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COMBUSTION-ASSISTED CO₂ CAPTURE USING MECC MEMBRANES

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ABSTRACT

Mixed Electron and Carbonate ion Conductor (MECC) membranes have been proposed as a means to separate CO₂ from power plant flue gas. Here a modified MECC CO₂ capture process is analyzed that supplements retentate pressurization and permeate evacuation as a means to create a CO₂ driving force with a process assisted by the catalytic combustion of syngas on the permeate side of the membrane. The combustion reactions consume transported oxygen, making it unavailable for the backwards transport reaction. With this change, the MECC capture system becomes exothermic, and steam for electricity production may be generated from the waste heat. Greater than 90% of the CO₂ in the flue gas may be captured, and a compressed CO₂ product stream is produced. A fossil-fueled power plant using this process would consume 14% more fuel per unit electricity produced than a power plant with no CO₂ capture system, and has the potential to meet U.S. DOE's goal that deployment of a CO₂ capture system at a fossil-fueled power plant should not increase the cost of electricity from the combined facility by more than 30%.

1.0 INTRODUCTION

Mixed Electron and Carbonate ion Conductor (MECC) membranes [1] have been proposed as a means to perform post-combustion CO₂ capture [2] from fossil-fueled power plant flue gas. A MECC membrane is composed of a porous metal support infiltrated with a carbonate salt mixture (i.e., lithium carbonate (Li₂CO₃), potassium carbonate (K₂CO₃), sodium carbonate (Na₂CO₃)). The metal support is conductive to the flow of electrons, and the carbonate salt, when molten, is conductive to the flow of carbonate ions. The transport of carbon dioxide (CO₂) across the membrane requires oxygen (O₂) at the retentate side and the transport driving force depends on the difference in CO₂ and O₂ partial pressures across the membrane. Fig. 1 shows a schematic of an MECC cell and the half-cell reactions that occur on each side of the membrane.

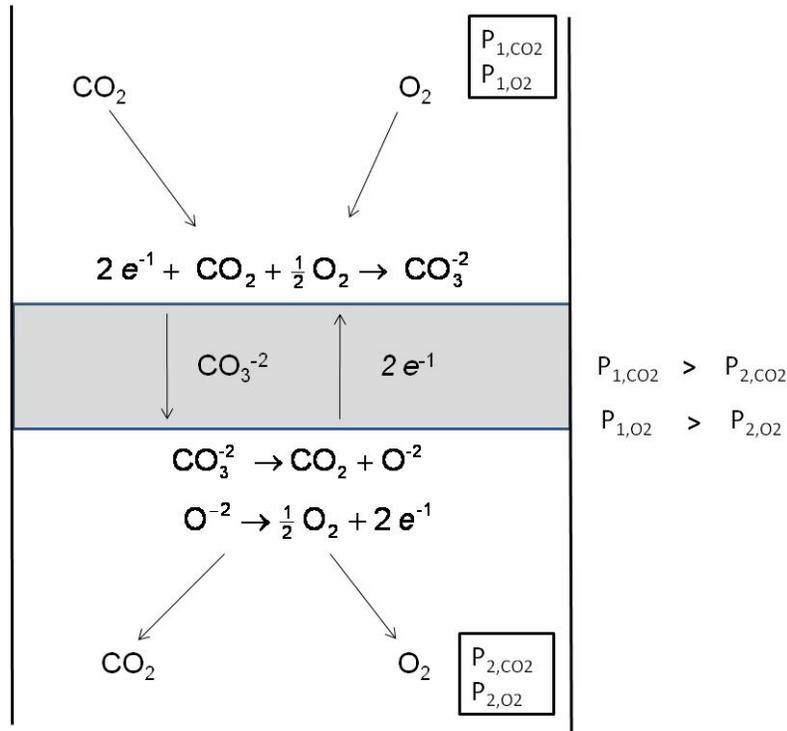


Fig. 1. MECC Membrane CO₂ Transport Half-Cell Reactions.

Because the pores of the metal support structure are filled with molten carbonate, membranes of this type tend to have high CO₂+O₂ selectivity and very low permeability to substances that aren't soluble in the carbonate. High CO₂ selectivity, in addition to high permeance, has been identified as a prerequisite for a successful membrane-based CO₂ separation system [3].

According to Chung et al. [1], CO₂ permeance is determined by the following expression.

$$F_{\text{CO}_2} = \frac{9RT}{16F^2L} \sigma_{\text{ion}} \ln \left(\frac{P_t^i}{P_t^{ii}} \right) \frac{\epsilon}{\tau} \left[\frac{1}{(P_t^i - P_t^{ii})} \right] \quad (1)$$

In Equation (1), F_{CO_2} is the CO₂ permeance, R is the ideal gas constant, T is the temperature in Kelvin, F is Faraday's constant, L is the thickness of the membrane, σ_{ion} is the membrane ionic conductance, P_t^i and P_t^{ii} are the CO₂ + O₂ total pressure on the

retentante and permeate sides of the membrane, ϵ is the porosity of the membrane, and τ is the membrane tortuosity. The electrical conductance of the membrane, signified by σ_{elec} , is usually much greater than the ionic conductance, σ_{ion} , under normal operating conditions, and therefore F_{CO_2} is not assumed to be a function of σ_{elec} in Equation 1. Permeance increases when the ionic conductance of CO_2 across the membrane increases, the pressure difference of CO_2+O_2 across the membrane increases, when the membrane porosity increases, and when the membrane tortuosity decreases. In this case, membrane porosity and tortuosity refer to the characteristics of the metal support structure since the molten carbonate component is liquid at operating temperature.

The permeance of MECC membranes at atmospheric pressure and over a range of temperatures has been measured. Fig. 2 shows a plot of permeance data from Chung et al. [1] and permeance values that were calculated from CO_2-O_2 flux data ($ml/min\ cm^2$) from Xu et al. [4] for sintered stainless steel membranes having the same thickness ($L=1.57\ mm$), porosity ($\epsilon = 0.25$), tortuosity ($\tau = 5.94$), and prepared using similar Li, Na, and K carbonate mixtures. In [1], a pressure driving force of 1 atm across the membrane was created by drawing a vacuum on the permeate side of the membrane, while in [4], a helium sweep gas at 1 atm was used to remove the permeate and create a partial pressure driving force of 1 atm across the membrane. In [1], a Li/Na/K carbonate mixture was used to prepare the membrane, while in [4], two membranes were tested, one membrane infiltrated with a Li/Na carbonate salt, and another membrane infiltrated with a Li/K carbonate salt.

