PRODUCTION OF HYDROGEN AND ELECTRICITY FROM COAL WITH CO_2 CAPTURE

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ABSTRACT

This paper summarizes a series of studies examining the prospective performance and cost of facilities that convert coal to H_2, co-product electricity and a stream of concentrated CO_2 (for sequestration). Synthesis gas is produced via oxygen-blown, entrained flow coal gasification, quench cooled and shifted to (primarily) H_2 and CO_2 via sulfur-tolerant water-gas shift (WGS) reactors. Our focus is on separating H_2 from the syngas and processing the carbon-bearing raffinate/purge gas to produce electricity and CO_2. We explore the use of novel inorganic membrane reactors for H_2 separation and compare their performance and cost with conventional gas separation technologies: CO_2 capture via solvent absorption followed by H_2 purification using pressure swing adsorption (PSA). This work highlights potential economic benefits of high system pressure, low H_2 purity, and co-sequestering CO_2 with sulfur-bearing waste gases, H_2S and SO_2.

I. INTRODUCTION

Carbon-free energy carriers, H_2 and electricity, are likely to play a critical role in a world with severe constraints on greenhouse gas emissions. Most fossil fuel decarbonization studies to date have focused on CO_2 capture in central-station electric power generation, which accounts for only ~30% of global CO_2 emissions. Much less is known about the prospects for producing CO_2–free H_2 from fossil fuels. This paper provides a *very brief* introduction to our research on the performance and cost of technologies for producing H_2 and/or electricity from coal with CO_2 capture; four detailed papers will be available [1,2,3]. Coal is a feedstock of particular interest because of its relative abundance, high carbon intensity, and low cost. Coal-to-H_2 plants based on gasification have been studied by Doctor et al. [4] and the Parsons Power Group [5]. The former investigated "conventional" gas separation technologies - CO_2 capture via glycol absorption and pressure swing adsorption (PSA) for H_2 purification - while the latter considered an inorganic (ceramic microporous) H_2 separation membrane reactor (HSMR) to produce H_2 and CO_2. Our work compares these two approaches in a consistent thermodynamic and economic framework and explores the design space for HSMR-based plants, seeking to understand the conditions that might lead to lower cost for H_2 from coal. The study focuses on one particular inorganic membrane technology, a 60/40% Pd/Cu dense metal film (for which experimental permeance data are available [6]) and explores how the efficiency and H_2 costs for such plants are affected by design parameters such as: H_2 recovery (i.e. percentage of H_2 extracted by the membrane), H_2 purity, raffinate turbine blade cooling, and system pressure. For the base case calculations, the plant products are: H_2 suitable for use in a PEM fuel cell (99.999% purity, 60 bar), co-product electric power, and dry (20 ppmv H_2O) supercritical CO_2 (at 150 bar) for pipeline transport and sequestration.

II. SYSTEM DESCRIPTION

The membrane-based plant is shown schematically in Fig. 1. In the conventional technology variant (not shown, but for base case parameters, see Table 1), the components downstream of the high-temperature WGS reactor are replaced by, in order: syngas cooler, low-temperature WGS reactor, H_2S and CO_2 physical absorption units, pressure swing adsorption (PSA) module, purge gas compressor, and gas turbine combined cycle (GTCC) for co-product electricity generation. The systems studied use an abundant, low cost feedstock and have essentially zero emissions. We seek simplified designs that minimize the cost of H_2 rather than maximize the system efficiency (e.g., by using syngas cooling via quench rather than more efficient and expensive high-temperature radiant/convective syngas coolers). System performance was modeled using Aspen Plus [7], GS [8], and Fortran (for membrane modeling).

