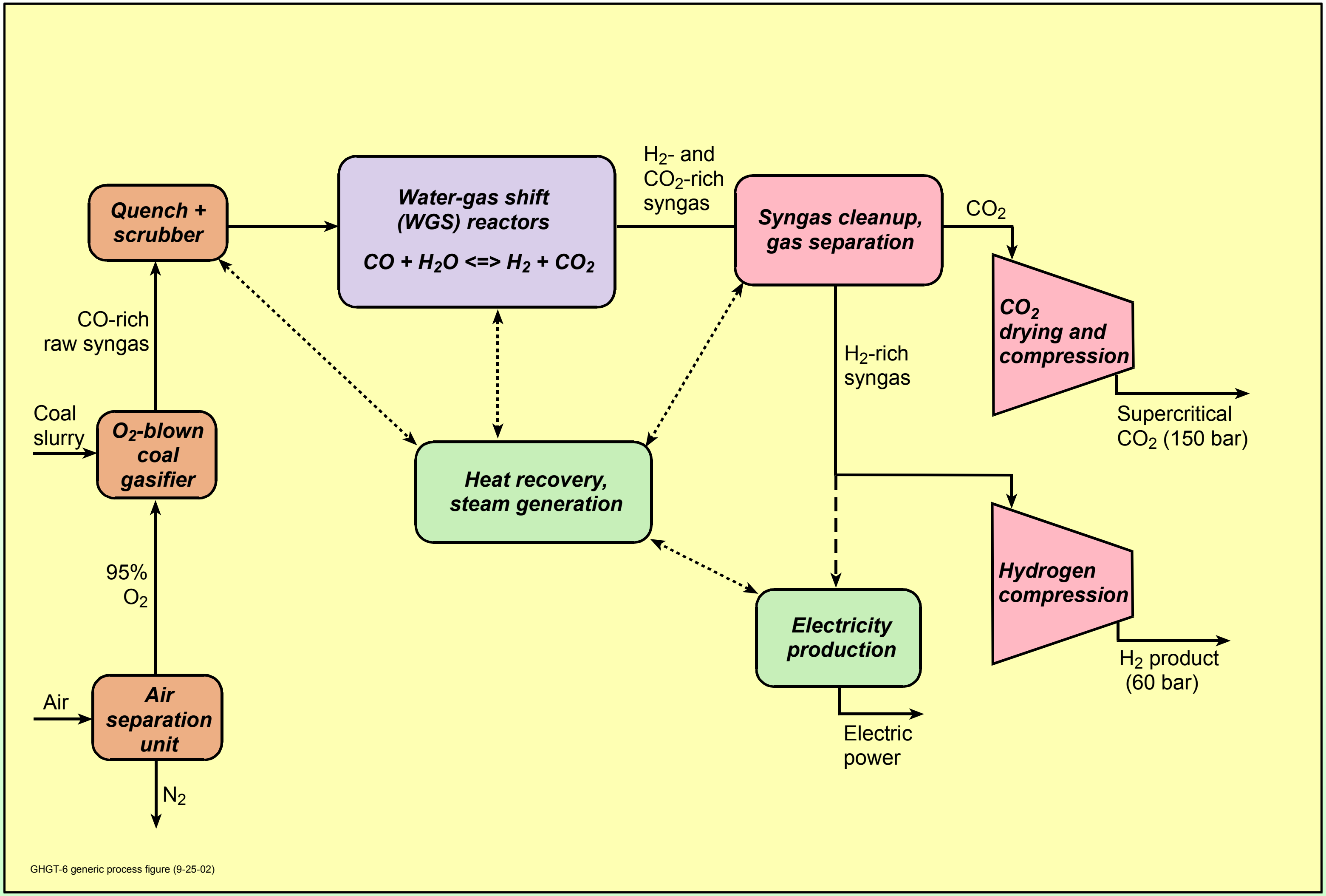
	Natural Gas	Coal & Residuals
CO_2 Venting	Almost all H_2 produced today	Refineries, chemicals, NH_3 production in China 2) "Conventional technology"
CO_2 Capture	1) FTR vs. ATR with CC	2) "Conventional technology" 3) Membrane reactors 4) Overview

H_2 *from Coal: Motivation*

- Distributed energy use (transportation and heating) responsible for ~2/3 of global CO_2 emissions
- CO_2 capture, compression, dehydration, and pipeline transport from distributed sources is *very* expensive.
- Low carbon energy carriers are needed: electricity*...and hydrogen?*
- If CO_2 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_2, Electricity, and CO_2



- All work presented here is based on O_2-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_2 production systems (both conventional and membrane-based)

