Carbon Dioxide Capture for Storage in Deep Geologic Formations – Results from the CO_2 Capture Project

Capture and Separation of Carbon Dioxide from Combustion Sources

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Chapter 18

DESIGN, SCALE UP AND COST ASSESSMENT OF A MEMBRANE SHIFT REACTOR

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ABSTRACT

The objective of the design, scale up and cost assessment of membrane shift reactor project was to produce a detailed design and cost estimate of a commercial scale membrane water gas shift (MWGS) reactor. The requirements for the reactor were:

- retentate dry CO_2 molar content—90%;
- permeate LHV—150 Btu/SCF;
- hydrogen extraction $>90\%$;
- feed/retentate pressure drop <2.76 bar (40 psid);
- sweep/permeate pressure drop <0.34 bar (5 psid).

The flux of hydrogen through the membrane was approximately 234 MMSCFD.

Two feasible MWGS reactor designs have been developed, which use either a planar or a tubular hydrogen separation membrane. The planar membrane is composed of a curved membrane supported by a corrugated Type 430 stainless steel sheet. Finite element analysis which considered the pressure, gravity, and differential thermal expansion loadings indicates that it is structurally adequate for 41.1 bar (600 psid) pressure loading at 450 °C (842 °F). A second MWGS reactor concept is based on a tubular membrane sized appropriately to contain the high pressure inside the tubes.

An analysis tool to permit examination of different arrangements for the MWGS reactor was developed and bench-marked against the model developed in Phase I. This analysis tool determined the membrane area required for each reactor concept. The planar membrane reactor has the following characteristics:

- a multi-pass cross flow arrangement;
- forty stacks of 159 membrane wafer panels, 2 m (6.55 ft) long by 3.05 m (10 ft) tall by 0.305 m (1 ft) wide;
- total active membrane surface area of 5357 m^2 (57,662 ft^2);
- catalyst placement between membrane stacks, catalyst gap of 0.15 m (6 in.);
- length is approximately 26.8 m (88 ft).

The tubular membrane reactor concept has the high-pressure feed gas inside the tubes and the sweep gas flowing across the tube bank. The tube length was varied to meet the feed-side pressure drop constraint for a given tube diameter, and the tube pitch and baffle arrangement were varied to meet the sweep-side pressure drop constraint. The characteristics of the tubular reactor include:

- four separate membrane reactors interstaged with catalyst reactors;
- each membrane reactor has 9730 U-tubes, 1.07 cm (0.424 in.) ID, 4.2 m (13.8 ft) long;
- total active membrane surface area of 5685 m^2 (61,193 ft^2);
- each membrane reactor is about 7.6 m (25 ft) long and 3.2 m (10.5 ft) diameter.

The baseline planar design places the membrane internals inside of a conventional pressure vessel. The tubular membrane reactor concept, which was not designed as rigorously as the planar options, was based on standard shell and tube construction. The vessels are designed according to Section VIII, Division 1 of the ASME Boiler and Pressure Vessel Code, for an internal pressure of 41.4 bar (600 psig) at a vessel metal temperature of 454 °C (850 °F). The estimated order-of-magnitude cost to fabricate the baseline planar reactor is approximately $19 million. The estimate is based on input from various suppliers of materials and services, as well as manufacturers specializing in the fabrication of components specified for the reactor. In many cases, where detailed information is not yet developed, rough cost estimates were provided by vendors based on similar work and standard cost models. The alternative tubular concept was estimated at approximately $12 million.

INTRODUCTION

The objective of the design, scale up and cost assessment of membrane shift reactor project was to design and estimate the cost of a membrane water gas shift (MWGS) reactor. The two products from this reactor will be: (1) a high-purity hydrogen stream, which could be used in boilers and furnaces and (2) a concentrated, high-pressure CO_2 stream, which can be sent to sequestration. The performance requirements for the MWGS reactor were as follows:

- retentate dry CO_2 molar content—90%;
- overall carbon recovery—90%;
- permeate LHV—150 Btu/SCF;
- hydrogen extraction $>90\%$;
- feed/retentate pressure drop <2.76 bar (40 psid);
- sweep/permeate pressure drop <0.34 bar (5 psid).

The flux of hydrogen through the membrane was approximately 234 MMSCFD.

The MWGS reactor would combine the WGS and CO_2 removal steps into one process. The potential benefits are lower capex and opex, and a simplified process. In addition, since the CO_2 is produced at an elevated pressure, sequestration compression costs will be lower.

STUDY METHODOLOGY

Structural Analysis Methodology

Finite element analyses of the membranes and support structure were accomplished. Models of a membrane wafer repeat unit and a wafer stack assembly repeat unit were accomplished.

1. *Membrane wafer repeat unit model*: The wafer repeat unit model was utilized for the detailed pressure and differential thermal expansion loading analysis. A typical repeat unit model mesh is shown in Figure 1.
2. *Wafer stack assembly repeat unit model*: The model geometry is shown in Figure 2. It is a region within the middle of a wafer stack assembly bounded by the mid-plane of a wafer and the mid-plane of the gap between wafers in the longitudinal direction, the plenum cover and the mid-plane of the wafer in the transverse direction, and the mid-plane of the gap between wafers and the mid-plane of the second adjacent gap between wafers in the vertical direction.

A pressure of 41.4 bar (600 psid) was applied to the exterior syngas wetted surfaces, which conservatively assumes that the sweep side is vented to atmosphere during startup or in a faulted condition. The syngas duct was exposed to 60 psid differential pressure which assumes that the internal syngas side is at design pressure, and the external syngas side is 50% beyond the nominal pressure differential expected from inlet to outlet of the vessel. The operating temperature was assumed to be 450 °C (842 °F). Mechanical properties for the alloy to be used for the membrane material are not yet available. Therefore, the properties of commercial purity membrane material were used. Type 430 stainless steel was preliminarily selected for

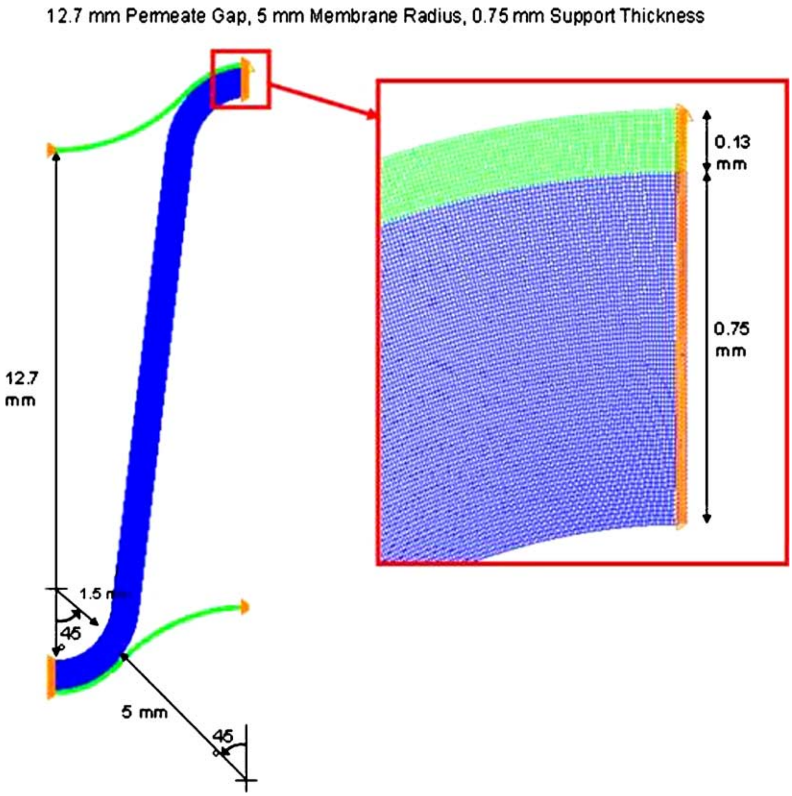
12.7 mm Permeate Gap, 5 mm Membrane Radius, 0.75 mm Support Thickness

Figure 1: Membrane wafer repeat unit model.

the support structure stainless steel (in order to minimize differential thermal expansion concerns). The material properties used are shown in Table 1.