Production of Shifted Syngas. Common to both conventional and membrane-based systems are system components designed to produce partially shifted syngas. High volatility Colorado bituminous coal (73.4% C, 5.1% H, 6.5% O, 1.3% N, 0.6% S, 1.4% moisture, 11.7% ash; HHV=29.58 MJ/kg) is gasified in an O_2-

blown, entrained flow, slurry-fed, slagging gasifier (Texaco) operating at 70 bar, modeled here using chemical equilibrium. Oxygen (95% purity) is produced in a stand-alone air separation unit (ASU). The raw syngas (~1330 C) passes through a quench cooler where it is scrubbed of particulates and water-soluble species, cooled to ~250 C, and saturated with water to a steam-to-carbon ratio (S/C) of at least 2.1 to avoid carbon formation in the WGS reactors downstream. The syngas passes through a high-temperature (~450 C) adiabatic WGS reactor that converts ~87% of syngas CO to H_2 and CO_2 (at exit: 3.0% CO, 23.3% CO_2, 34.2% H_2), raising the temperature by ~200 C. All WGS reactors employ sulfur-tolerant (fully sulfided) cobalt molybdate shift catalyst; H_2S traverses unaffected while COS is converted to H_2S.
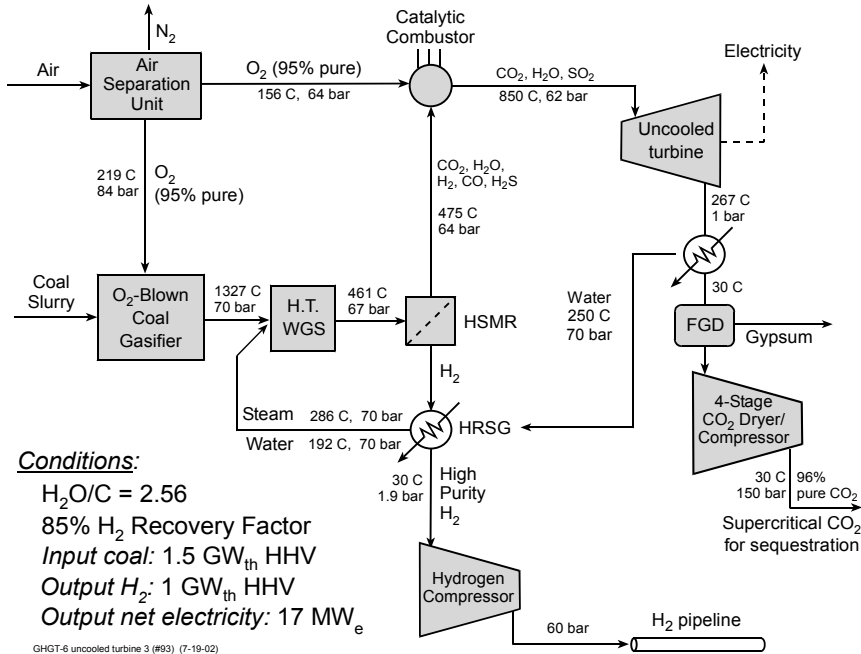

Fig. 1 Schematic of base case plant for producing hydrogen, electricity, and CO_2 from coal using a WGS hydrogen separation membrane reactor (HSMR).

Table I. Base case system parameters, inputs, and outputs (pure CO_2 sequest.).

	Conv. tech.	HSMR system
Gasification pressure, bar	70	
S/C ratio at H.T. WGS reactor	2.20	2.56
H_2 recovery factor, *HRF* (%)	85	
H_2, CO_2 final pressure, bar	60, 150	
Coal thermal input, GW (HHV)	1.50	1.48
H_2 production, GW (HHV)	1.00	
CO_2 disposed, ktonne CO_2/yr	2842	3265
Carbon emitted, ktonne C/yr	65.9	-
Air separation, MW	-40.8	-50.3
O_2 compression, MW	-24.8	-30.6
Gasification auxiliaries, MW	-14.4	-14.4
Other auxiliaries, MW	-13.5	-5.3
CC or raffinate turbine, MW	161.0	211.2
CO_2 compression, MW	-36.6	-51.5
H_2 compression, MW	-	-42.0
Net power output, MW	*30.9*	*17.1*