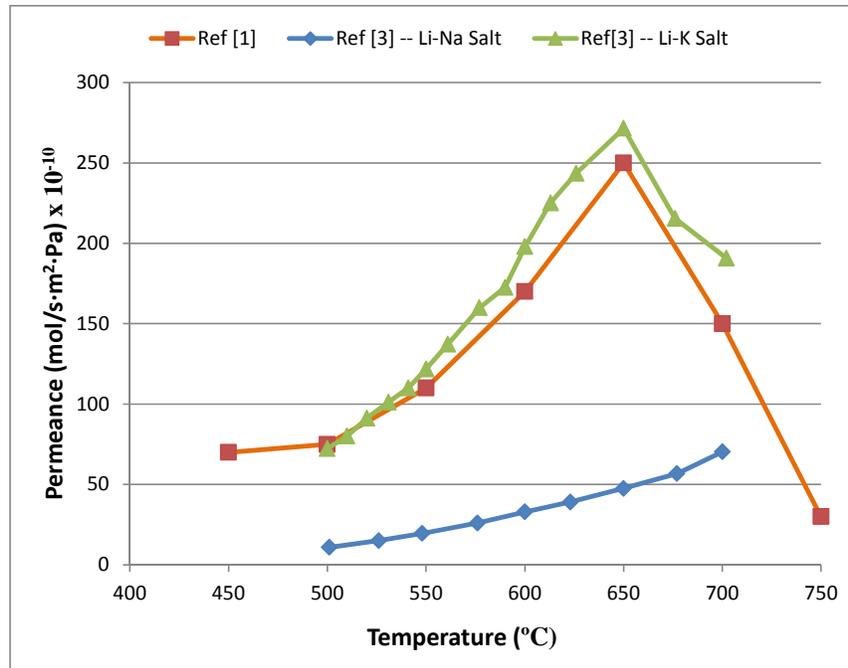


Fig. 2. MECC membrane permeance versus temperature

In Fig. 2, the permeance measurements from [1] rise to a maximum of 250×10^{-10} mol/s·m²·Pa at 650°C and then falls at higher temperatures. In [4], the measured permeance varies with temperature, and with the type of salt used to prepare the membrane. When a Li/Na carbonate salt was used to prepare the membrane, the permeance increased monotonically with temperature, and was lower in value at all temperatures than that measured in [1]. When a Li/K carbonate salt was used to prepare the membrane, the permeance behaved in a similar manner as measured in [1], reaching a maximum value of about 270×10^{-10} mol/ s·m²·Pa at 650°C. In [1], the drop in membrane performance above 650°C was attributed to the reaction of Li in the molten carbonate with Fe in the metal membrane support, leading to the formation of LiFeO₂. LiFeO₂ is electrically non-conductive, and formation of this substance in the membrane would increase the electrical resistance of the membrane and decrease

membrane permeance. Under these circumstances, σ_{elec} becomes an important factor in limiting CO_2 transport, and Equation 1 must be changed to include σ_{elec} .

MECC membranes are similar in construction to carbonate-ceramic dual-phase membranes as described by Anderson and Lin [5], except that the metal support structure found in MECC membranes is replaced by a mixed conducting oxide ceramic. Replacement of the support structure by a mixed oxide conducting ceramic allows CO_2 to permeate the membrane even when oxygen is absent in the retentate, but at a lower rate than when oxygen is available in the retentate [6].

For MECC-based membranes, stoichiometric amounts of O_2 must be present in the flue gas to facilitate CO_2 transport, and additional air or O_2 may need to be mixed with the flue gas to remove the desired amount of CO_2 . A partial pressure difference must be established across the membrane, which usually requires deployment of energy-intensive gas compression on the retentate side of the membrane, and gas evacuation on the permeate side of the membrane. The permeate is a mixture of CO_2 and O_2 , and further gas separation steps are needed to produce a pure CO_2 stream. Since the MECC membranes operate at higher temperatures ($T > 450^\circ\text{C}$), there are also safety concerns in handling high-temperature oxygen-enriched permeates.

A modified MECC process may be designed to mitigate some of these concerns. A reactive gas such as syngas (mixture of hydrogen (H_2) and carbon monoxide (CO)) or H_2 may be used as a sweep gas on the permeate side of the membrane. The sweep gas, aided by a combustion catalyst, reacts with O_2 in the permeate to make CO_2 and water vapor (H_2O), which reduces the O_2 partial pressure in the permeate. Additional CO_2 is produced by the combustion of CO , but the CO_2 produced by combustion of CO

is on the “correct” side of the membrane to achieve capture. The heat released by the combustion reactions may be used to maintain membrane temperature, preheat feed streams, and to make steam for additional electricity production.

In this work, the mole balances of several combustion-assisted MECC CO₂ capture processes are compared in order to determine whether natural gas or coal are suitable feed stocks. The implications of selecting syngas or H₂ as the reactive gas are explored. Then, a detailed process flow sheet, along with its mass and energy balances, is developed that uses a syngas sweep stream produced from methane. Process energy costs are determined from the detailed process flow sheet and compared to a base case power plant with no CO₂ capture. Some information on the cost of the membranes is also determined.

2.0 PROCESS MOLE BALANCES

Syngas, which is a mixture of CO and H₂, or H₂ produced from syngas, are initially proposed as reactive gas inputs for a combustion-assisted MECC CO₂ capture process. Nearly all syngas streams produced industrially [7] are made from the steam reforming of coal (51%), petroleum (26%), or natural gas (21%), and about 95% of H₂ produced industrially in the United States is purified from syngas produced from natural gas [8]. The selection of the reactive gas (syngas or H₂) and the reactive gas feedstock (coal, petroleum, natural gas) affects operational costs and the amount of CO₂ generated by the CO₂ capture process. To minimize operational costs, the feedstock must be inexpensive, and the syngas H₂:CO ratio must be maximized to reduce the amount of CO₂ generated by the CO₂ capture process itself.