Disaggregated Cost of H_2 Production

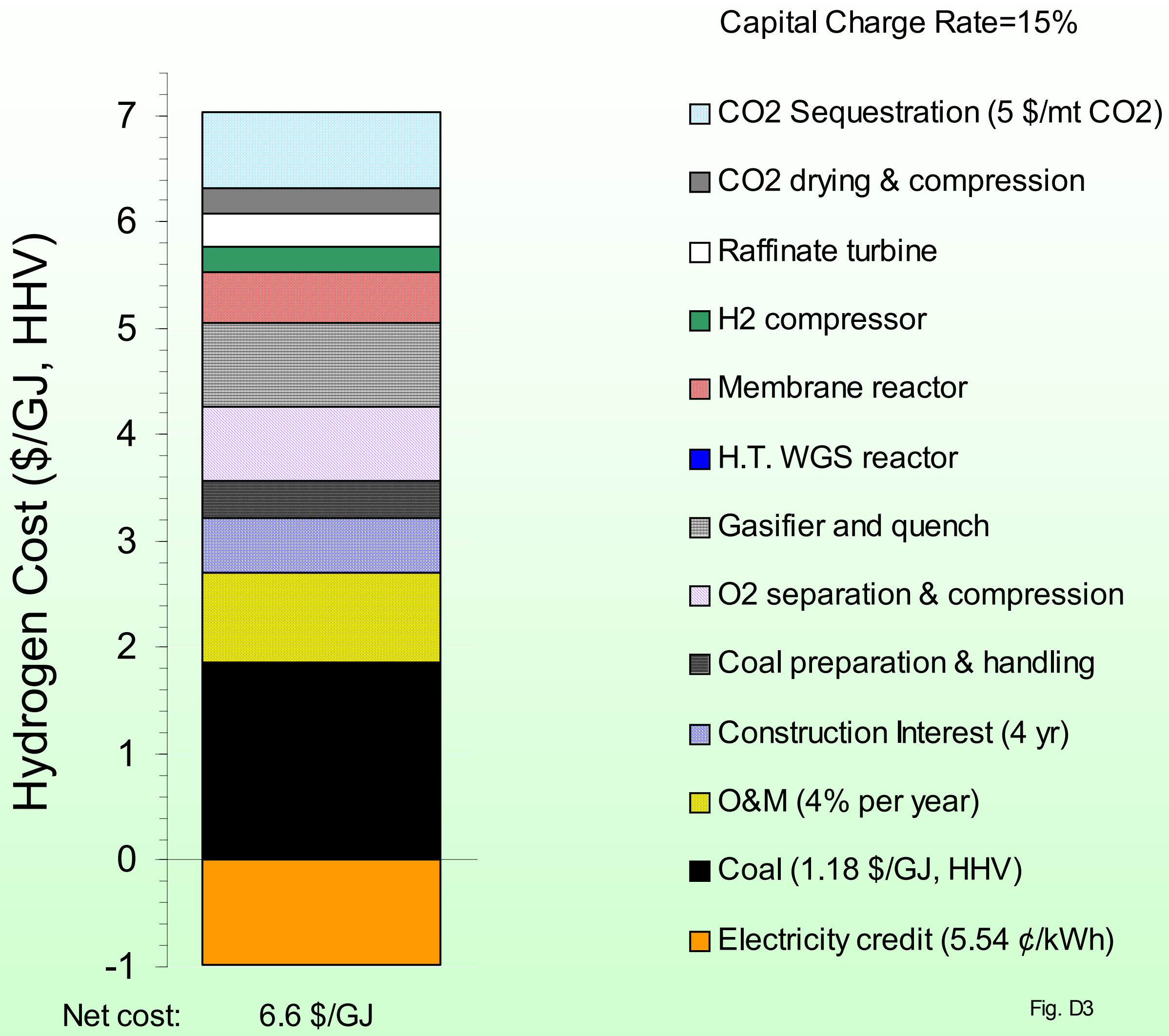


Fig. D3

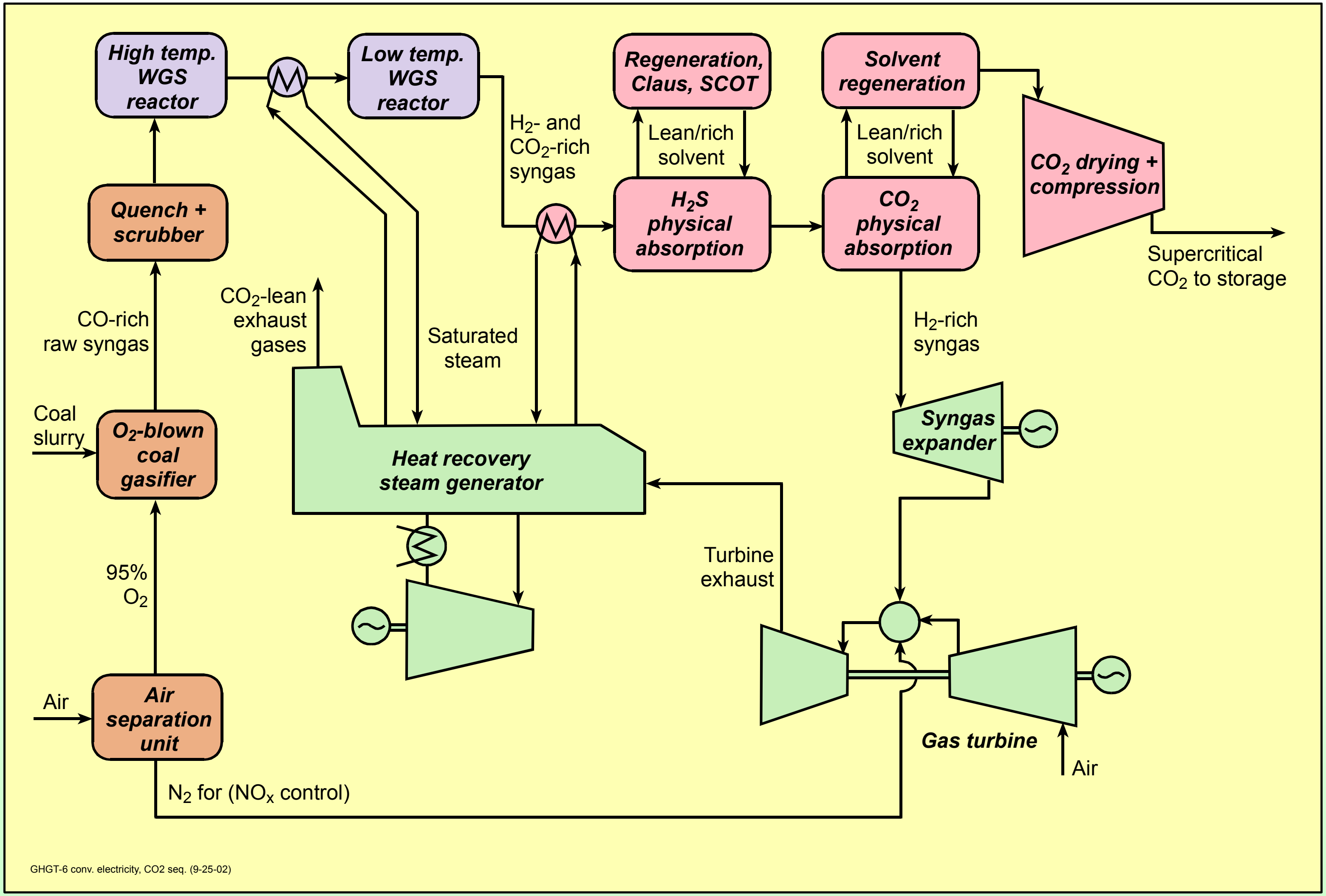
- 70 bar gasifier, 83.2% HRF, uncooled raffinate turbine, scale: 1 GW_{th} H_2 (HHV)

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_2 storage cost *	5 $/mt CO_2 (~0.7 $/GJ H_2 HHV)
U.S. dollars valued in year	2002
Plant scale	1 GW_{th} H_2
Coal Type	Illinois #6

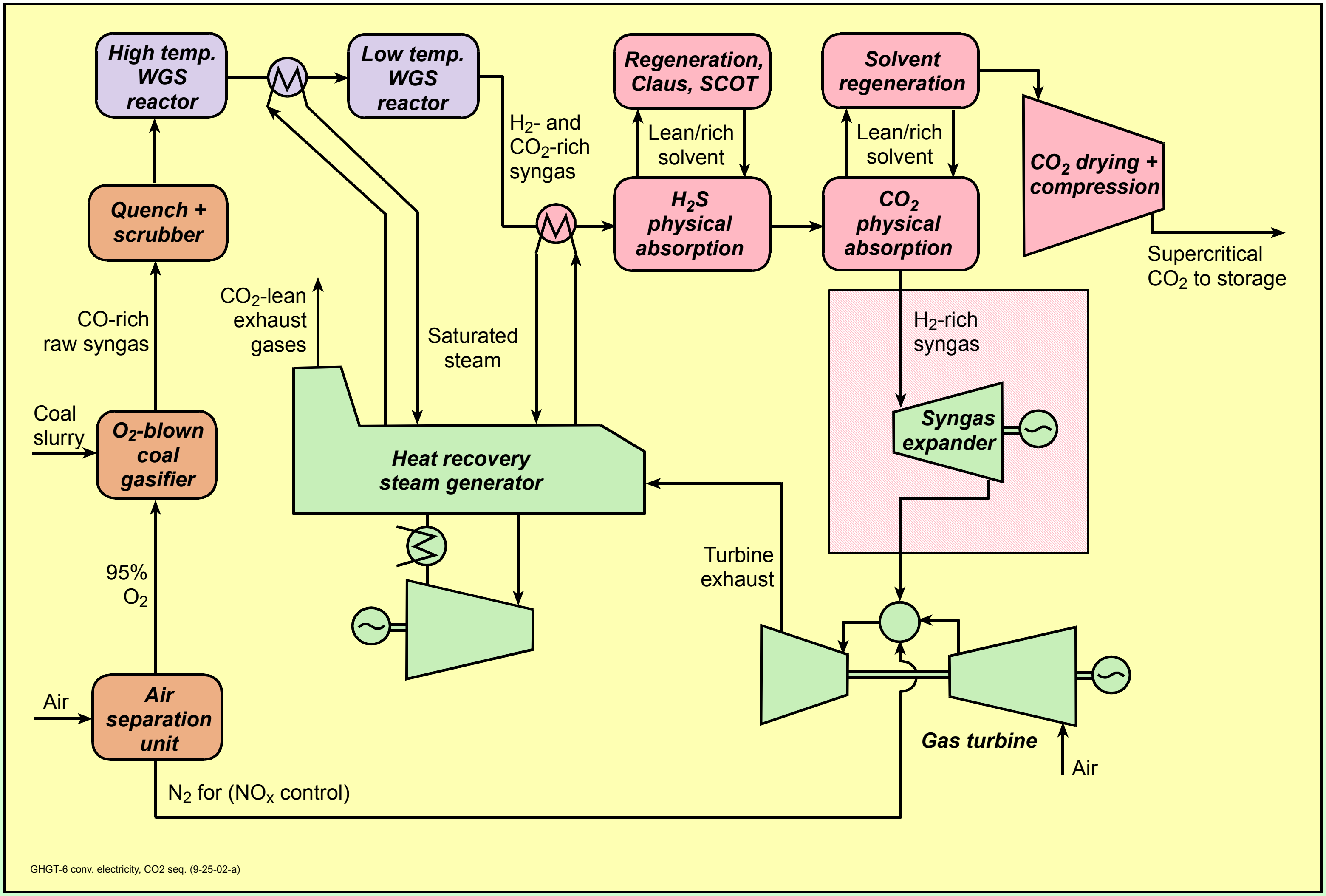
- "Best case" cost estimate for: 16,000 tonne/day CO_2, 100 km pipeline, 2 km deep injection well (layer thickness > 50 m, permeability > 40 mDa)