Conventional Gas Separation. In the conventional technology system, the shifted syngas is cooled to 200 C (in syngas coolers generating high- and low-pressure steam), shifted further in a low temperature WGS reactor, and cooled to 35 C in preparation for acid gas removal. Roughly 99% of the H_2S is removed from the syngas by physical absorption and converted to elemental sulfur via Claus and SCOT plants. A PSA unit separates 85% of the H_2 from the sulfur-free syngas; the H_2 exits at 60 bar and 99.999% purity. In the CO_2 venting case (included for comparison), the PSA purge gas is combusted to raise steam for a steam cycle that generates 144 MW$_e$ of electric power. In the CO_2 sequestration case, 92.7% of the CO_2 is scrubbed from the syngas by a secondary absorption tower (Selexol) located just downstream of the sulfur capture system. Because the PSA purge gas has a significantly higher calorific value than in the venting case, it is compressed to 18.2 bar and burned in a GTCC, generating 161 MW$_e$ of electricity. A CO_2-H_2S co-sequestration case was also investigated, where H_2S is captured along with the CO_2 in a single Selexol absorption unit; PSA purge gas is processed as in the pure CO_2 sequestration case. The estimated cost savings from co-sequestration (~\$0.25/GJ; see Tables II and III) are significantly smaller than would be expected using high-sulfur coals typical of integrated gasifier combined cycle (IGCC) plants; such coals (e.g. Pittsburgh #6, Illinois #6) typically have 4-7 times more sulfur than the Colorado coal employed here.

Membrane-based Gas Separation. In contrast to conventional technology, the membrane-based systems employ a WGS H_2 separation membrane reactor (HSMR) to shift the syngas and extract H_2. The maximum operating temperature of ~475 C insures fast chemical kinetics, and good WGS equilibrium performance is obtained by continuous removal of the H_2 product. Of the three major classes of inorganic H_2 permeable membranes: 1) ceramic molecular sieving, 2) dense ceramic ion transport, and 3) dense metal, we focus here on the third, in a shell-tube configuration, operating as an adiabatic plug flow reactor. The membrane is a sulfur-tolerant 10 μm dense metal film of Pd alloyed with 40% Cu, supported by a porous metallic tube, with an oxide interlayer to resist intermetallic diffusion between the Pd/Cu film and the substrate. Such membranes have been tested at temperatures of 300-600 C, pressures up to 35 bar, and H_2S concentrations as high as 10% in H_2 [6]. Based on permeation data obtained at 500 C for H_2 contaminated by 1000 ppmv H_2S, we model the HSMR using a H_2 permeance of 0.058 moles-m^{-2}-s^{-1}-bar$^{-0.5}$ [1]. Since data at higher H_2S levels are unavailable, we restrict our feedstock to relatively low sulfur bituminous coal (<0.6%wt) so that the H_2S concentration is below 1000 ppmv throughout the HSMR.

We employ here a one-dimensional, quasi-steady state HSMR model, with ideal gas behavior, and infinite H_2 selectivity (i.e. 100% H_2 purity). (We have also pursued more extensive 2D membrane modeling, examining the effects of: 1) radial dispersion of mass and heat, 2) reactor tube aspect ratio, 3) boundary layer

effects at the wall, and 4) sweep gas configuration [9].) The rate of H_2 permeation is assumed to be proportional to $P_r^n - P_p^n$, where P_r and P_p are the partial pressures of H_2 on the raffinate and permeate sides of the membrane, and $n=0.5$ (Sievert's law). As the syngas flows along the membrane reactor, the partial pressure of H_2 (and CO) falls - roughly exponentially with distance - as CO is converted to H_2 and permeates through the membrane. To quantify the extent of H_2 extraction from the syngas, we define a "H_2 recovery factor", *HRF*, as the ratio of moles of permeated H_2 to the moles of (H_2 + CO) in the entering syngas. As increasingly high *HRF* levels are sought, a point of diminishing returns is reached where investing in a larger membrane area yields insufficient gains in H_2 recovery. Fig. 2 shows an example where the falling average permeance flux with *HRF* leads to a dramatic increase in average membrane cost above *HRF*~95%. In this system, with a raffinate turbine incorporated to take advantage of a significant value for byproduct electricity, a very high *HRF* is generally found *not* to be necessary to obtain low H_2 costs.