Four cases are considered that span the range of potential feedstock and reactive gas types. The feedstock types and assumptions for each case are shown in Table 1.

Table 1. Case Descriptions

	Case #1	Case #2	Case #3	Case #4
Feed Type	Methane	Methane	Coal	Coal
Feed Formula $C_xH_yO_z$, (x,y,z)	(1,4,0)	(1,4,0)	(1,0.7,0.1)	(1,0.7,0.1)
Water-Gas Shift Unit Deployed?	No	Yes	No	Yes
Reformer Thermal Efficiency	70%	70%	70%	70%
Molar Conversion of Feedstock in the Reformer	100%	100%	100%	100%
Water-Gas Shift Molar Conversion	---	75%	---	75%
H ₂ Purification Efficiency	---	89%	---	89%
Moles CO ₂ captured per mole feed	2.1	1.4	0.45	~0.3

In Case #1, methane (CH₄), is used as a feedstock, and syngas is used as the reactive gas in the MECC CO₂ capture module, as shown in Fig. 3.

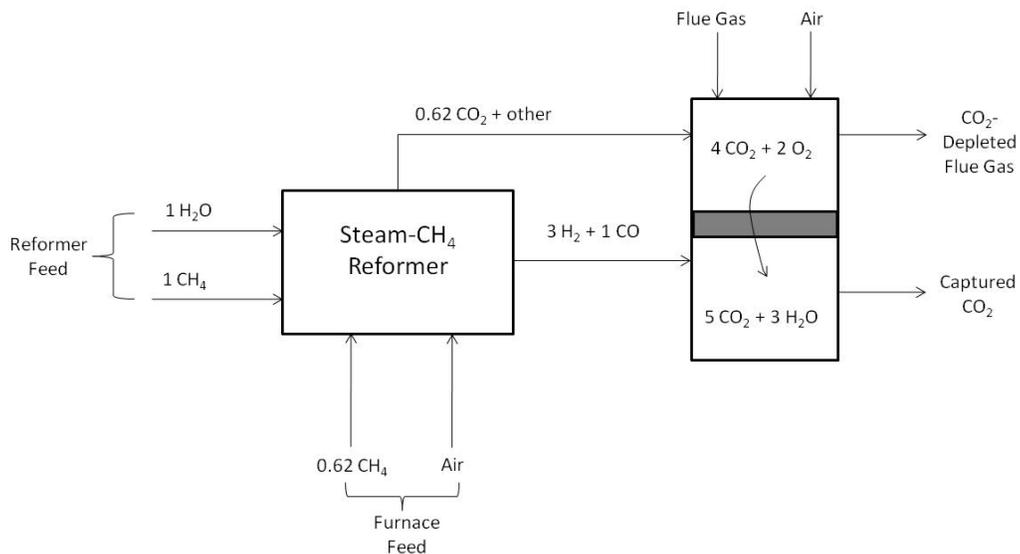


Fig. 3. Block flow diagram of Case #1.

In the figure, CH₄ and water (H₂O) are fed to a steam-methane reformer, where it is catalytically transformed into syngas having a 3:1 H₂:CO ratio [9]. The steam-methane

reformer is heated using a methane-fired furnace. The reformer exhaust is mixed with the power plant flue gas so that CO_2 in the reformer exhaust may be captured. Full conversion of CH_4 into syngas results in the creation of 3 moles H_2 and 1 mole CO for every mole of CH_4 feed. According to the process mole balance, up to 3.4 moles of CO_2 in the power plant flue gas may be captured for every 1.6 moles of CH_4 consumed by the CO_2 capture process, or about 2.1 moles CO_2 in the power plant flue gas for every mole of CH_4 consumed.

In Case #2, the syngas stream produced by the steam-methane reformer is further transformed by the Water-Gas Shift reaction [10] to make H_2 . The water-gas shifted syngas stream is then purified, and pure H_2 is used as the reactive gas in the MECC CO_2 capture module, and the impure gas stream from the H_2 purification unit containing CO , H_2 , and CO_2 is recycled back to the steam reformer furnace to be burned. A diagram of Case #2 is shown in Fig. 4. According to the process mole balance, about 2.4 moles of CO_2 from the power plant flue gas are captured for every 1.4 moles of CH_4 consumed, or about 1.7 moles of CO_2 are captured from the power plant flue gas for every mole of CH_4 consumed.

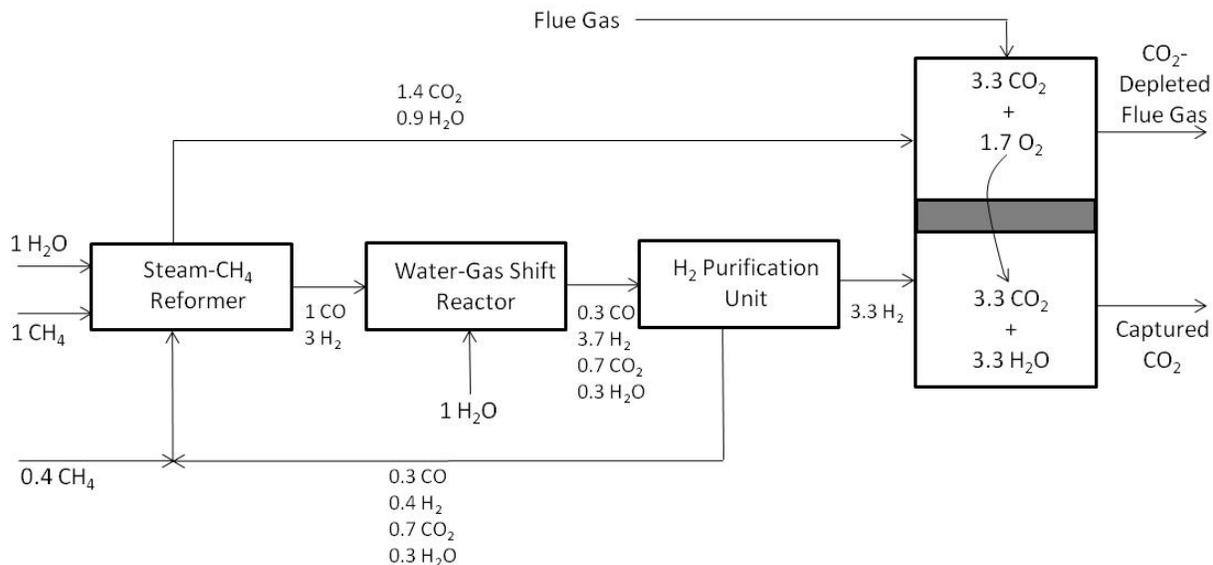


Fig. 4. Block flow diagram of Case #2.