Benchmark: IGCC Electricity with CO_2 Capture



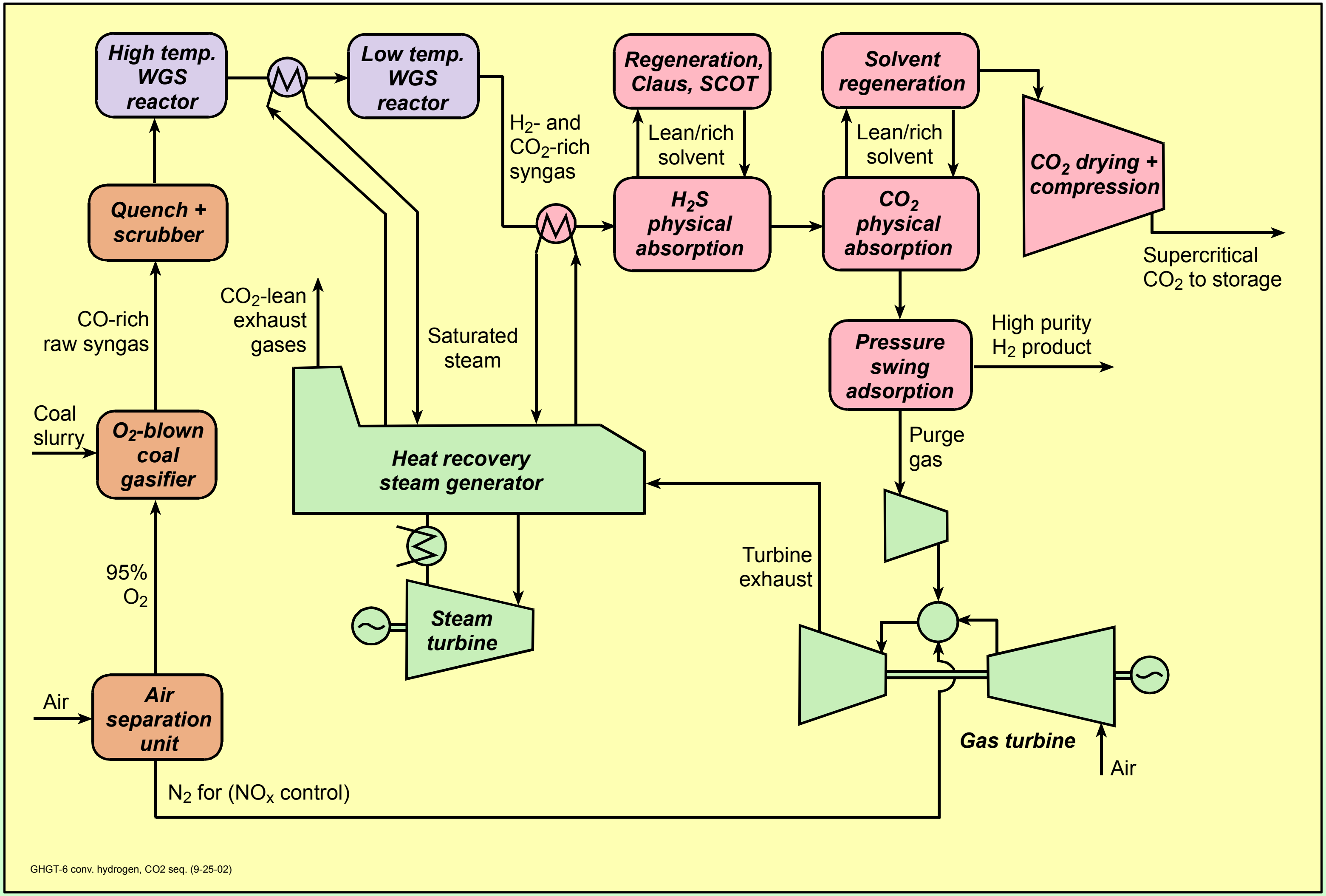
• Cost: 6.4 ¢/kWh, efficiency: 34.9% (HHV). (70 bar gasifier, scale: 362 MW_e)

H_2 Production: Add H_2 Purification/Separation



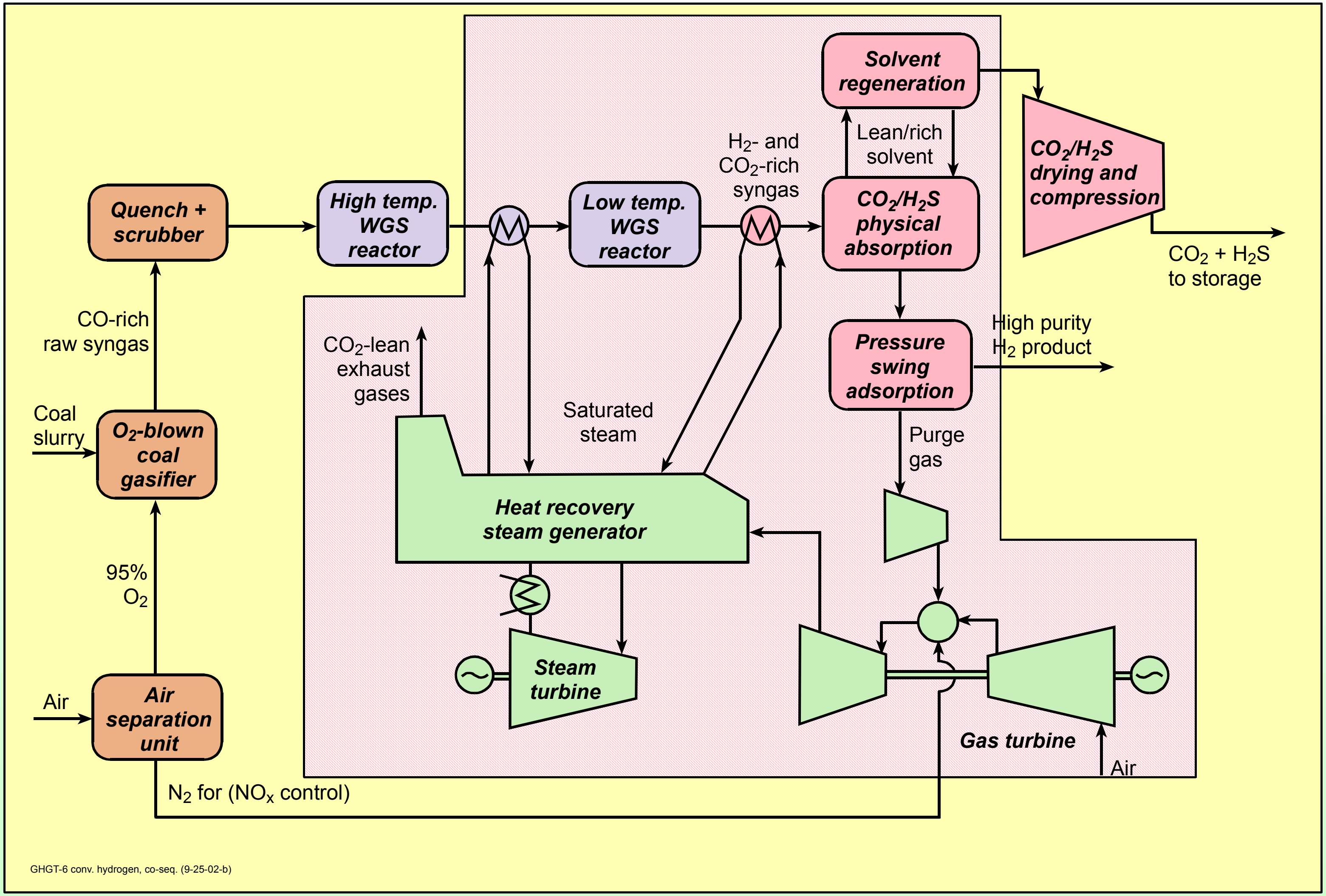
- Replace syngas expander with PSA and purge gas compressor.