A flow of inert (e.g. N_2 or steam) "sweep" gas on the permeate side – particularly in a counter-current configuration – can significantly increase the average flux of H_2 through the membrane by reducing P_p. A counter-current sweep also removes the sharp falloff in average flux shown in Fig. 2, enabling higher *HRF* values. The resulting H_2-sweep gas mixture is well suited for producing CO_2-free electricity in a GTCC (not discussed here) where hydrogen dilution is required to reduce NO_x emissions [2]. However, because this complicates the production of pure H_2 (albeit less so with steam than N_2), sweep gas is not used here.

The average H_2 flux through the membrane, and thus its area and cost, are determined by *HRF*, P_r, and P_p. Given a desired *HRF* and a fixed value for P_r, P_p controls the size and cost of the HSMR. In practice, we optimize the value of P_p to minimize the cost of the 60 bar H_2 product, balancing the capital and operating costs associated with compressing the permeate H_2 (lower at high P_p) against the capital cost of the HSMR (lower at low P_p). For the Pd/Cu membrane studied, the optimal value of P_p is typically 2-3 bar.

HSMR Configuration. Employing a single, adiabatic WGS HSMR for syngas shifting and H_2 removal affords both conceptual simplicity and potential capital cost savings. However, the WGS reaction kinetics are so rapid at these temperatures that most of the reaction and heat release are found to occur within the first ~20% of the reactor length, before a significant fraction of the H_2 can permeate through the membrane. In effect, the initial section of the HSMR acts much like a traditional adiabatic WGS reactor. We choose therefore to "split off" this section of the HSMR as a separate upstream adiabatic WGS reactor, shielding the relatively delicate membrane reactor from large temperature gradients, frequent catalyst changes, and trace contaminants in the syngas. The performances of these two configurations - labeled in Fig.2 "HSMR" and "HT-WGS+HSMR"
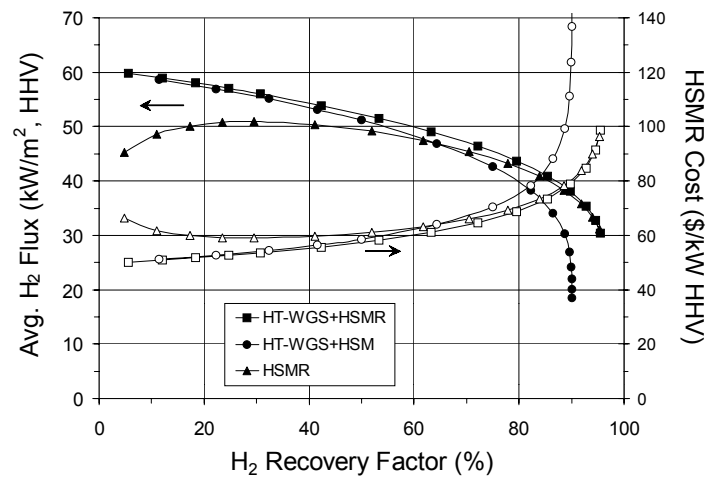

Fig. 2 Performance of the WGS and H_2 membrane separation system for various configurations (67.2 bar raffinate, $P_p = 1$ bar, membrane material cost = \$1500/m^2, no sweep).

respectively - are seen to be quite close above *HRF*~50%; the capital costs are also expected to be similar. Furthermore, the low CO conversion efficiency of the HSMR in the "split" configuration (since so little CO enters it) casts doubt on the necessity of a membrane *reactor* to follow the upstream HT-WGS reactor. Consider the effect of converting the HSMR into a pure H_2 separation membrane (HSM) permeator by replacing its catalyst bed with inert material. The performance of this configuration, "HT-WGS+HSM" in Fig. 2, is seen to be only slightly lower than that of the membrane reactor configurations below *HRF* ~80%; only at the highest *HRF* values (without sweep gas) is the performance unacceptable. These insights suggest that, if a very high *HRF* (significantly above 85%) is not required - and it generally is *not* for low H_2 cost - then a membrane <u>reactor</u> *per se* does *not* appear to be a key component of this intermediate temperature Pd/Cu membrane-based hydrogen separation system.