MWGS Reactor Performance Model

A model of the MWGS reactor was developed to facilitate design activities and sensitivity studies of important design parameters. The model included:

- membrane kinetics;
- catalyst kinetics for a commercially available bulk catalyst;
- heat transfer between the feed and permeate streams.

A comparison of the output from this model (SOFCo) was compared to the output from the ASPEN-based model developed in Phase I of the program. The results, summarized in Table 2, show the agreement is adequate for design purposes.

RESULTS AND DISCUSSION

Structural Analysis of Hydrogen Separation Membrane

Structural analysis of the support structure for a hydrogen separation membrane was accomplished. The analysis considered pressure, and differential thermal expansion loading. Designs that satisfy stress and

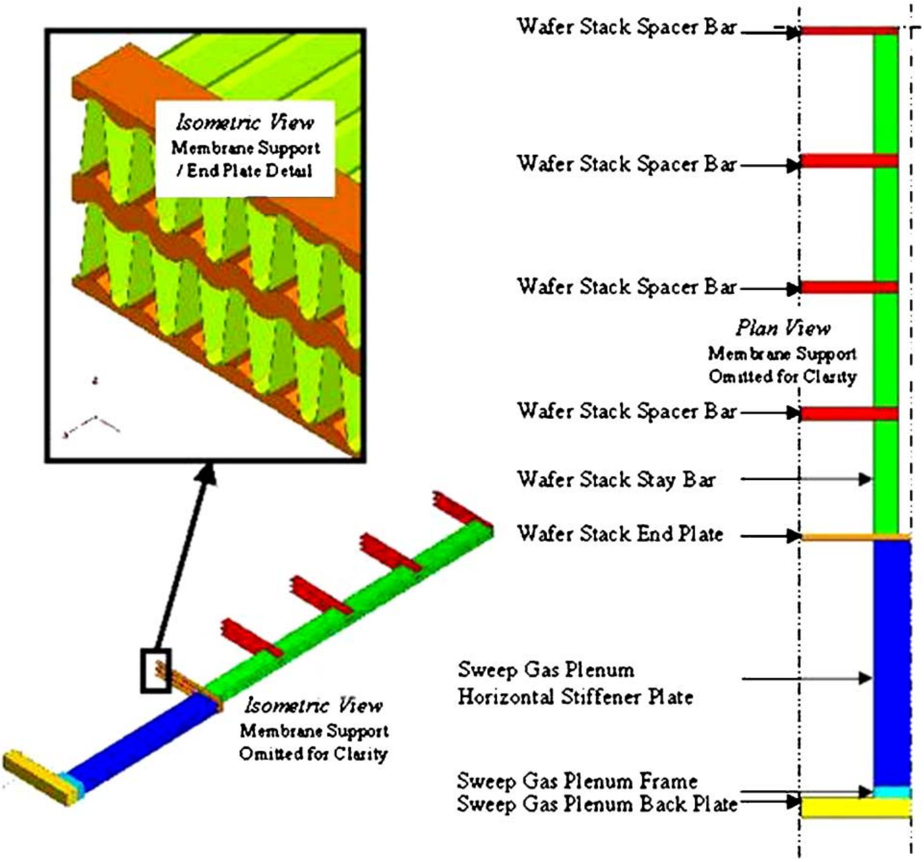

Figure 2: Wafer stack assembly repeat unit model.

instability constraints for several permeate gap heights were found. The analysis and resulting designs are summarized below.

Repeat unit analysis—pressure loading. Typical results for a repeat unit model subjected to pressure loading are shown in Figure 3.

Three permeate gap heights were considered: 2.54, 12.7, and 25.4 mm. For these cases, the membrane stress was relatively insensitive to the support thickness. A membrane mid-radius of 5 mm provided acceptable stress in the membrane. The required support thickness was 0.25, 0.75, and 1.05 mm for the three cases. The analysis results are summarized in Table 3.

Figure 4 shows the recommended support thickness as a function of the permeate gap height. It is noted that the relationship is not smooth since different regions are limiting throughout the range.

Additional analyses which considered plane strain conditions (i.e. no out-of-plane displacement) and friction $(mu = 1.0)$. It was found that these conditions were less limiting than the base conditions (plane stress and frictionless).

TABLE 1
MATERIAL PROPERTIES

Material property	**Value**	**Source**
Membrane material		
Modulus of elasticity	127.6 GPa	[1]
Poisson ratio		[1]
Thermal expansion		20–500 °C [2]
Density	6.1 g/cm^3	[1]
Yield strength	454 MPa	20 °C, annealed, sheet [3]
Ultimate strength	526 MPa	20 °C, annealed, sheet [3]
347 Stainless steel		
Modulus of elasticity		[4]
Poisson ratio		
Thermal expansion		[4]
Density	7.96 g/cm^3	[4]
Yield strength	139 MPa	454 °C, plate [5]
Ultimate strength	405 MPa	454 °C, plate [5]
430 Stainless steel	(Fe-17% Cr)	
Modulus of elasticity	160 Gpa	[5]
Poisson ratio	0.3	[2]
Thermal expansion	11.2 ppm/°C	[5]
Allowable primary membrane stress	99 MPa	454 °C, plate [5]

TABLE 2
COMPARISON OF SOFCO MWGS MODEL OUTPUT TO ASPEN MODEL

	Baseline 315 °C			**400 °C Case 1**			**400 °C Case 2**		
	ASPEN	**SOFCo**	**% Diff.**	**ASPEN**	**SOFCo**	**% Diff.**	**ASPEN**	**SOFCo**	**% Diff.**
Operating conditions									
Membrane area, m^2	17,325	17,325		11,410	11,410		11,780	11,780	
Catalyst volume to area, m^3/m^2	0.100	0.100		0.005	0.005		0.005	0.005	
Nitrogen sweep gas, kmol/h	9100	9100		9100	9100		9100	9100	
Steam sweep gas, kmol/h	8800	8800		8800	8800		10,200	10,200	
Feed-side pressure, bara	35.00	32.20		32.20	32.20		32.20	32.20	
Sweep side pressure, bara	3.00	3.35		3.35	3.35		3.35	3.35	
Performance comparisons									
Average H_2 flux (mol/m^2 s)	0.186	0.185	−0.4%	0.275	0.277	0.7%	0.272	0.274	0.6%
H_2 recovery, %	95.3%	95.0%	−0.3%	93.3%	93.9%	0.7%	95.2%	95.7%	0.5%
CO_2 purity (dry)	90.2%	88.90%	−1.3%	86.86%	86.84%	0.0%	90.04%	89.97%	−0.1%
CO out, ppm	995	1000	−0.5%	3000	4077	−35.9%	2000	3063	−53.1%
Permeate outlet temperature, °C	347.5	346.5	0.3%	419.9	417.3	0.6%	421.9	420.4	0.4%
Retentate outlet temperature, °C	327.7	329.0	−0.4%	421.8	422.7	−0.2%	418.0	418.9	−0.2%