Conventional H_2 Production with CO_2 Capture



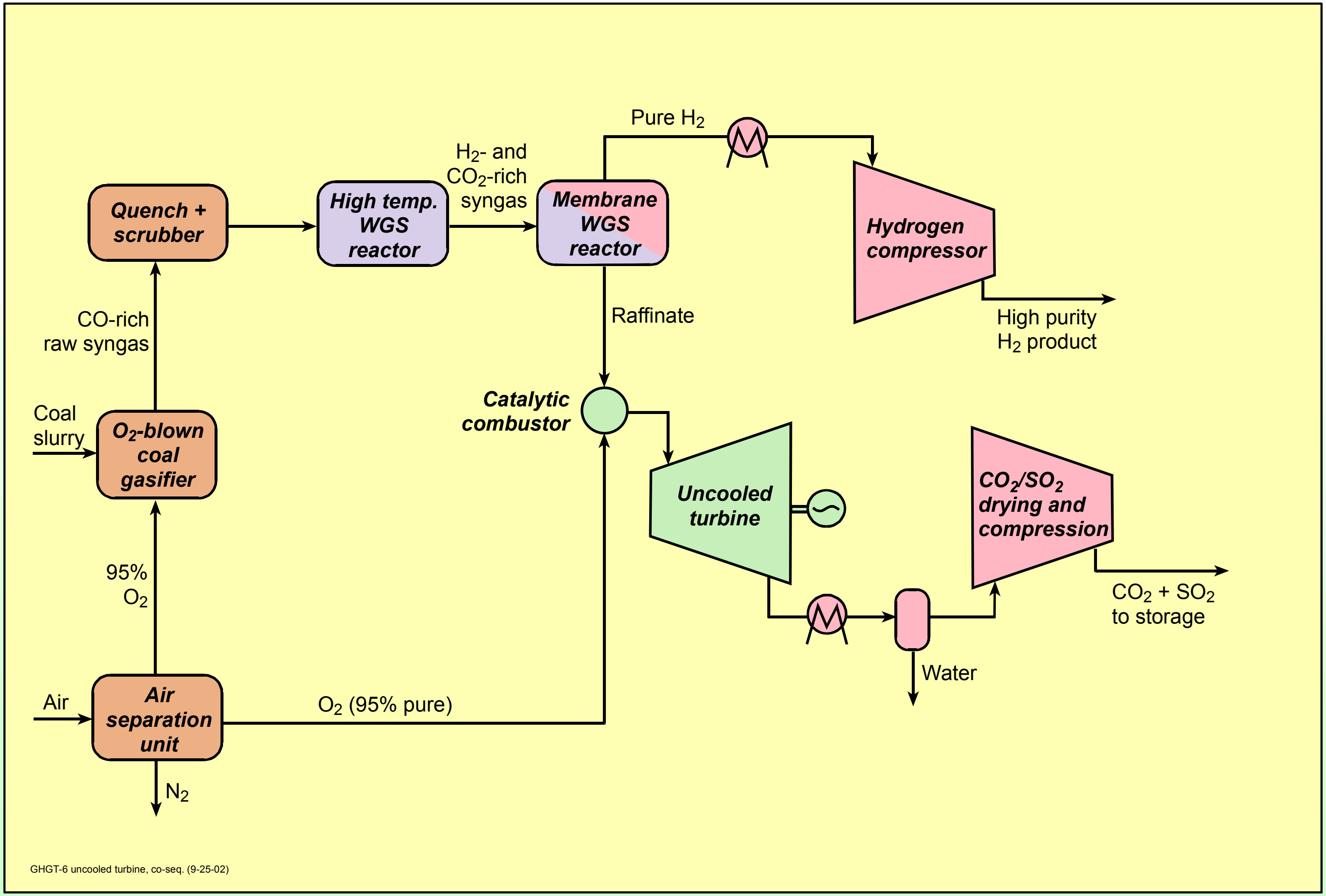
- H_2 cost: 7.3 \$/GJ (HHV). (85% HRF, scale: 1 GW_{th} H_2 HHV, @ 6.4 ¢/kWh)

Change H_2-CO_2 Gas Separation Scheme



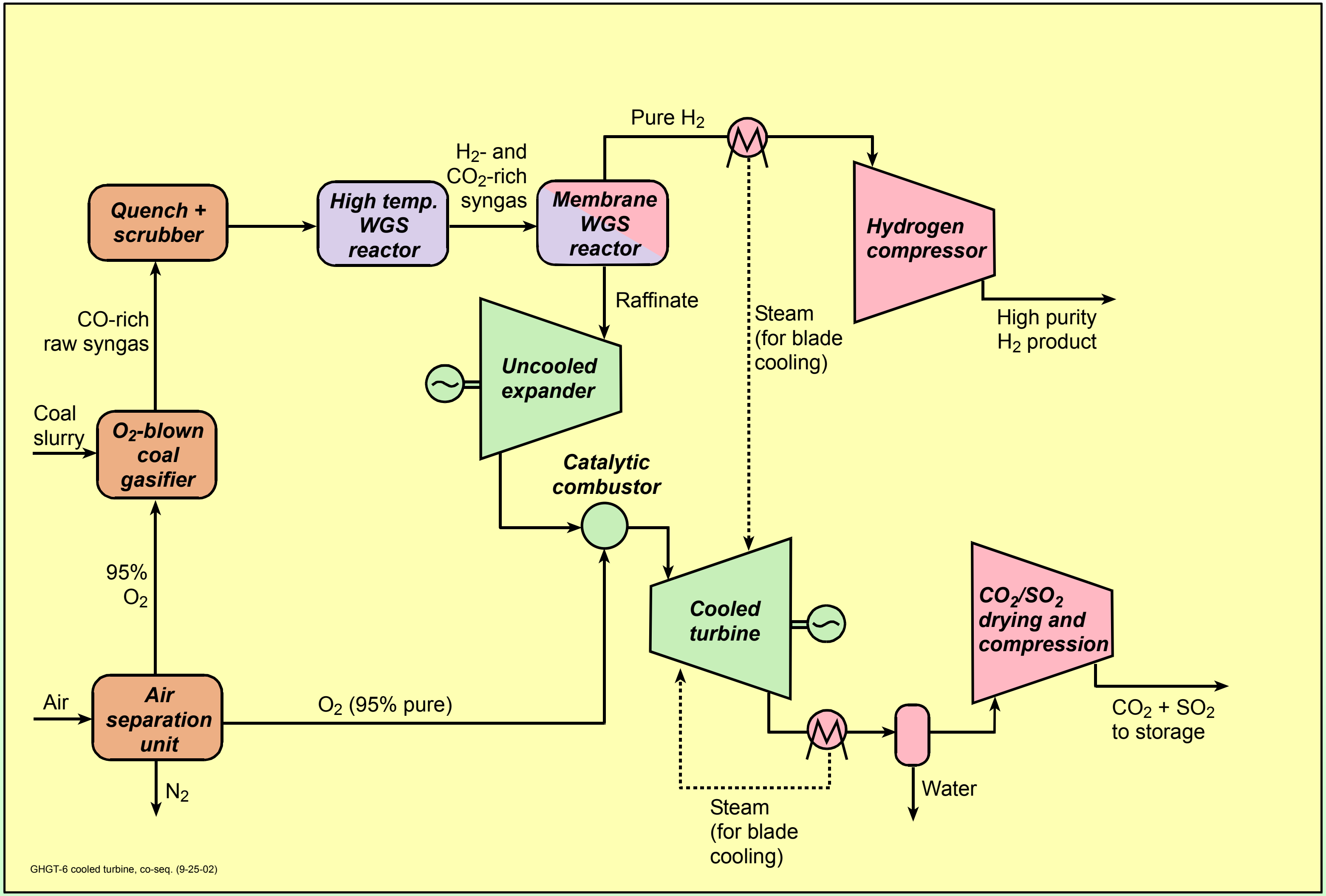
- This work uses a membrane to separate H_2 from the syngas instead of CO_2.