In Case #3, coal is substituted for methane in Case #1. If it is assumed that coal is represented by the hypothetical compound CH_{0.7}O_{0.1}, which corresponds to the approximate molar composition of bituminous coal minus moisture and other impurities [11], then only about 1.0 moles of CO₂ from power plant flue gas may be captured for every 2.2 moles of coal consumed by the steam reformer (1.0 moles coal for the steam reforming reaction, 1.2 moles coal for the reformer furnace), or about 0.45 moles of CO₂ captured for every mole of coal consumed by the reformer.

Case #4 is similar to Case #3 except that the syngas stream produced by the steam reforming of coal is further converted into a pure hydrogen stream using a Water-Gas Shift unit, and the results are even worse. Only about 0.3 moles of CO₂ may be captured for every mole of coal consumed.

Among the cases considered, using CH₄ as a process feedstock and syngas having a 3:1 H₂:CO ratio as the reactive gas appears to be the most desirable option. Up to 2.1 moles of CO₂ in power plant flue gas may be captured for every mole of CH₄

consumed, which is higher than the other cases. The use of a purified H₂ stream produced from methane might be considered as a back-up option, however, if the increased feedstock costs might be offset by potential operational advantages associated with combusting only H₂ in the MECC CO₂ capture units (e.g., lower CO₂ partial pressure in the permeate). Using coal as a feedstock for this process is not at all feasible because the CO₂ capture plant would consume more fuel and generate more CO₂ than the original power plant.

3.0 DETAILED PROCESS MODELING

Even in the best case, the feedstock requirements for the CO₂ capture process are high. For a natural gas-powered electrical plant of fixed size, for example, the amount of natural gas needed to operate the CO₂ capture process on a molar basis would increase the amount of fuel supplied to the combined facility by about 52%, with the additional amount of natural gas used to make syngas to drive the CO₂ capture process. That is, for every mole of methane burned at the electrical power plant, about 0.52 moles of methane are needed to drive the CO₂ capture process if 100% of the CO₂ generated by the overall plant is to be captured. Alternatively, if the natural gas feed molar rate were fixed, then the amount of power generated by the power plant would have to be reduced by 32% because a portion of the natural gas provided to the power plant complex would need to be diverted to the steam-methane reformer to make syngas instead of burning it to make steam for electricity production. At these feedstock consumption rates, the parasitic load of the CO₂ capture plant exceeds the 20% target

established by the U.S. DOE for CO₂ capture processes [11,12], and the process would not be acceptable.

The utility of the proposed CO₂ capture system may be greatly improved, however, if the CO₂ capture system also serves as a supplemental power plant. The combustion of syngas in the MECC CO₂ capture units releases copious quantities of heat, and the heat may be used to make steam for electricity production. In this manner, some of the thermal value of methane fed to the CO₂ capture plant may be recovered as electrical power, and the fuel penalty associated with operating the CO₂ capture plant may be lessened.

Calculating how much supplemental power the CO₂ capture plant might generate requires a more detailed process model than previously discussed. Mass and energy balances are needed, and integration of thermal flows will be required to determine how much energy is needed to operate the CO₂ capture process, and how much excess thermal energy is available for steam production. In building a process model, physical and chemical constraints must also be recognized, so that the modeled conditions are not overly optimistic compared to what might be possible in an actual process.

3.1 Process Assumptions and Constraints

3.1.1 Process Chemistry

The following chemical compositions, conditions, and constraints are used in the detailed process model.

- A simplified list of chemical species is used – Ar, O₂, CH₄, CO, CO₂, H₂, H₂O, and N₂.

- The power plant flue gas is assumed to contain [13]: 13% CO₂, 16% H₂O, 3% O₂, and 68% N₂; and is delivered to the CO₂ capture plant at 150°C at 1 atm.
- The CO₂ capture plant uses CH₄ as a power source and a syngas feedstock, regardless of the fuel used to generate electricity in the electrical power plant (coal, oil, or natural gas).
- The following chemical reactions are assumed. Equations 1-3 are the H₂, CO, and CH₄ combustion reactions. Equation 4 is the steam reforming reaction of methane. All are assumed to go to completion.



- Heated air is used as an O₂ source for the MECC CO₂ capture units, and is mixed at a flow rate sufficient to facilitate 100% transport of CO₂ in the power plant flue gas and the steam reformer furnace exhaust.
- Combustion of H₂ and CO in the MECC CO₂ capture modules is catalyzed, and requires at least 2 mol% excess O₂ to proceed.
- The steam-methane reformer is assumed to operate at ~20.5 atm and 850°C [14,15].

3.1.2 *Physical Constraints and Requirements*

The following physical constraints and requirements are assumed:

- The membranes are assumed to be 100% selective for CO₂ and O₂, and are impermeable to N₂ and other gases.
- The CO₂ capture process is sized to remove 90% of the CO₂ in the combined flue gas stream (i.e., power plant flue gas + reformer furnace exhaust gas + make-up air). 100% removal is not performed because a CO₂ + O₂ partial pressure driving force must be maintained across the separation membranes in order to achieve separation.
- The fuel requirements and power output of the electrical power plant are fixed, and the CO₂ capture plant is assumed to be an add-on to an existing facility. Therefore, the CO₂ capture plant requires a dedicated fuel feed stream separate from the power plant fuel feed stream.
- The entry temperature of gases into the MECC membrane units is 500°C on both sides of the membrane to facilitate effective CO₂ transport, and the maximum allowable exit temperature of gases from the MECC membrane units is 700°C to avoid changes in the chemical and physical properties of the membranes [1]. Although membrane degradation in [1,4] occurred above 650°C, it is assumed for this analysis that good membrane performance could be extended to 700°C (or beyond) without large changes to membrane materials and construction.
- There is 10% loss of thermal energy to the environment from the steam methane reformer. All other thermal energy is available for driving the steam reforming reaction (Equation 4) and for preheating feed streams.
- The MECC CO₂ capture units are well insulated and operate adiabatically.