Co-Product Electricity Generation. We have studied a number of different strategies to produce electricity and sequesterable CO_2 from the hot, pressurized raffinate stream. 1) The configuration used here considers an O_2-fired (O_2 flow rate 5% above stoichiometric) combustor, followed by an uncooled turbine expander with a turbine inlet temperature (TIT) of 850 C. The turbine exhaust stream (35% CO_2, 63% H_2O) is readily processed into a relatively pure CO_2 stream for disposal. Catalytic combustion is required in the base case because the heating value of the raffinate stream is only 6.5 kJ/mole (LHV). 2) In order to raise the raffinate conversion efficiency, we have also investigated using a syngas expander followed by a raffinate turbine with open circuit steam cooling of the blades, enabling a significantly higher TIT (1250 C). While this configuration increases the electrical efficiency, the TIT can be achieved only by lowering the *HRF* to ~59% (to raise the raffinate heating value). 3) Since most of the water flowing in the turbine is separated by condensation before CO_2 compression, the raffinate conversion efficiency can also be increased by expanding to a sub-atmospheric pressure. However, because this design entails more expensive equipment (larger raffinate turbine and CO_2 compressor, vacuum heat exchangers and FGD, etc.), the effect on H_2 cost is unclear.

Turbine Exhaust Condensation and Optional Desulfurization. The base case turbine exhaust composition is 63% H_2O, 35% CO_2, 0.58% SO_2, and 93 ppmv (est.) SO_3 (plus non-condensable gases). The exhaust stream is first cooled with a standard heat exchanger to 180 C, just above its dew point, thereby avoiding formation of corrosive sulfurous and sulfuric acids, and then (optionally) desulfurized, dehydrated, and compressed. If desulfurization is chosen, we employ limestone slurry with forced oxidation (LSFO) flue gas desulfurization (FGD) to remove 98% of the SO_2 from the CO_2 stream down to 300 ppmv. If CO_2-SO_2 co-sequestration is chosen instead, a corrosion-resistant condenser is used to remove the acidic condensate (pH ~2.1) at 30 C from the raffinate turbine exhaust. (All SO_3 is assumed to condense out as sulfuric acid, but only 2.9% of the SO_2 is removed from the exhaust gas.) The exit stream, containing 85% CO_2, 9.6% H_2O, and 1.4% SO_2 (plus non-condensable gases), is sent on for compression and dehydration.

CO_2 Compression and Dehydration. Prior to pipeline transport, the CO_2 stream is dehydrated to 20 ppmv H_2O by flowing through a circulating triethylene glycol desiccant. Before dehydrating, the gas is compressed to 43 bar in three intercooled, centrifugal compressor stages. Selected components of the compressor and intercoolers are made of stainless steel to reduce corrosion from the acidic condensate. The dehydrated gas stream is compressed in a fourth compressor stage to the final 150 bar pressure, a typical value for supercritical CO_2 pipelines. The final stream composition is, in the membrane case with FGD sulfur removal, 95.9% CO_2, 2.0% Ar, 1.3% N_2, 0.7% O_2, 300 ppmv SO_2, and 20 ppmv H_2O.

III. SYSTEM ECONOMICS

The economic analysis is kept simple and transparent in order to facilitate re-evaluation by the reader using different assumptions. The cost of coal is $0.94/GJ (HHV), the US Energy Information Administration (EIA) estimate for the year 2020, when such a plant might be in operation. Interest during construction (IDC) is 16.1% of overnight capital (OC), based on a 4-year construction schedule and a discount rate of 10%/yr. The capital charge rate, applied to OC+IDC, is 15%/yr. Operation and maintenance (O&M) costs are 4% of OC per year. The capacity factor is 80%. The cost of aquifer CO_2 sequestration is $5 per metric ton of CO_2 (for a 100 km pipeline and 2 km deep injection well at this scale [2]). All plants are sized to produce 1000 MW H_2 (HHV), taking advantage of scale economies. Byproduct sulfur, slag, and gypsum are assumed to have no market value. All costs are reported in year 2001 U.S. dollars.