H_2 Separation Membrane Reactor System



- Employ a H_2 permeable, thin film (10 μm), 60/40% Pd/Cu (sulfur tolerant) dense metallic membrane, configured as a WGS membrane reactor.

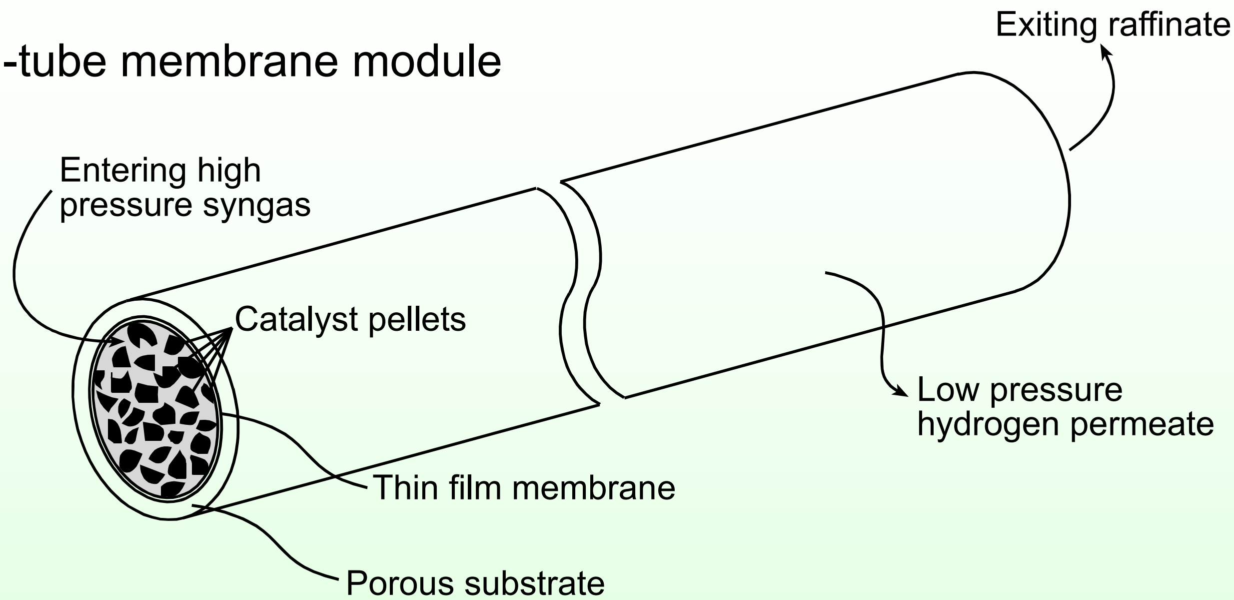
Membrane System with Cooled Raffinate Turbine



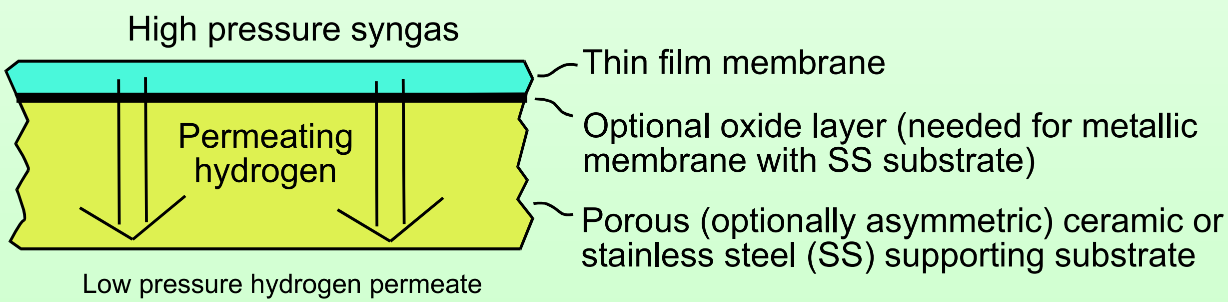
- Blade cooling with steam enables higher TIT (1250 C vs. 850 C), and higher electrical conversion efficiency. Requires much lower HRF (~60%).

Hydrogen Separation Membrane Reactor (HSMR) Concept

Shell-tube membrane module



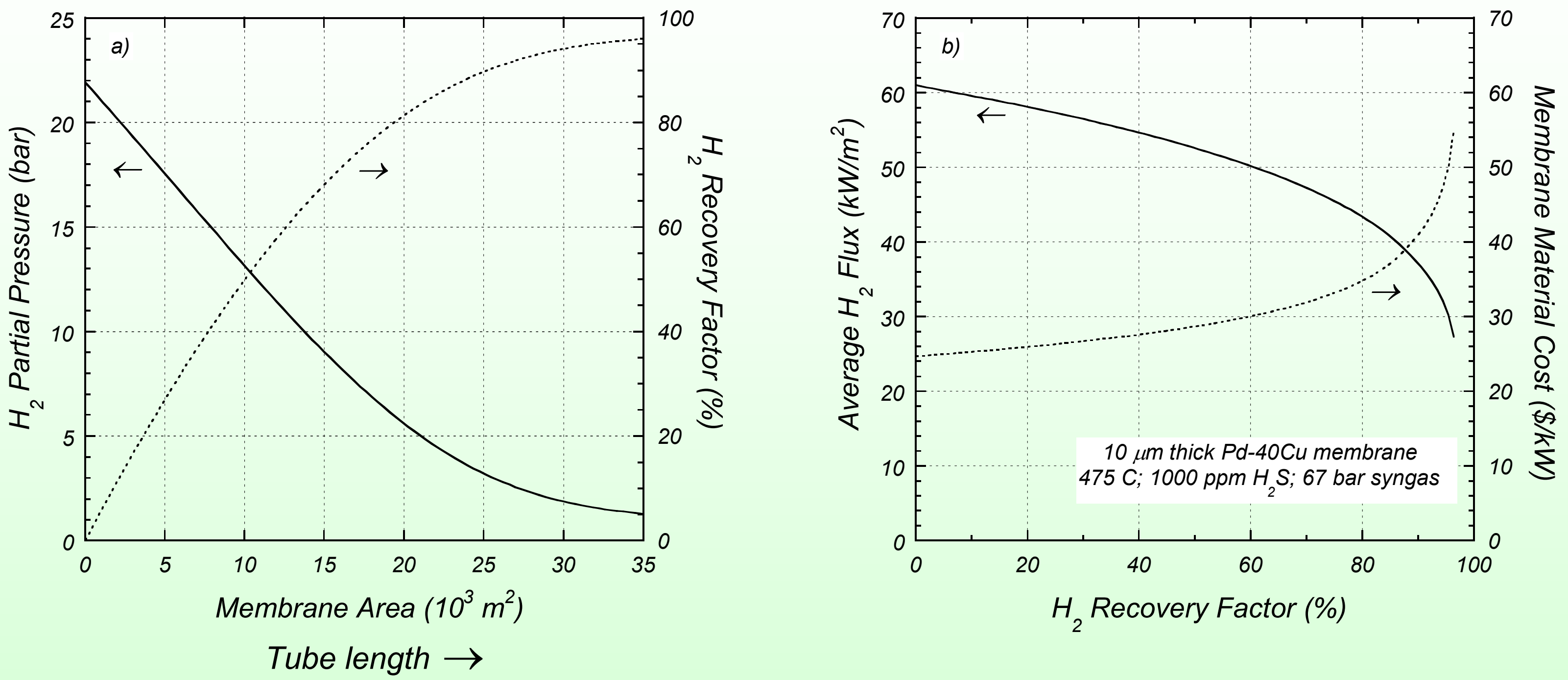
Membrane Structure:



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

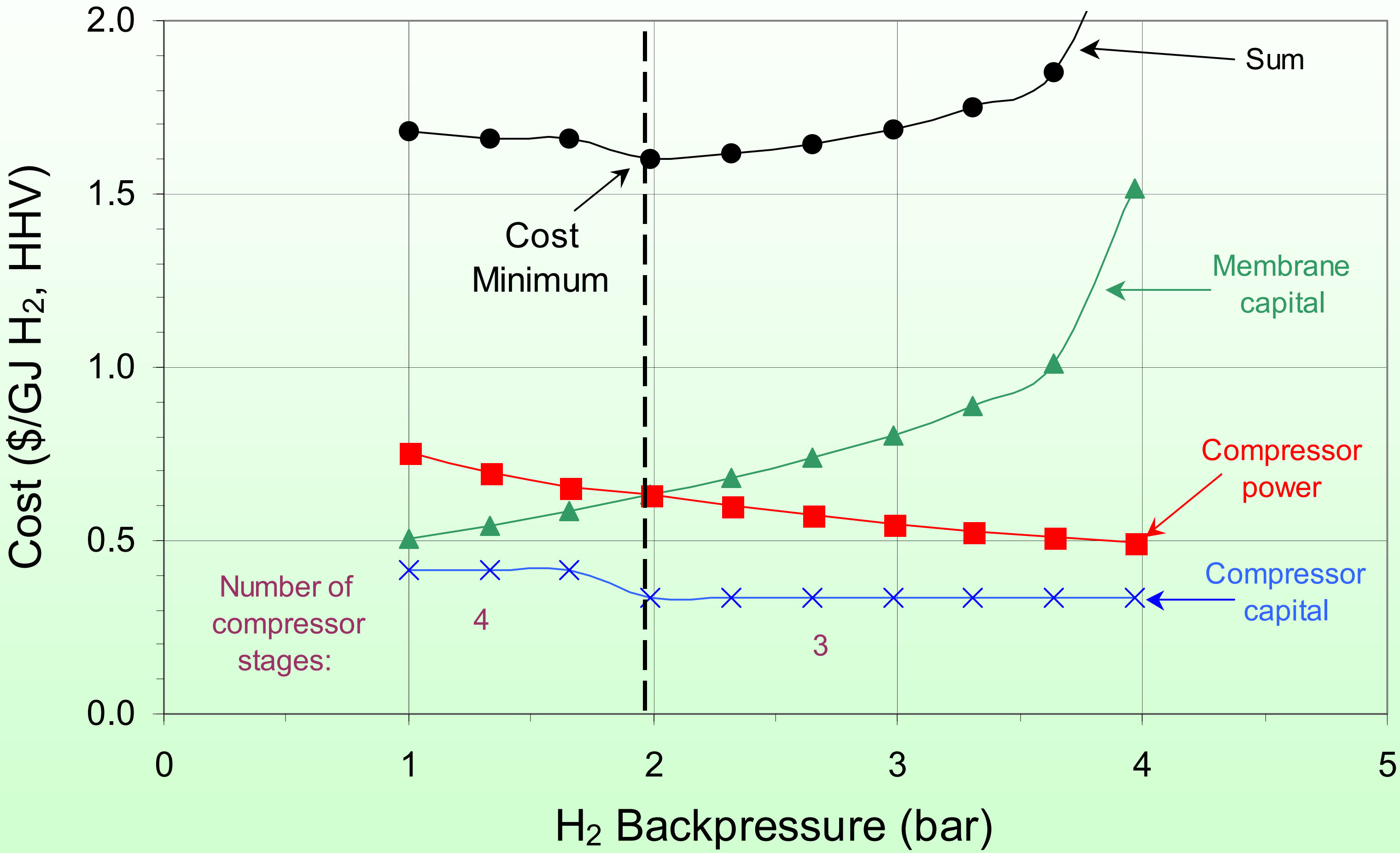


Fig. G4b

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

System Parameter Variations

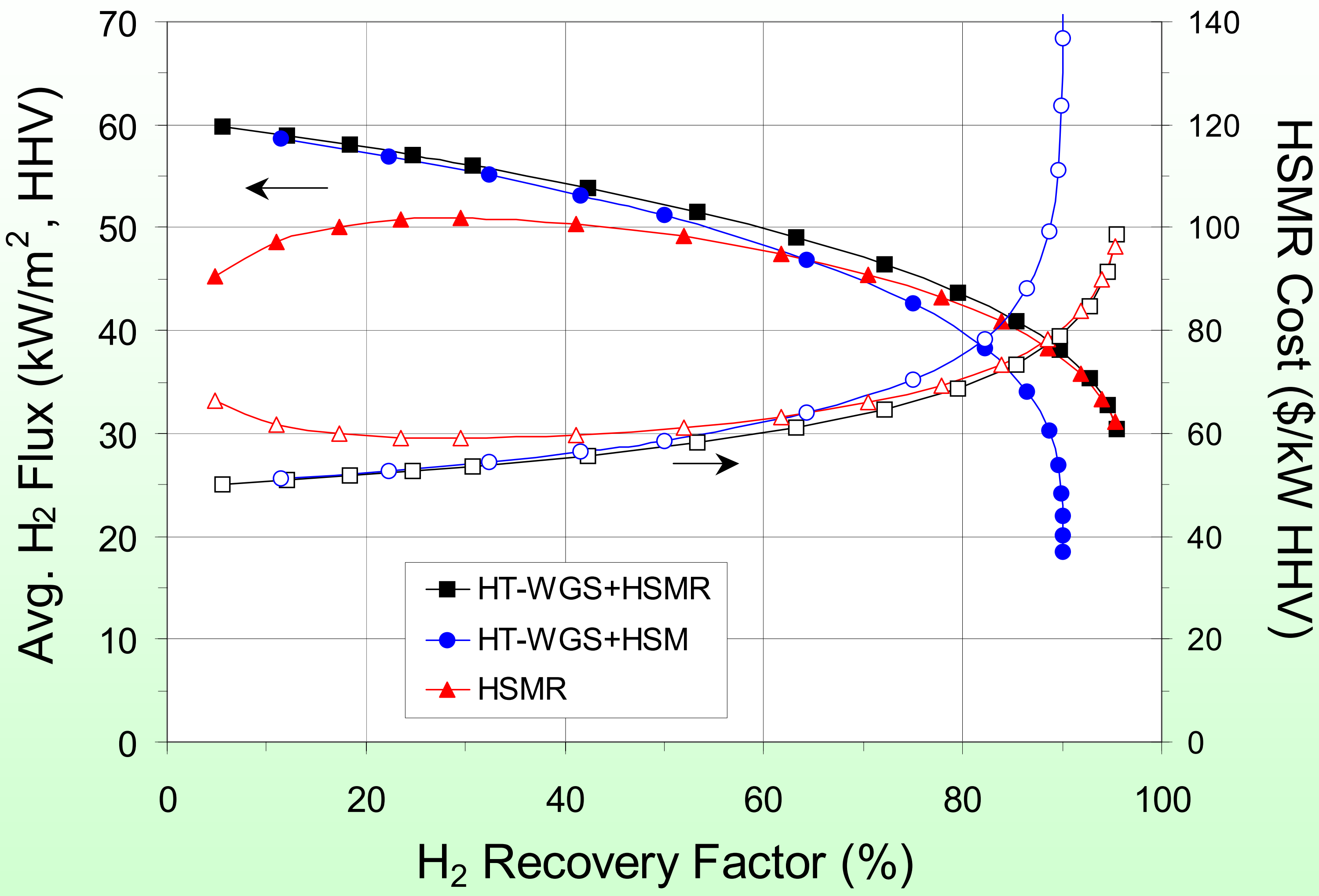
System Performance:

- 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_2 co-sequestration

Effect of HSMR Configuration



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

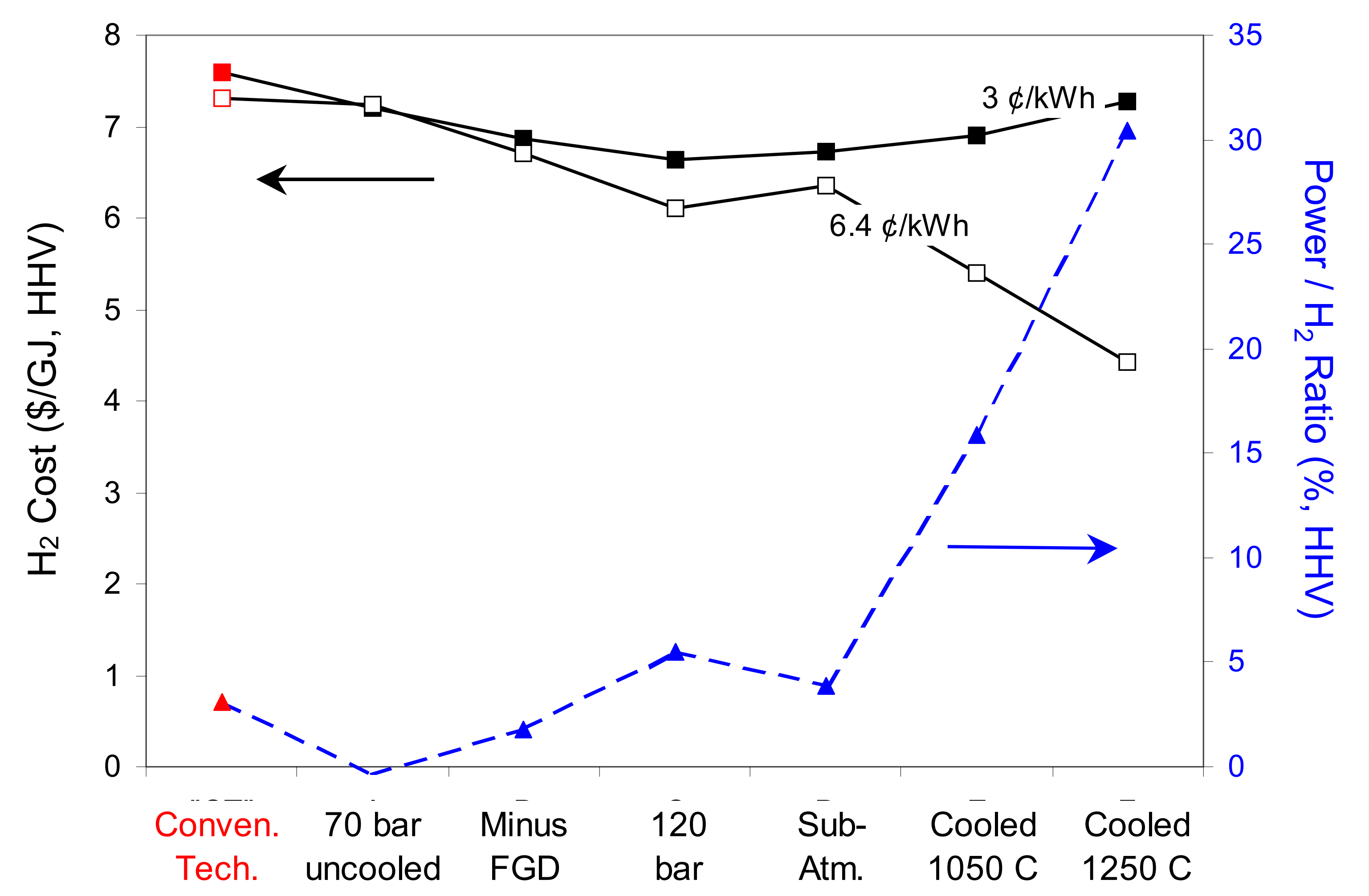
Modeling Results: Parametric Variations

	Description of system and/or change from one system to the next (in sequence). (Scale=1 GW_{th} HHV coal input)	*Hydrogen Recovery Factor (%)*	*"Effective" system efficiency (%, HHV)**	*CO_2-free H_2 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_2 output / HHV (coal input – coal saved**)

** Coal saved based on IGCC with CO2 capture, 34.9% HHV efficiency

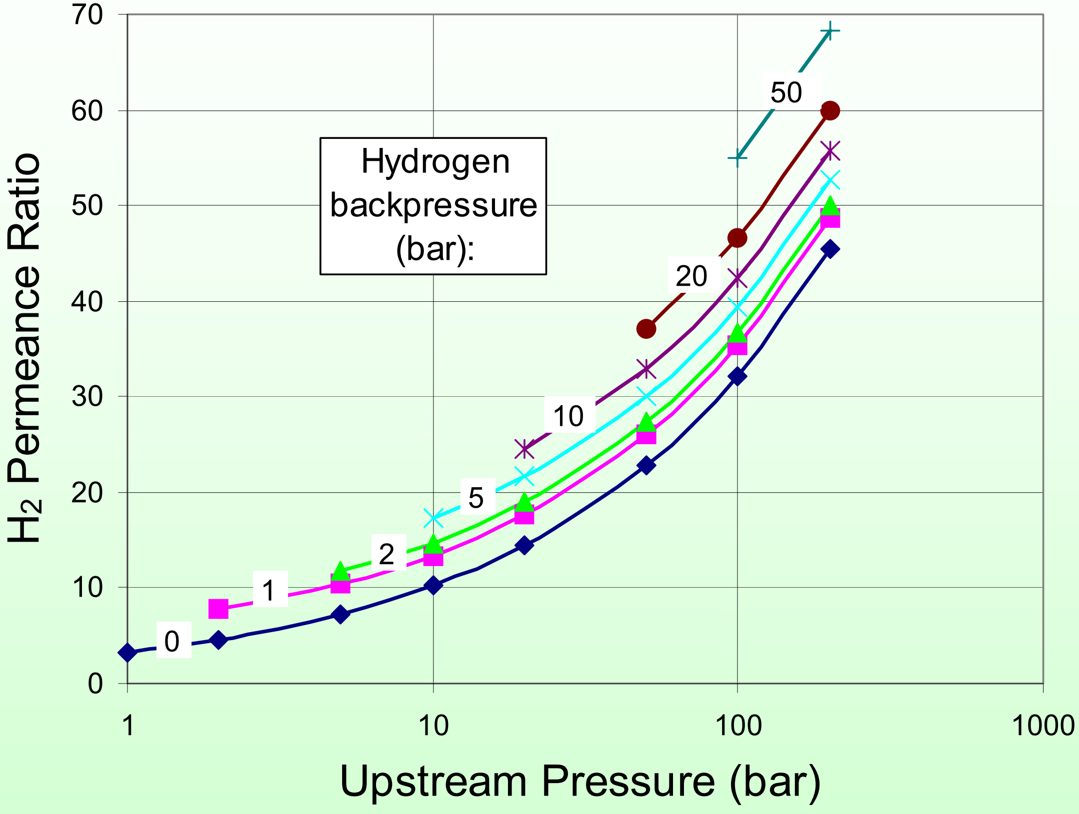
Modeling Results: Parametric Variations



- H_2 costs via membranes comparable to costs via “conventional technology”
- Power / H_2 ratio and electricity price are key to H_2 costs