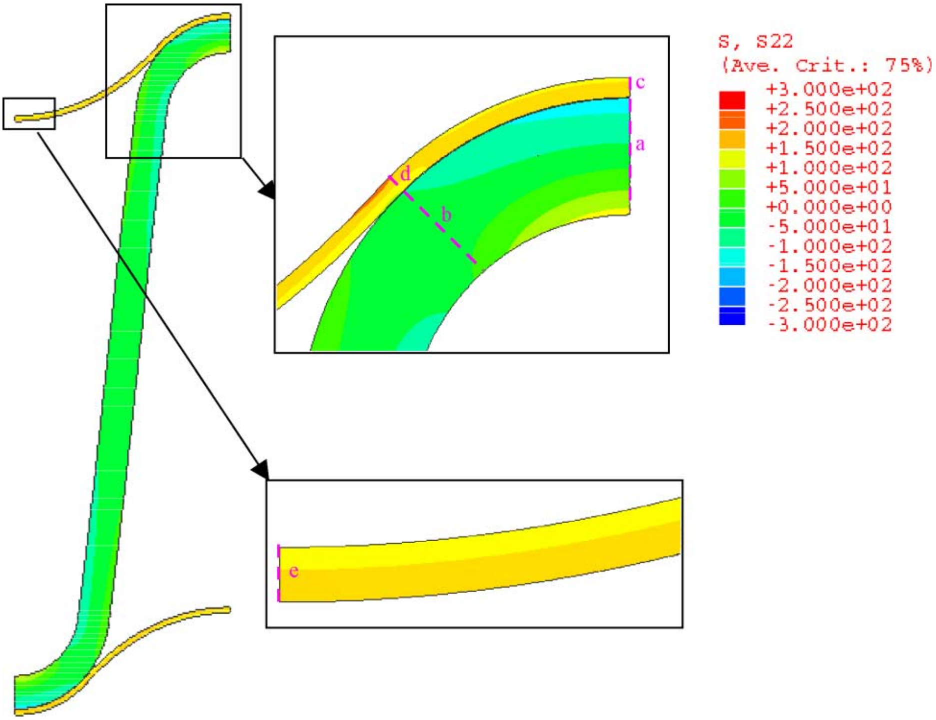

Figure 3: Pressure loading analysis.

TABLE 3
ANALYSIS RESULTS FOR PRESSURE LOADING

Permeate gap height (mm)	**Membrane radius (mm)**	**Support thickness (mm)**	**Location in cross-section**	**Fraction of allowable stress (%)**				
				Support		**Membrane**		
				a (top)	**b (edge)**	**c (top)**	**d (edge)**	**e (bottom)**
2.54	5.0	0.25	Center	93	95	99	99	90
			Bottom	41	66	77	54	65
			Top	83	63	56	78	54
12.7	5.0	0.75	Center	18	40	94	95	89
			Bottom	73	26	68	39	68
			Top	97	35	57	88	51
25.4	5.0	1.05	Center	14	35	93%	95	89
			Bottom	83	26	66	35	69
			Top	99	40	58	92	50

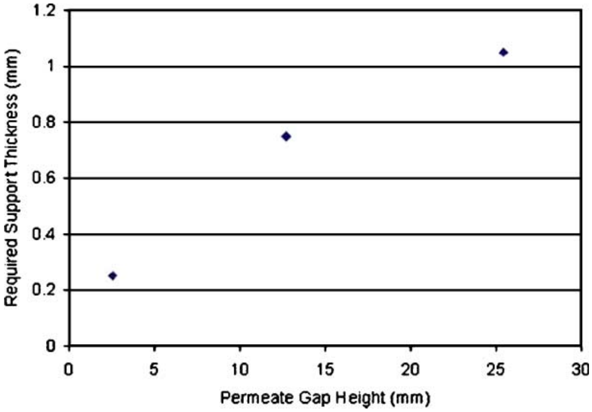

Figure 4: Effect of permeate gap height on required support thickness.

MWGS Reactor Design Basis

The flux of H_2 through the membrane is given by the expression:

$$N_{H_2} = -k'(P^{0.5}_{H_2,\mathrm{ref}} - P^{0.5}_{H_2,\mathrm{perm}})$$

where: k', permeance $= 0.5A_0 \exp(-E_A/RT)$ (mol/m^2 s Pa$^{1/2}$); A_0 is pre-exponential factor (mol/m^2 - s Pa$^{1/2}$); E_A is activation energy (J/mol) and R is 8.314 J/mol K.

The flux parameters were initially based upon the Phase I results for preliminary sizing estimates. Later in the program, the flux for the membrane material was expected to be much higher and so the values were increased. The preliminary sizing estimates discussed here were based on the Phase I flux, but the final design was based on the mid-Phase II flux. The numbers are summarized in Table 4.

TABLE 4
FLUX PARAMETERS FOR H_2 SEPARATION MEMBRANES

	A_0 Pre-exponential factor mol/m^2 s Pa$^{1/2}$	**E_a Activation energy J/mol**	**k' Average permeance mol/m^2 s Pa$^{1/2}$**
Phase I	2.87E-02	24,896	4.312E-04 (at 440 °C)
Mid phase II	1.49E-03	2420	9.789E-04 (at 425 °C)

The flow and pressure drop requirements for the MWGS reactor are shown in Table 5.

MWGS reactor options. Using the MWGS reactor model, a number of studies were performed to examine the required amount of membrane and catalyst for different conditions and reactor arrangements. Four different configurations were considered:

1. counter-flow;
2. baffled counter-flow;
3. cross-flow;
4. multi-pass cross-flow.

TABLE 5
FLOW STREAMS FOR MWGS REACTOR

Flow stream	Temperature	Pressure	Constituent	Mole flow (kmol/h)	
				In	Out
Sweep side	400 °C in, 415.2 °C out	3.34 bara in, 3.0 bara out	H_2	0	11,666
			H_2O	8800	8800
			N_2	9100	9100
			CO	908.434	65.856
Feed side	400 °C in, 425.1 °C out	35 bara in, 32.2 bara out	H_2O	10,135.650	9,293.072
			H_2	11,222.23	518.521
			CO_2	4,837.563	5,680.141
			N_2	34.561	34.561
			CH_4	3.474	3.474
			Other	8.409	8.409

The multi-pass cross-flow arrangement was chosen for development to a conceptual level design based upon the comparison of perceived advantages and disadvantages shown in Table 6.