- Thermal energy released by the CO and H₂ combustion reactions is shared between the retentate and permeate streams in the MECC CO₂ capture units, but the permeate exit temperature is assumed to be 10°C warmer than the retentate exit temperature due to heat transfer limitations.
- Excess heat not used to preheat unit feed streams is used to make high-pressure superheated steam (~20.5 atm, 426°C) or lower pressure superheated steam (~5 atm, 155°C) for electrical power production.
- No pressure drop through the MECC CO₂ capture units is assumed, and the cells are assumed to operate at atmospheric pressure on both sides of the MECC membrane.
- The high-pressure and lower-pressure steam turbines have a 0.8 isentropic efficiency and an exit pressure of 1 atm.
- All process compressors have an isentropic efficiency of 0.8 and a thermal efficiency of 1.0.
- All process pumps have a volumetric efficiency of 0.8 and a mechanical efficiency of 1.0.

3.2 Process Simulation

Process flow sheets were developed using the process simulator ProSimPlus version 3.1.2.1.2. Initially, the basic process steps were established in the simulator, and then the feed and product streams were linked by heat exchangers, as needed, to preheat unit feed streams. The flow sheet configuration was then adjusted to ensure that the MECC CO₂ capture modules did not overheat as a result of the CO and H₂

combustion reactions. Simplified block flow diagrams of the process sections were drawn using SmartSketch 2007 version 05.02.03.09 Service Pack 3 by Intergraph Corporation.

4.0 RESULTS AND DISCUSSION

A high-level block flow diagram of the CO₂ capture process is shown in Fig 5.

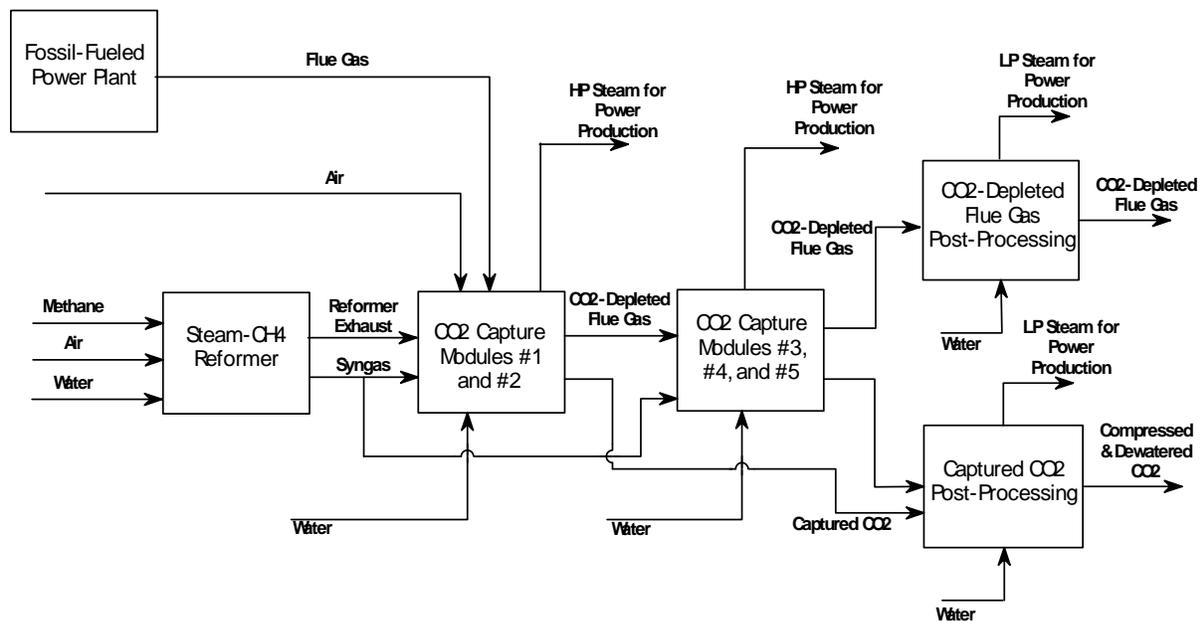


Fig. 5. Block flow diagram of proposed combined power plant/CO₂ capture facility.

In the diagram, the significant process sections are the Fossil-Fueled Power Plant; the Steam-CH₄ Reformer; CO₂ Capture Modules #1 and #2; CO₂ Capture Modules #3, #4, and #5; CO₂-Depleted Flue Gas Post-Processing; and Captured CO₂ Post-Processing. The individual unit operations are grouped into process sections according to function and thermal flows. The process sections are described below.

4.1 Steam-CH₄ Reformer Process Section

The Steam-CH₄ Reformer process section consists of a direct-fired steam-CH₄ reformer unit and heat exchangers that are used to recuperate heat from the furnace exhaust and syngas product streams. Fig. 6 shows a detailed block flow diagram of the Steam-CH₄ Reformer process section.