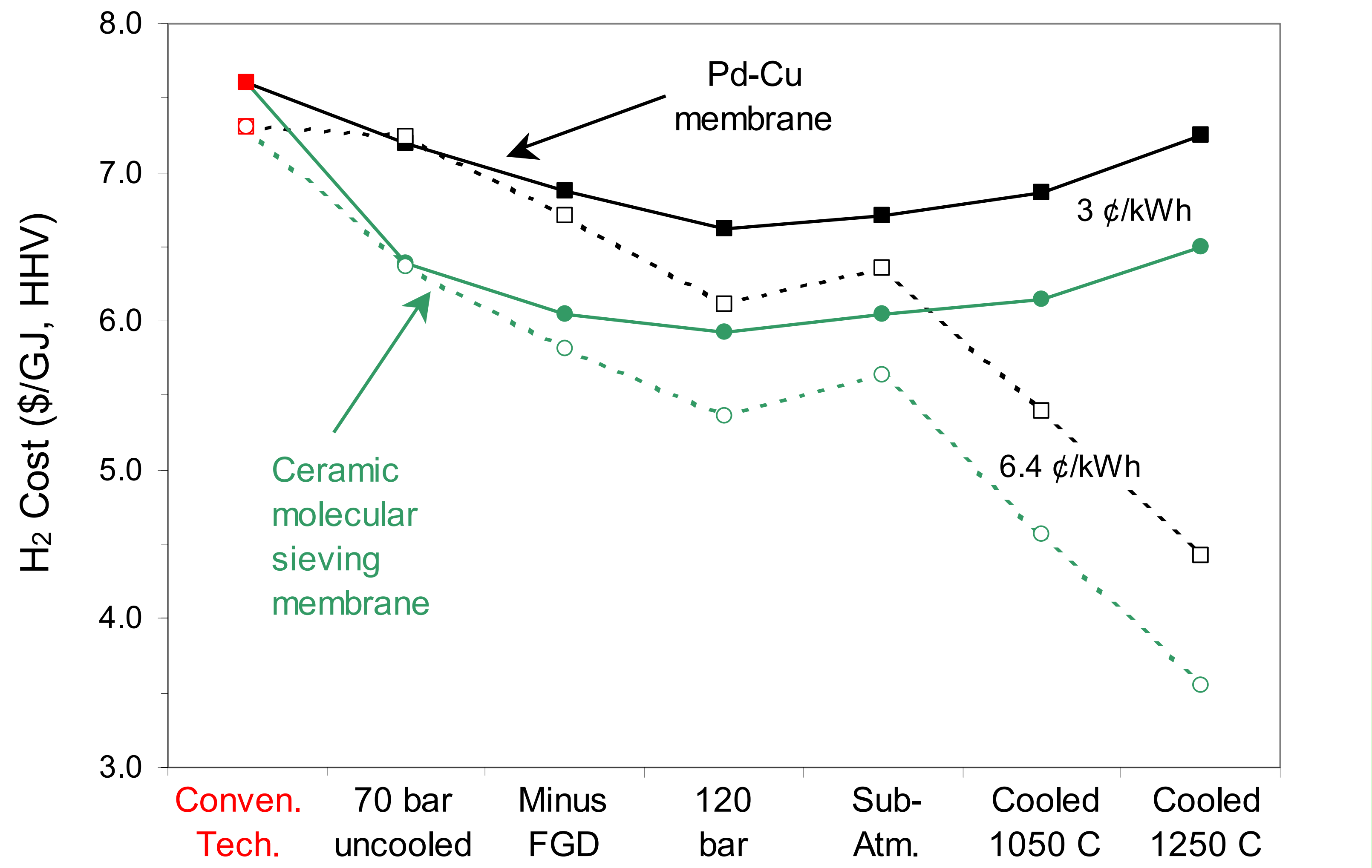
Oak Ridge Molecular Sieving Membrane

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



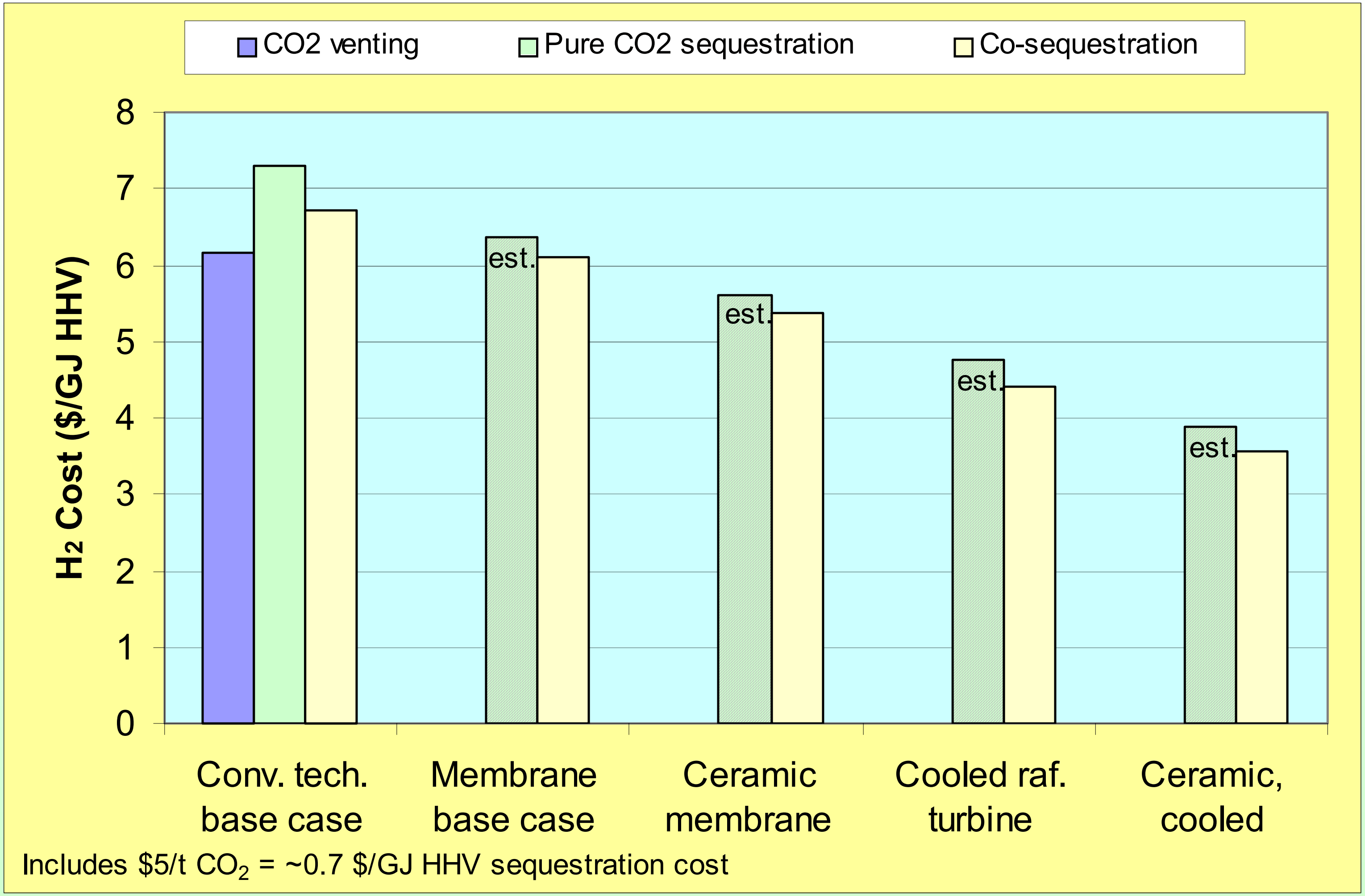
- Membrane permeance pressure dependence:
  - ceramic: $(P_{high} – P_{low})$ vs. metallic: $(\sqrt{P_{high}} – \sqrt{P_{low}})$
- Permeance increase up to factor of ~50 possible. Reduced purity.

Metallic vs. Ceramic Membrane reactors



- The increased permeance of ceramic membranes allows for potentially lower H_2 costs. (Same membrane unit price assumed, $3,021/m^2.)

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 $CO \rightarrow H_2$ conversion efficiency may not be a design goal.
- High *HRF* yields high system efficiency and low H_2 costs that are relatively insensitive to the electricity prices; v*ery* 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 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_2-CO_2 co-sequestration raises the system efficiency, may lower the cost of H_2, and may provide environmental benefits (Hg, etc.).
- Good system design can yield H_2 costs that are relatively insensitive to HSMR cost and H_2 recovery.
- Relative to "conventional technology" membrane reactors might lower the cost of H_2 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_2 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.