Capital Cost Estimation. Capital cost estimates for each component of the system (Table II) are taken from a variety of detailed studies in the open literature, mostly relating to coal-fired IGCC systems. Since the capital-intensive components of the H_2 production systems studied here are largely shared by IGCCs, we are able to calibrate our system cost and performance models by modeling comparable IGCC power plants (not discussed here). In this work, we adopt the capital cost estimates given in a recent comparison of advanced IGCC systems by Holt [10], based on detailed IGCC system studies by EPRI. The installed capital cost of the HSMR is assumed to be $3000/m^2, half of which is the cost of Pd (at $140 per troy oz). The cost of the raffinate turbine (combustor, expander, generator) is assumed to be 66% of the cost of a commercial gas turbine having the same *expander* power. "H_2S removal" costs include Claus and SCOT plants. The cost of the FGD unit is estimated via IECM [11], accounting for both the increased CO_2 concentration and specified SO_2 removal efficiency relative to a standard unit. Following Holt, for components within the gasification island (GI), our balance of plant (BOP) costs are 23% of GI capital, engineering fees (EF) are 15% of GI+BOP, process and project contingency are 15% of GI+BOP+EF (total= 1.63×GI). Power block components (e.g. turbines) and ASU include BOP, engineering and a 5% contingency (no further project contingency is included in OC). Other costs such as owners fees, royalties, start-up and pre-production costs, initial inventory, working capital, spare parts, and land are *not* included in this analysis. Table II presents a comparison of the base case system costs (for the pure CO_2 sequestration case). "Error bars" or uncertainty limits for our cost model, based on the disparity among equally credible estimates of component capital costs in our cost database, are estimated to be ±11% (of OC) or ±$0.55/GJ (HHV). Note in Table II that, since the capital costs of gas separation and power generation comprise less than 30% of the total capital cost, it is not surprising that these two different gas separation strategies do not yield markedly different costs for H_2.

Table II. Base case system economics (pure CO_2 sequestration case). Installed capital costs include apportioned BOP and general facilities, engineering, and process/project contingencies.

System Component Cost (Million $)	Conv. tech.	HSMR system
Coal prep and handling	52.9	52.4
Air separation unit	129.9	150.7
O_2 compressor	21.2	24.5
Gasifier and quench/scrub	125.9	124.8
Sour WGS reactor(s)	35.4	11.6
H_2S removal or FGD	31.3	27.4
Selexol CO_2 recovery	36.5	-
PSA or HSMR	44.8	89.2
GTCC or raffinate turbine	105.7	28.6
CO_2 compression	46.5	65.5
H_2 compression	-	50.7
Overnight capital (OC)	*630.0*	*625.4*
Interest during construction	100.9	100.2
Total Capital Requirement	**730.9**	**725.6**
H_2 Cost Component	**$/GJ (HHV)**	
Capital (at 15%/yr)	4.35	4.31
O&M (at 4%/yr of OC)	1.00	0.99
Coal (at 0.94 $/GJ HHV)	1.41	1.39
Electricity (at 3.0 ¢/kWh)	-0.26	-0.14
CO_2 disposal (at 5 $/t CO_2)	0.56	0.66
Total H_2 Production Cost	**7.06**	**7.20**

Value of Co-produced Electric Power. The primary economic metric in this study is the cost of H_2, which depends on the price of co-product electricity. This price strongly influences the optimal system design, especially the H_2-to-electricity ratio or *HRF*. We bracket the range of possibilities by considering: 1) a low price, 3.0 ¢/kWh, characteristic of natural gas-fired GTCC (Frame 7H) with CO_2 venting and average gas price in 1999 of 2.5 \$/GJ HHV, and 2) a relatively high price, 5.5 ¢/kWh, characteristic of coal IGCC (Frame 7H) with conventional CO_2 capture and sequestration (calculated using the performance and cost models described here). The low price is employed below in the "base case" economic analyses.

IV. RESULTS AND DISCUSSION

Effect of Hydrogen Recovery Factor. In broad terms, the H_2 separation unit - either membrane reactor or PSA - splits the shifted syngas into separate streams of H_2 and electricity (via the raffinate turbine or PSA purge gas GTCC). The extent of H_2 production, quantified by the *HRF*, strongly affects the both the efficiency and economics of H_2 production. For this two-product system, it is useful to introduce an "effective hydrogen production efficiency", η^{eff}, defined as the HHV of the H_2 product divided by the HHV of *net* coal, where net coal is the input coal minus the coal that would otherwise be required to produce the electricity (via a coal-fired Frame 7H IGCC with CO_2 sequestration operating at a HHV efficiency of 37.1%). In the membrane system, η^{eff} rises monotonically with increasing *HRF* (Fig. 3) because the

Fig. 3 Efficiency and cost of hydrogen production as a function of *HRF* (HSMR, pure CO_2 sequestration case).