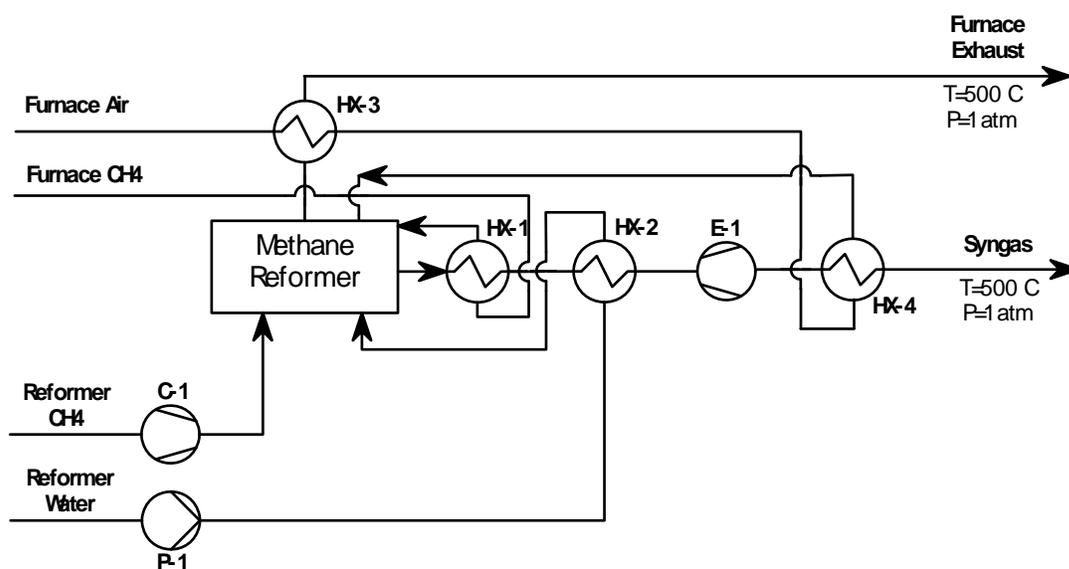


Fig. 6. Detailed block flow diagram of "Steam-CH₄ Reformer" process section.

The heat exchangers are arranged to preheat reformer feed streams to the greatest extent possible, and to cool the syngas and furnace exhaust streams to 500°C, the entry temperature of the MECC CO₂ capture units. Heat exchangers HX-1 and HX-2 cool syngas from 825°C to 304°C, preheat CH₄ for the reformer furnace from 25°C to 300°C, and preheat water at 25°C and 20.4 atm to make superheated steam at 400°C and 20.4 atm. Heat exchanger HX-3 cools the reformer furnace exhaust from 950°C to 500°C and

preheats the furnace air stream from 25°C to 607°C. The preheated reformer furnace air stream warms the syngas stream to 500°C in heat exchanger HX-4, while the preheated air stream is cooled from 607°C to 363°C before it enters into the Steam-CH₄ Reformer furnace. Compressor C-1 and pump P-1 pressurize the reformer CH₄ and water streams to 20.4 atm, and expander E-1 reduces the syngas pressure from 20.4 atm to 1 atm while recovering some of the mechanical energy to make electricity. The mechanical action of compressor C-1 preheats the CH₄ reformer stream from 25°C to 404°C.

A stream table for the Steam-CH₄ Reformer Section is shown in Table 2.

Table 2. Partial Stream Table, Reformer Section

	Steam-CH ₄ Reformer, Reforming Section		Steam-CH ₄ Reformer, Furnace Section		Output, Syngas mol/h	Output, Furnace Exhaust mol/h
	Input, CH ₄ Stream mol/h	Input, H ₂ O Stream mol/h	Input, CH ₄ Stream mol/h	Input, Air mol/h		
Ar				5.3		5.3
O ₂				119		10.8
CH ₄	90		54			
CO					90	
CO ₂	1.4		0.9	0.2	1.4	55.1
H ₂					270	
H ₂ O		90		10.2		118
N ₂	1.4		0.9	438	1.4	438

In the simulation, the steam and methane feed rates is set at 90 mol/hour, while the feed rate of methane to the steam reformer furnace is adjusted to 54 mol/hour to ensure that enough combustion heat is available to drive the steam reforming reactions and to preheat the reformer feed streams. 10% excess air is supplied to the furnace to achieve efficient CH₄ combustion.

4.2 MECC CO₂ Capture Modules

For the simulation, the CO₂ capture process is divided into five stages with intercooling between each stage. The first four stages capture 80% of the CO₂ available in the combined power plant flue gas stream, while the last stage captures 10% of the available CO₂. The intercoolers remove the heat of combustion between each stage. More stages with smaller capture fractions could be deployed, but capturing more than 20% of the CO₂ inventory in each stage was found to cause overheating of the membranes in the process simulation. Heat removed between each stage, and at the end of the last stage, is used to preheat unit feed streams and to make steam for electricity generation. The first and second CO₂ capture stages are grouped in the process section "CO₂ Capture Modules #1 and #2." The third, fourth, and fifth CO₂ capture stages are grouped in the process section "CO₂ Capture Modules #3, #4, and #5." Syngas from the steam reformer section is divided into five parallel streams, and each sub-stream is sent to a CO₂ capture module (i.e., Syngas Stream #1 to Module #1, Syngas Stream #2 to Module #2, etc.). The flow rate of syngas to each module is not evenly divided, and 2/9 of the total syngas stream is each provided to Module #1 through #4, and 1/9 of the total syngas flow is provided to Module #5. The flue gas stream is maintained as a single stream, and it flows through each MECC CO₂ capture module in serial fashion (i.e., first Module #1, then Module #2, etc.). This flow arrangement was chosen to maximize the CO₂ driving force in each membrane module while minimizing the volumetric flow rate on the CO₂ capture side of the membranes.

Other flow configurations are possible. Serial flow of both the flue gas stream and the syngas stream was analyzed, but serial flow of the flue gas stream and parallel

flow of the syngas stream allowed for a smaller membrane surface area. Initially, parallel distribution of flue gas and syngas was also pursued, but was abandoned in this analysis due to complications in managing membrane temperatures. In the dual parallel flow arrangement, the flue gas stream and the syngas stream are divided, and each CO₂ capture module operates as an independent unit. Each module therefore had to achieve 90% CO₂ capture on its own in order for the combined system to reach the CO₂ capture target. While this arrangement would minimize membrane surface area, it does not solve the problem of managing membrane temperature, and the membranes would quickly overheat without some means of cooling. In the serial flow arrangements, intercoolers are used to remove the heat of combustion, but in a completely parallel arrangement, some other means of cooling the membrane modules would be needed. For example, a pumped fluid such as water or steam could be used to cool the membrane modules. A more complex membrane module must then be constructed that contains flow paths for flue gas, syngas, a cooling fluid, and this could increase module capital costs if membrane costs are not the dominate contributor to the cost of the CO₂ capture modules. Certainly more work is needed on analyzing these alternative flow arrangements before heavy investment is made into any single flow arrangement.