TABLE 6
COMPARISON OF PERCEIVED ADVANTAGES AND DISADVANTAGES FOR REACTOR OPTIONS

Option	Advantages	Disadvantages
Counter-flow	Minimum membrane required	Large wafer plates difficult to braze Catalyst packing between plates
Baffled counter-flow	Minimum membrane required	Large wafer plates difficult to braze Baffle plates
Cross-flow	Assembly of manifolds Smallest wafer package	22% more membrane than counter-flow option
Multi-pass crossflow	Assembly of manifolds	8% more membrane than counter-flow option

MWGS Reactor Packaging and Performance

The reactor packaging required an iterative approach to satisfy the surface area, pressure drop, and structural design requirements. At this stage, the flux performance of the membranes was based upon the mid-Phase II results given in Table 4. The design activities are discussed below.

Pressure drop performance

The reactor flow conditions are shown in Table 6. Note that the outlet conditions are dependent on the particular reactor flow configuration, shown here are for the final multi-pass cross-flow configuration. On the feed/retentate side of the reactor, the allowable pressure drop was specified as 2.76 bar (40 psid). On the sweep/permeate side of the reactor, the allowable pressure drop was specified as 0.34 bar (5 psid). The flow paths through the reactor are represented in Figure 5.

Feed/retentate pressure drop

The feed/retentate side pressure drop is a function of:

- the pressure drop between the corrugated membrane panels;
- the pressure drop through bulk catalyst between stacks.

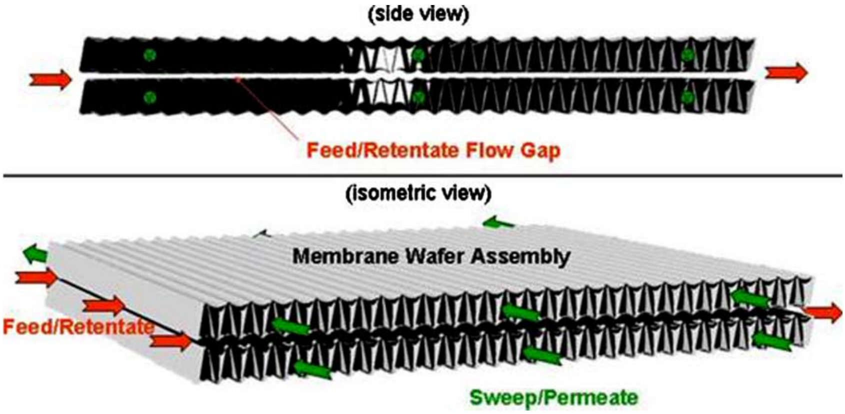

Figure 5: MWGS reactor flow path representation.

The packaging of the membrane is conceptualized as some number of stages down the flow direction of the reactor. This determines the amount of flow per unit area, which is inversely proportional to the number of stages. As the number of stages increases, the flow per unit area increases as well as the length of the flow path. The gap between the membranes was varied as needed to meet the pressure drop target.

The relationship between the number of stacks, the wafer panel pitch, and the approximate size of the reactor is shown in Figure 6. As shown, the required reactor internal diameter (approximated by the diagonal of the wafer stack and plenum) tends to asymptotically approach a lower limit as the number of stacks is increased. Based on this relationship, a package was based upon 40 stacks. This results in a stack with the following characteristics:

- wafer to wafer pitch is 1.905 cm (0.75 in.);
- 159 wafers per stack;
- feed/retentate flow gap is 0.521 cm (0.205 in.);
- wafer panel length is 2 m (6.55 ft);
- stack height is 3.05 m (10 ft);

Sweep/permeate side pressure drop performance
On the sweep side, the pressure drop target is met by increasing the sweep side flow area inside of the wafer. Figure 7 shows the sweep side pressure drop as function of the number of stacks corresponding to the package determined in Figure 6. Note, this is only for the flow within the wafer panels and does not include the plenum losses. In addition, only an average condition was considered, so the fact that the flow increases as more H_2 is permeated into the sweep gas is not captured (this is considered in the FATHOM analysis presented below). As shown, if the number of stacks is at least 40, we have a pressure drop near 4 psid, which would allow for about 1 psid loss in the sweep/permeate headers.

The concept the sweep/permeate plenum basically includes is a rectangular area with separation plates to channel the flow through eight banks of wafers at a time through five passes before exiting the reactor. This is shown schematically in Figure 8 and the concept represented in Figure 9.

A model of the reactor sweep flow path was constructed using AFT-FATHOM, a commercially available pressure drop code. This model represents the flow using interconnecting pipes and junctions. This model incorporates the following features:

- H_2 mass source at each wafer stack sweep pass. This varies according to performance predictions;
- structural supports added to the plenum to support the pressure loading;

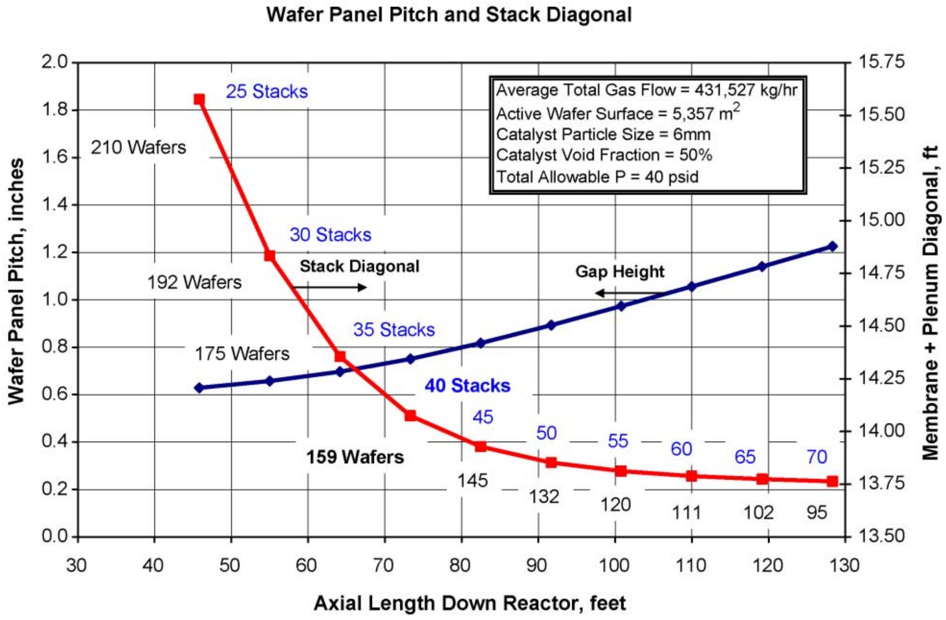

Figure 6: MWGS reactor wafer pitch versus reactor length.