~29% HHV efficiency (coal to electricity) of the raffinate turbine "pathway" is less than that of the IGCC that it displaces. (The raffinate turbine has both a significantly lower TIT and less efficient utilization of low quality heat than a standard GTCC.) Increasing *HRF* minimizes relatively inefficient electricity production and thereby increases η^{eff}. (This principle is likely to hold also for H_2-electricity plants based on conventional gas separation technology.) The *HRF* also affects the economics of H_2 production. In Fig. 3, for example, low electricity prices drive HSMR system design to high *HRF* values, while high electricity prices yield H_2 whose price is broadly insensitive to *HRF* over a wide range of values. (The discontinuity observed in the H_2 cost curves between *HRF*=52% and 57% reflects the savings from switching from 3 to 2 ASU trains as the oxygen requirement for the raffinate turbine falls with increasing *HRF*.)

We have chosen base-case configurations with a relatively high *HRF*=85%, consistent with our primary interest in H_2 (rather than electricity) production. Fig. 3 shows that *HRF* near this value yields high system efficiency and low H_2 costs that are insensitive to the electricity price. In contrast, systems having relatively low *HRF* are complicated by the strong coupling between H_2 cost and electricity price. Notably, the use of a cooled raffinate turbine increases the electrical efficiency by lowering the *HRF* to ~59% (as discussed earlier); because the overall "effective" efficiency of the system, η^{eff}, falls monotonically with decreasing *HRF* (Fig. 3), η^{eff} is lower and H_2 is much more expensive (Table III, line 4) than with an uncooled turbine.

Cost of H_2 Purity. In conventional technology systems described thus far, H_2 purity is very high (99.999%), matching that of the membrane-based system and suitable for use in a PEM fuel cell. Less pure "fuel grade" H_2 (93.5% H_2, 3.3% CO_2, 1.2% Ar, 1.0% N_2, 0.9% CO), adequate for H_2 combustion applications such as internal combustion engines or gas turbines, can be produced more cheaply by omitting from the conventional plant the PSA unit, purge gas compressor, and gas turbine, and downsizing the steam cycle. This reduces the OC by more than 25%, and converts the plant from a net exporter (31 MW_e) to a net importer (-49 MW_e) of power. At an electricity price of 3.0 ¢/kWh, the cost of delivered H_2 drops by ~1 \$/GJ HHV (Table III, line 2).

Table III. Base case thermodynamic and economic performance comparison between various membrane vs. conventional technology systems. (HSMR results for CO_2 venting, which involve raffinate combustion in air instead of O_2, are approximate.)

	Description	*CO₂ venting*		*Pure CO₂ sequestration*		*CO₂-S Co-seq.*
		η^{eff} *(%)*	*\$/GJ*	η^{eff} *(%)*	*\$/GJ*	*\$/GJ*
Conv. Tech.	*Base case*	71.6	5.55	69.4	7.06	6.79
	Fuel grade H_2	75.5	4.75	74.7	6.05	5.81
HSMR-Based System	*Base case*	75	5.3	69.1	7.20	6.97
	Cooled raf. turbine	66	4.9	57.8	8.45	8.14
	High perm HSMR	76	4.7	69.9	6.59	6.35