A partial stream table of the CO₂ capture stages is shown in Table 3. In the table, input retentate and permeate streams and output streams are listed. The retentate input Flue Gas stream is the power plant flue gas stream that is provided to the CO₂ capture process. The retentate input Make-up Air stream is the flow of air that is mixed with the flue gas in order to oxygenate it. The permeate input Syngas Feed is the flow of syngas provided individually to each of the five MECC CO₂ capture modules in the flow sheet.

The permeate output Captured CO₂ shows the flow rate of captured CO₂ from each MECC CO₂ capture module. The retentate output Flue Gas streams show the flow rates of the constituents in the depleted flue gas after emerging from each MECC CO₂ capture module in series, and the final state of the CO₂-depleted flue gas is shown in the rightmost column.

4.2.1 CO₂ Capture Modules #1 and #2

The detailed configuration of this process section is shown in Fig. 7. In this section, power plant flue gas, reformer furnace exhaust, and make-up air are blended in mixer MIX-1 to make a combined stream containing CO₂ and O₂. The heat exchangers HX-5, HX-6, HX-7, and HX-9 are used to set the input temperature of the retentate and permeate streams to approximately 500°C for each module, and to preheat the flue gas and make-up air streams using heat captured from hot exit streams. After preheating the flue gas and make-up air streams to 500°C, some excess heat is available, and this excess heat is used to make high-pressure superheated steam using a steam generator, HX-8. A pump P-2 provides water at 25°C to the steam generator, and a blower C-2 provides make-up air for the capture process.

Pure syngas enters the capture process as the permeate in MECC #1 and MECC #2, and it becomes diluted with captured CO₂ and water vapor as the capture process progresses. In this flow arrangement, the combustion process is syngas-limited, and the extent of combustion is limited by the supply of syngas to the membrane surfaces.

Table 3. Partial Stream Table, CO₂ Capture Section

	Retentate Input (total)	Retentate Input (total)	Retentate Input (total)	Permeate Input (each) to Modules 1, 2, 3, and 4 (for Module 5, multiply by 0.5)	Permeate Output (each) from Modules 1, 2, 3, and 4 (for Module 5, multiply by 0.5)	Retentate Output (from MECC Module #1)	Retentate Output (from MECC Module #2)	Retentate Output (from MECC Module #3)	Retentate Output (from MECC Module #4)	Retentate Output (from MECC Module #5)
	Flue Gas (mol/h)	Furnace Exhaust (mol/h)	Make-up Air (mol/h)	Syngas Feed (mol/h)	Captured CO ₂ (mol/h)	Flue Gas (mol/h)	Flue Gas (mol/h)	Flue Gas (mol/h)	Flue Gas (mol/h)	Flue Gas (mol/h)
Ar		5	5			10	10	10	10	10
O ₂	79	11	116		2	165	123	82	41	21
CH ₄										
CO				20	0.4					
CO ₂	342	55	0.2	0.3	101	318	238	159	80	40
H ₂				60	1.2					
H ₂ O	427	118	10		59	555	586	586	586	586
N ₂	1827	438	433	0.3	0.3	2699	2747	2747	2747	2747