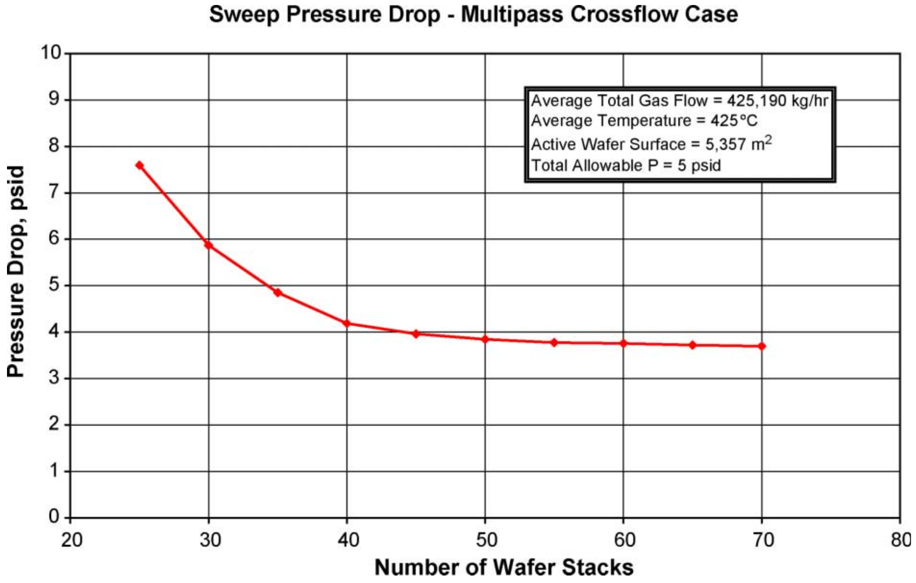

Figure 7: MWGS reactor sweep side pressure drop.

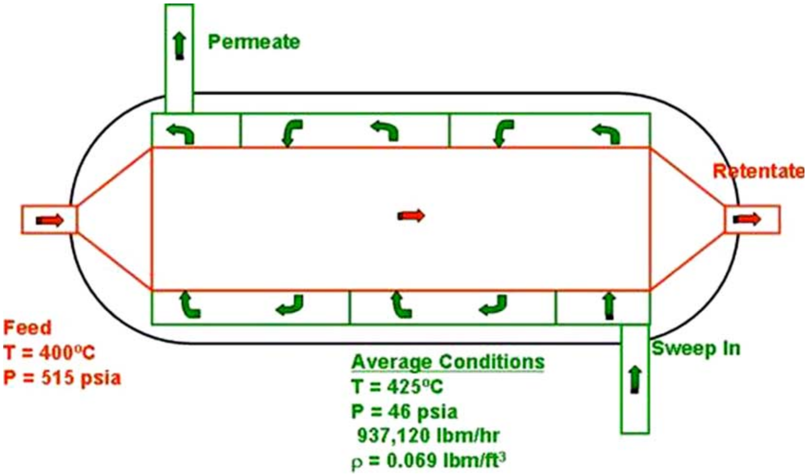

Figure 8: MWGS reactor sweep flow path schematic.

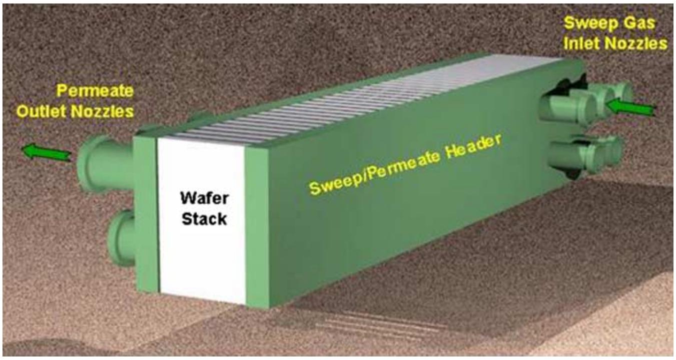

Figure 9: MWGS reactor sweep flow plenum concept.

- the wafer channels;
- inlet and outlet nozzles.

The total pressure drop predicted for this arrangement was found to be 5.75 psid, close to the target value. Further development of the flow plenums would need to be required in a future design effort to address the maldistribution and pressure drop.

Heat transfer design

The design basis for the insulation was to limit the heat loss to about 5% of the reaction energy released due to the water gas shift reaction. The reaction energy was calculated to be about 2.2 MW (7.6 million Btu/h). The allowable heat loss was therefore set at about 111 kW (380,000 Btu/h).

The insulation is to be placed externally on the vessel. This was chosen rather than to place it on the interior of the vessel because of:

- concern of convective heat loss through gaps in insulation at high pressure inside vessel;
- no mechanical strength advantage for the vessel material selected to keep it cool;

- no need to make special provision to insulate nozzle penetrations through the shell;
- low-cost materials.

Based on this heat transfer requirement, a thickness of 11.4 cm (4.5 in.) of Danser Vacuduct material was selected for estimating purposes. This is vacuum formed ceramic fiber insulation. The predicted heat loss for this material was ~100 kW (341,000 Btu/h). This amount of heat loss has minimal impact on the performance of the MWGS reactor.

Design performance
The design which was selected for detailed design and estimating has the following characteristics:

- 40 stacks of 159 membrane wafer panels, 2 m (6.55 ft) long by 3.05 m (10 ft) tall by 0.305 m (1 ft) wide;
- total active membrane surface area of 5357 m^2 (57,662 ft^2);
- a catalyst gap of 15.24 cm (6 in.) between each membrane stack.

The temperature and recovery profiles and the H_2 flux performance for the design case are shown in Figures 10 and 11.

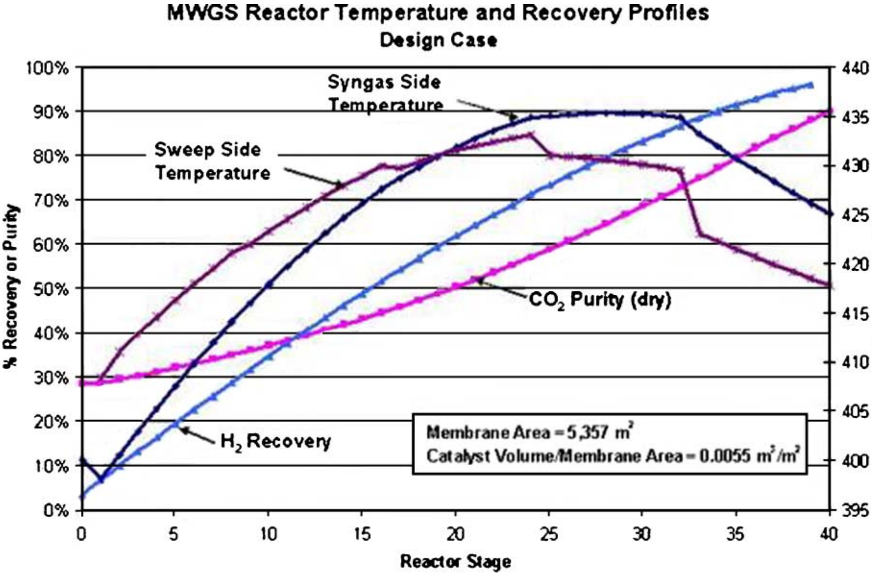

Figure 10: Design case temperature and recovery profiles.

Structural Analysis of Reactor Components
Structural analyses of the wafer stack assembly for pressure loading were accomplished to support the design effort. The resulting design satisfies the structural requirements except in a limited number of regions. The analysis results and recommendations are summarized below.