Effect of Membrane Reactor Cost and Type. The prospective cost of the membrane reactor is less well known than that of any other system component. Varying the installed cost of the HSMR between \$1,000/m² and 5,000/m² is found to alter the H_2 cost by less than \$0.9/GJ HHV. There exists an "optimal" *HRF* value at which the cost of product H_2 is lowest. Above some high *HRF*, the H_2 cost rises because of the rapidly increasing cost of the membrane (in the absence of sweep gas). Expensive membranes encourage
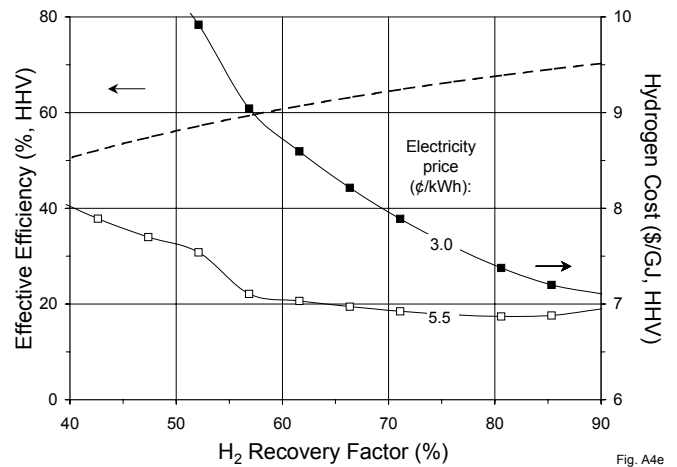
systems with relatively low *HRF* values, while low-cost membranes allow larger membrane reactors to recover more H_2 from the syngas. Preliminary calculations indicate that replacing the 60/40 Pd/Cu membrane with a high permeance ceramic molecular-sieving membrane (~10× as permeable at optimized P_p; also costing \$3,000/m^2) might reduce the cost of H_2 by as much as \$0.6/GJ HHV (Table III, line 5) due to lower costs for both H_2 separation and compression. Such microporous membranes involve a tradeoff between permeance and selectivity; in a single-stage configuration, H_2 purity will suffer (99.5%, est. [5]), while multiple-stages yield lower average H_2 flux. Note also that, since n=1 for these membranes, raising the system pressure (e.g. to 120 bar, see discussion below) will significantly increase the improvement in permeance afforded by this technology. We are currently researching systems that best utilize such membranes.

Effect of Gasifier Pressure. Thermodynamic arguments suggest that increasing the system pressure will improve overall efficiency. As with a steam cycle, gasification benefits from relatively low cost pressurization of an incompressible feedstock followed by turbine expansion of the hot product gases (although, in gasification, oxygen must also be compressed), a situation that clearly favors high system pressures. Moreover, the gas separation technologies employed here to produce high pressure H_2 and CO_2 are generally more efficient and less costly with increasing pressure. Our calculations confirm these concepts, exhibiting effective efficiencies of membrane-based H_2 production that rise monotonically with increasing gasifier pressure between 30 and 120 bar. Further study will be required to understand better the relation between pressure and cost, and how high the pressure can be raised before materials issues, corrosion, etc. become limiting factors. Recent studies highlight economically promising IGCC systems at 85 and 110 bar [12].

V. CONCLUSIONS

We have constructed a consistent framework for estimating the performance and cost of plants that convert coal to H_2 and electricity with CO_2 sequestration and have used it to investigate systems using both membrane-based and conventional H_2 separation. We plan to extend this methodology to study other feedstocks (petroleum residuals, natural gas, biomass, etc.) and other novel technologies. Our parametric investigation of membrane-based plants indicates that: a) the 60/40 Pd/Cu membrane does not appear to be a likely candidate for commercialization because of its limited potential for cost reductions relative to conventional technology, b) high permeance microporous membranes offer greater (although still modest) potential cost reductions, albeit with reduced H_2 purity, and c) when following an upstream HT-WGS reactor, the Pd/Cu WGS membrane *reactor* appears to have little advantage over the membrane *permeator*. In H_2 plants with co-product electricity, high *HRF* yields high system efficiency and low H_2 costs that are relatively insensitive to the electricity prices. However, *very* high *HRF* is typically not required to obtain low H_2 costs. For systems with CO_2 capture, high H_2 purity is costly; "fuel grade" H_2 can be produced with conventional technology at significantly lower cost. Higher gasifier pressures yield higher system efficiencies and potentially lower H_2 cost. Finally, co-sequestration of sulfur-bearing species and other pollutants with CO_2 may provide key economic and environmental benefits. The merits of co-sequestration are largely unexplored; the savings from reduced in-plant sulfur treatment must be compared with possible added compression and pipeline costs, and consequences at the storage site must be better understood.

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