Wafer stack assembly repeat unit model. The overall stress distribution and deformed shape is shown in Figure 12. The predicted stress at key locations within the various components is shown in Table 7.

It is seen that the stress in the wafer stack end plate and the wafer stack spacer bars exceed the allowable limits. Further detailed elastic–plastic analyses of these regions should be accomplished to assess their acceptability. It is also seen that the wafer stack stay bar is far below the allowable stress. A reduction of stay bar width should be considered in future analyses to reduce system cost.

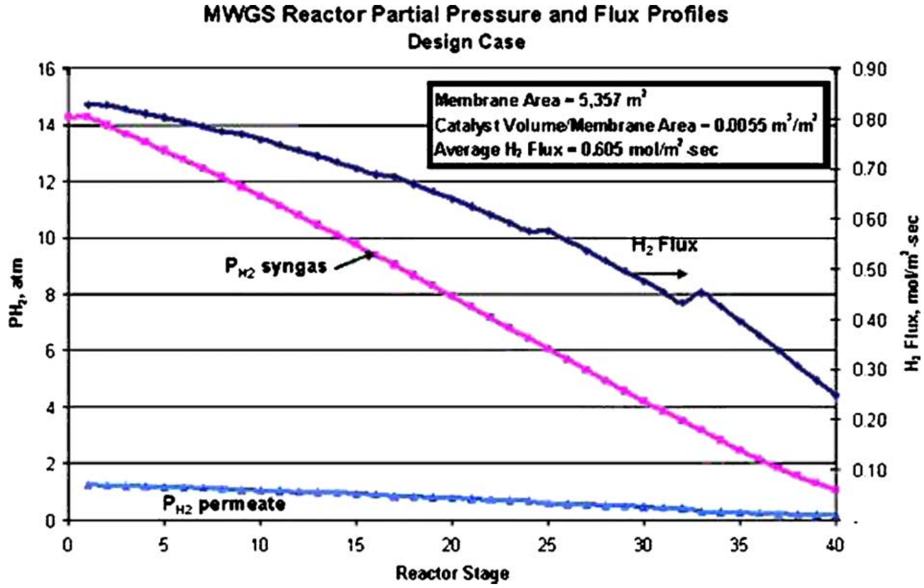

Figure 11: Design case partial pressure and H_2 flux profiles.

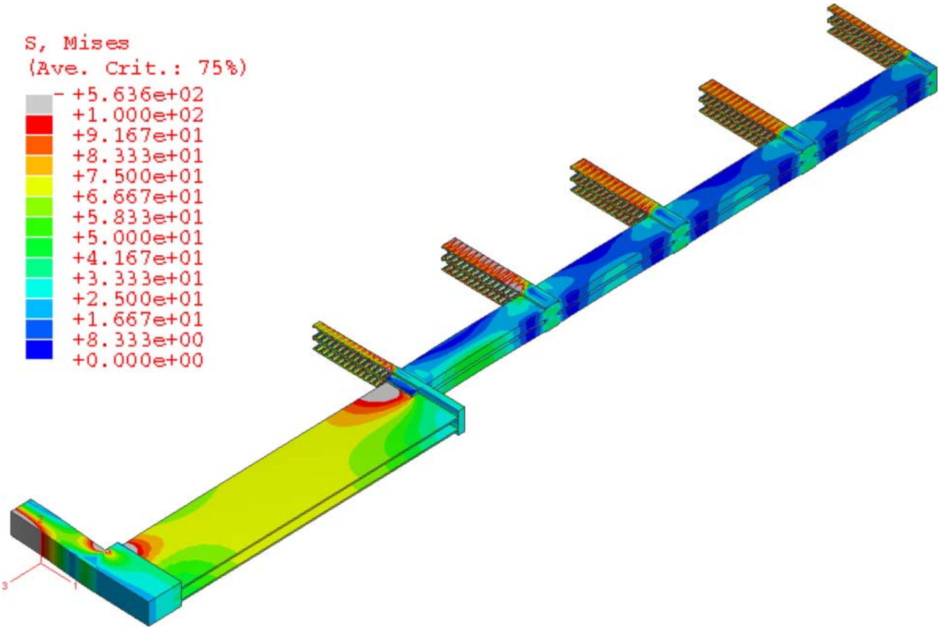

Figure 12: Wafer stack assembly stress distribution.

TABLE 7
PLENUM AND WAFER REPEAT UNIT MODEL RESULTS

Location	Fraction of allowable stress (%)	
	Membrane	Membrane + bending
Sweep gas plenum back plate	64	98
Sweep gas plenum horizontal stiffener		
Inboard end (AA)	79	77
Outboard end (BB)	78	92
Wafer stack end plate	119	157
Wafer stack stay bar	17	34
Wafer stack spacer bar		
Vertical section (AA)	112	139
Horizontal section (BB)	102	131

Reactor Vessel Design Concept

The reactor is a horizontally oriented steel pressure vessel resting on four saddle supports, as illustrated in Figure 13. Characteristics of the vessel are as follows:

- length is approximately 26.82 m (88 ft);
- inside diameter is 5.5 m (18 ft);
- welded construction;
- designed according to Section VIII, Division 1 of the ASME Boiler and Pressure Vessel Code;
- the vessel is designed for an internal pressure of 41.4 MPa (600 psig) at a vessel metal temperature of 454 °C (850 °F).

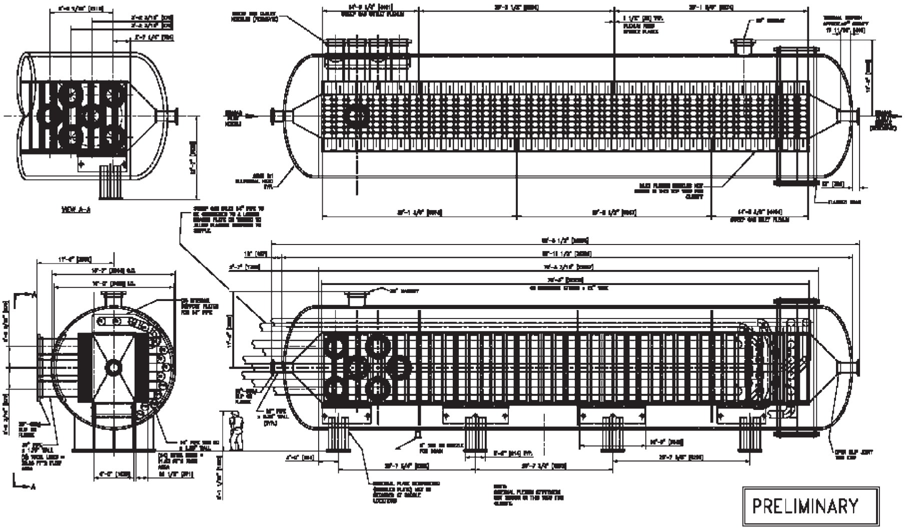

Figure 13: Baseline reactor vessel design.

The vessel houses 40 stacks of planar (corrugated) membrane panels. Each stack comprises 159 membrane panels, which are aligned and spaced vertically apart to permit syngas to flow over the outer surfaces of the membrane panels along the longitudinal axis of the vessel. The 40 stacks are arranged in line along the longitudinal axis of the vessel, and are each separated by a 15.2 cm (6 in.) thick bed of catalyst. Flow manifolds attached to the sides of the 40 stacks direct sweep gas through the inside of the membrane panels in a cross-flow direction, normal to the axial direction of syngas flowing over the panels. Divider plates inside the flow plenums are located such that sweep gas passes through the panels of a group of eight stacks at a time in five alternating cross-flow paths across the 40-stack membrane panel assembly.

Reactor Fabrication Cost Estimate

The estimated cost to fabricate the reactor vessel is approximately $19 million. The estimate is based on input from various suppliers of materials and services, as well as manufacturers specializing in the fabrication of components specified for the reactor. In part, the estimates developed by these vendors were based on detailed information provided to them, such as drawings or written processes and specifications. In many cases, where detailed information is not yet developed, rough cost estimates were provided by vendors based on similar work and standard cost models. As such, the $19 million estimate represents an order-of-magnitude cost to fabricate the reactor.

The reactor fabrication cost estimate is broken down into four major cost categories:

1. membrane panel stack assemblies—$4.3 million;
2. assembly of the reactor internals—$6.0 million;
3. reactor pressure vessel—$7.7 million;
4. catalyst beds and vessel external insulation—$1.0 million.

Tubular MWGS Reactor Concept

An alternative concept was developed in less detail after the completion of the planar vessel cost estimate. The alternate reactor concept is a tubular membrane vessel in which the high-pressure syngas is contained within thin membrane tubes, which are sized accordingly to meet the pressure requirements. The tubes are packaged in a pressure vessel in a U-tube arrangement which resembles standard shell-and-tube construction.

Tubular H_2 separation membrane design basis. The diameter of the tube is set by the material properties, the internal pressure, and the wall thickness. The design considered a 127 μm (0.005 in.) wall thickness, with the membrane material properties presented in Table 5. This resulted in a tube inside diameter of 10.77 mm (0.424 in.). Note that later information from Eltron suggested that a membrane thickness of 250 μm (0.0098 in.) may be feasible. Such a wall thickness would permit tubes up to 21.18 mm (0.834 in.) in diameter.

To facilitate catalyst placement, it was decided to separate the process into four separate reactors with catalyst reactors staged in between. This avoided potential tube damage due to catalyst placement around the tubes as a result of bed lock up. The process analysis also assumed that the sweep gas was partitioned into four separate paths as shown in Figure 14. The change in the sweep flow arrangement requires that the membrane surface area be increased to 5685 m^2 (61,193 ft^2).

The pressure drop on the feed/syngas side needed to account for loss through tubes, catalyst, and piping. For design purposes, the catalyst was arbitrarily assigned 10 psid, and the membrane tubing and interconnecting piping (100 ft of 16 in. pipe) was assigned 30 psid. Based on the 10.77 mm (0.424 in.) inside diameter tubes and the required total surface area of 5685 m^2 (61,193 ft^2) the arrangement calls for each of 4 vessels to have 9730 U-tubes, each with 13.8 ft long active membrane.

The arrangement of the tubes on the sweep/permeate side is based on the need to maintain a 5 psid pressure drop. The design was based on the sweep flow making two passes in each vessel over a tube bundle with a 6.35 mm (0.25 in.) minimum tube spacing.

The tubular reactor concept, therefore, has the following characteristics:

- Four separate membrane reactors interstaged with catalyst reactors.
- Each membrane reactor has 9730 U-tubes, 1.07 cm (0.424 in.) ID, 4.2 m (13.8 ft) long.

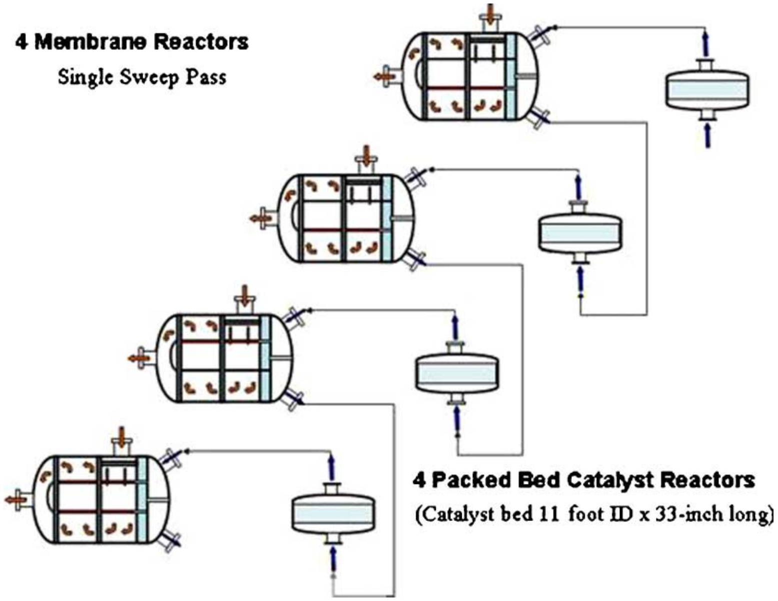

Figure 14: Staged tubular membrane vessel arrangement.

- Total active membrane surface area of 5685 m^2 (61,193 ft^2).
- Each membrane reactor is about 7.6 m (25 ft) long and 3.2 m (10.5 ft) diameter.

This design is fashioned after a standard U-tube type shell and tube heat exchanger, in which the membranes are of a tubular form and are an integral part of U-tube assemblies. The ends of the U-tube assemblies are joined to a single tubesheet with syngas flowing inside the tubes (high pressure tube-side of the reactor). Sweep gas flows across the outside of the membrane tubes in the lower pressure shell side of the reactor.

Figure 15 shows the assembly of the tubular reactor. The syngas feed enters the flanged, removable head of the reactor, flows through 9730 U-tubes containing straight active lengths of membrane tubes, and exits as retentate through an outlet nozzle in the removable head. The syngas feed and retentate flows are separated by means of a partition plate in the high-pressure reactor head. The tubesheet is gasketed between the head and shell flanges and is, therefore, removable to facilitate installation of the U-tube assemblies. Each of the 9730 U-tube assemblies includes two straight 2.13 m (84 in.) long sections of “active length” membrane tube. One end of each membrane tube is brazed to a stainless steel U-bend tube section, and the opposite ends are brazed to straight, 1.12 m (44 in.) long transition sections of stainless steel tube. The straight transition tube sections pass through the tubesheet and are expanded into the tubesheet holes to form the tube-to-tubesheet joints. Baffle plates and support plates positioned within the U-tube bundle support the tubes and direct the sweep gas flow across the outer surfaces of the straight membrane tubes, as indicated by the flow arrows in the reactor assembly drawing. The baffle and support plates are held in position by tie bars mounted to the tubesheet.

Four membrane reactors are required to achieve the hydrogen separation capacities of the program. However, unlike the baseline concept in which syngas catalyst beds are an integral part of the reactor, the U-tube membrane reactor concept does not include a provision for containing catalyst. As such, the catalyst beds are contained in four separate catalyst reactor vessels external to the membrane reactors. These catalyst reactor vessels are interstaged with the membrane reactor vessels as shown in the process flow schematic on the assembly drawing in Figure 15. The vessel is supported on a cylindrical skirt and is sized to accommodate and 3.35 m (11 ft) diameter, 84 cm (33 in.) thick catalyst bed within the shell portion

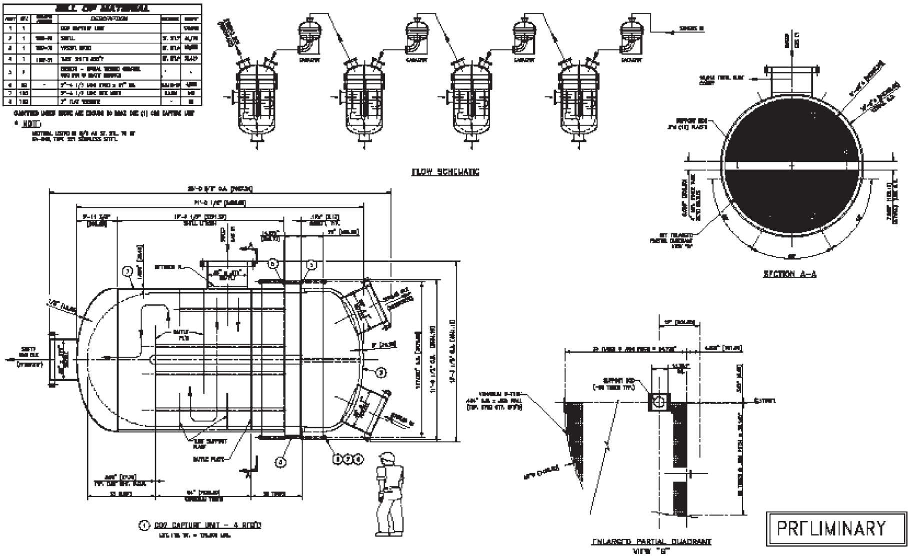

Figure 15: Tubular membrane reactor assembly.

of the vessel. Syngas enters the vessel from the top inlet nozzle, flows through the bed and exits the vessel through the lower head and elbow.

The material selected for construction of the catalyst reactor vessels, as well as the membrane reactor vessel shells, tubesheets and removable heads is Type 321 austenitic stainless steel. This material selection is based on its high strength, elevated temperature rating, and corrosion resistance characteristics. This material is also compatible with the process gas compositions and environments to which it will be exposed during reactor operation.

The estimated cost to fabricate and assemble four single U-tube membrane reactors and four catalyst reactors required for the tubular membrane plant concept is approximately $12 million. This estimate does not include the cost for interconnecting piping between the eight vessels, nor does it include fabrication development functions, such as for membrane tube forming, brazing and other joining processes, tooling, building prototypes and assembly trials. The $12 million cost estimate comprises the following major cost categories:

1. membrane tubes—$2.26 million;
2. membrane U-tube assemblies—$2.2 million;
3. membrane reactor vessels—$4.8 million;
4. catalyst reactor vessels—$2.3 million;
5. external insulation for eight vessels—$0.41 million.

The above summaries provide a breakdown of the costs estimated to produce the tubular membrane reactor concept. This estimate does not include costs for required fabrication development functions or for shipping completed reactor vessels of these sizes from a manufacturer to an end-use site.

CONCLUSIONS

Two feasible MWGS reactor designs have been developed, which use either a planar or a tubular hydrogen separation membrane. The planar reactor design uses a membrane composed of a curved membrane supported by a corrugated Type 430 stainless steel sheet. Finite element analysis which considered the pressure, and differential thermal expansion loadings indicates that it is structurally adequate for 41.4 bar (600 psid) pressure loading at 450 °C (842 °F). A second MWGS reactor concept is based on a tubular membrane sized appropriately to contain high pressure inside the tubes.

A performance analysis tool was developed to permit examination of different arrangements for the MWGS reactor and bench-marked against the model developed in Phase I. This analysis tool determined the membrane area required for the planar and tubular reactor concepts.

The baseline planar design places the membrane internals inside of a conventional pressure vessel. An alternative planar design uses an externally stayed structure to house the membrane panels. The planar membrane reactors have the following characteristics:

- a multi-pass cross-flow arrangement to meet the performance and pressure drop requirements;
- catalyst placement in a catalyst gap of 0.15 m (6 in.) between each membrane stack;
- forty stacks of 159 membrane wafer panels, 2 m (6.55 ft) long by 3.05 m (10 ft) tall by 0.305 m (1 ft) wide;
- total active membrane surface area of 5357 m^2 (57,662 ft^2);
- length is approximately 26.8 m (88 ft).

Detailed stack design structural analyses of the planar wafer stack assembly for pressure loading were accomplished to support the design effort. The resulting design satisfies the structural requirements except in a limited number of regions.

The tubular membrane reactor concept has the high-pressure feed gas inside the tubes and the sweep gas flowing across the tube bank. The tubular membrane reactor concept, which was not designed as rigorously as the planar options, was based on standard shell and tube construction. The tube length was set to meet the feed-side pressure drop constraint for a given tube diameter, and the tube pitch and baffle arrangement were set to meet the sweep-side pressure drop constraint. The characteristics of the tubular arrangement include:

- four separate membrane reactors interstaged with catalyst reactors;
- each membrane reactor has 9730 U-tubes, 1.07 cm (0.424 in.) ID, 4.2 m (13.8 ft) long;
- total active membrane surface area of 5685 m^2 (61,193 ft^2);
- each membrane reactor is about 7.6 m (25 ft) long and 3.2 m (10.5 ft) diameter.

The estimated order-of-magnitude cost to fabricate the reactor vessel is approximately \$19 million. The estimate is based on input from suppliers of materials and services, as well as manufacturers specializing in the fabrication of components specified for the reactor. In many cases, where detailed information is not yet developed, rough cost estimates were provided by vendors based on similar work and standard cost models.

The tubular membrane MWGS reactor was estimated with three sub-options in which vessels were combined. The alternate arrangement cost estimates are summarized in Table 8.

TABLE 8
MWGS REACTOR ESTIMATE SUMMARY

Vessel option	**Estimated cost**
Baseline planar concept, separate internals and pressure vessel	$19,054,600
Shell and tube vessel with tubular membranes	
4 Membrane vessels + 4 catalyst vessels	$11,810,400
2 Membrane + 4 catalyst	$11,690,400
2 Membrane + 1 catalyst	$11,190,400

RECOMMENDATIONS

The feasibility and cost to manufacture either the planar or tubular membranes as conceived in this study should be investigated. In particular, the cost of the operations as well as the assembly procedures must be proven out to have more confidence in the cost to produce the reactor.

The following additional investigations are recommended for the membrane design:

- Re-evaluation of the membrane stress (or allowable tube diameter) when mechanical properties of the membrane material alloy are available and final membrane thickness is selected.
- Acceptability of Type 430 stainless steel considering operating environment and interaction with membrane material.
- Vibration testing of the wafer panel assembly or tube bundle.
- Mass transfer limitations on a given membrane arrangement.

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