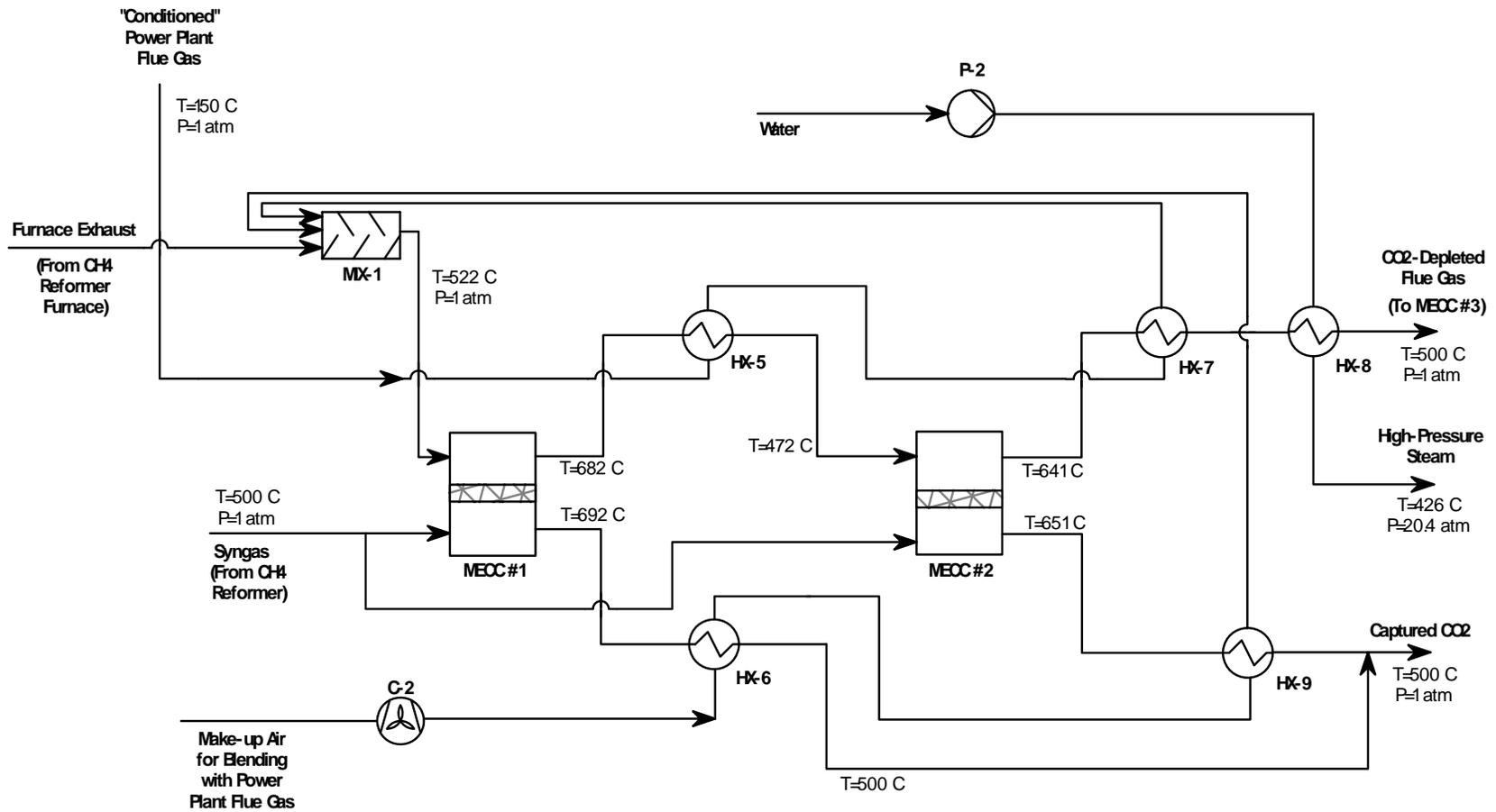


Fig. 7. Detailed block flow diagram of "CO₂ Capture Modules #1 and #2"

4.2.2 CO₂ Capture Modules #3, #4, and #5

The detailed configuration of this process section is shown in Fig. 8. In this section, three CO₂ capture modules are used to capture the rest of the CO₂ in the retentate stream. Since all of the process streams have already been preheated at this point in the process, the excess heat from the combustion process is used to make steam for electricity generation. Steam generators HX-10 through HX-15 are used to boil water to make superheated steam for power production. The exit temperature of the retentate stream ("CO₂-Depleted Flue Gas") and the permeate stream ("Captured CO₂") from this process section is 500°C, and the streams must be further cooled downstream to facilitate safer handling.

4.3 CO₂-Depleted Flue Gas Post-Processing

This process section is labeled in Fig. 5 as "CO₂-Depleted Flue Gas Post-Processing." The CO₂-Depleted Flue Gas stream was cooled from 500°C to 106°C before being released from the process. The heat recovered from the CO₂-Depleted Flue Gas stream is used to generate low-pressure steam at 155°C and 5 atm using a steam generator. The low-pressure steam then drives an electrical turbine to generate electricity.

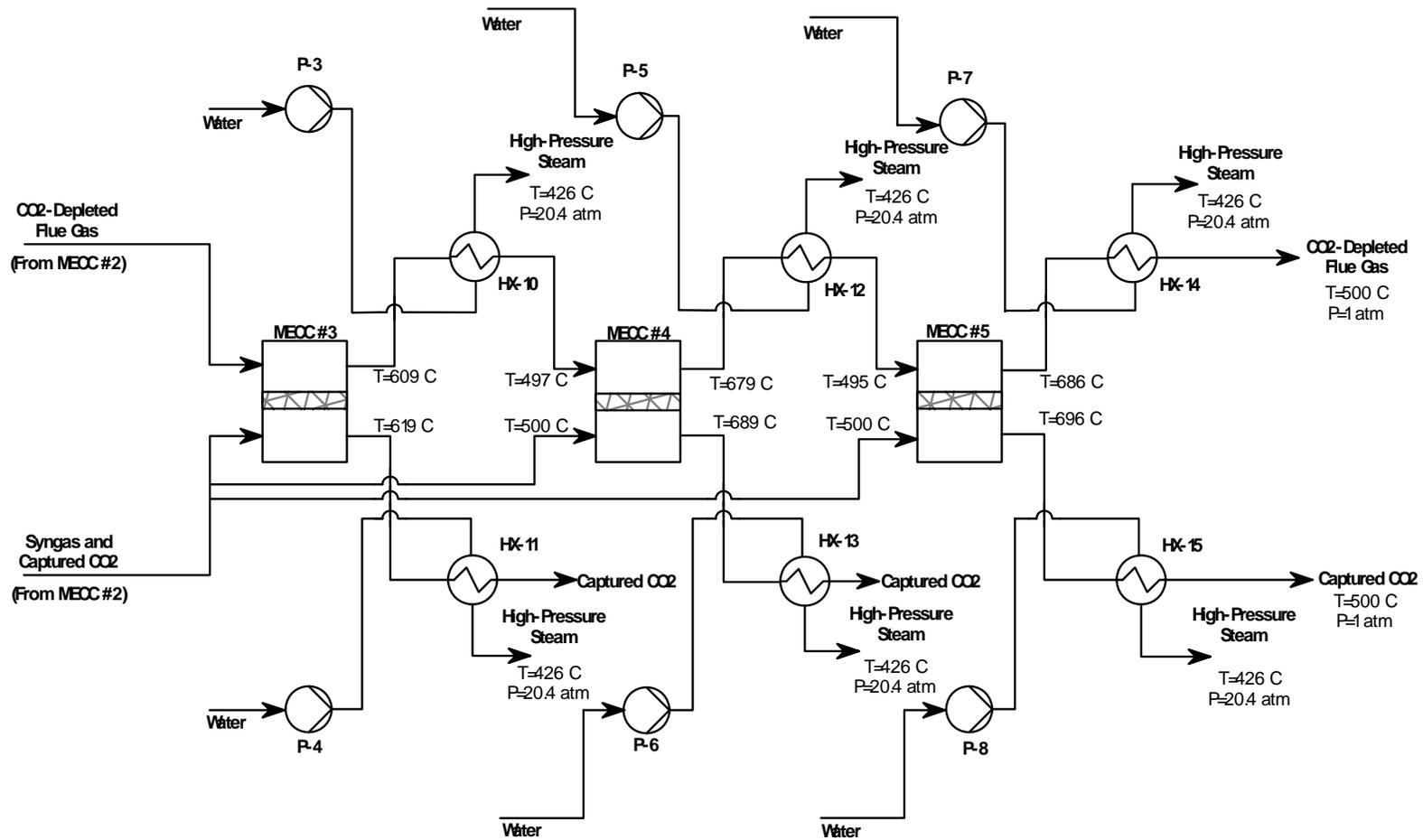


Fig. 8. Detailed block flow diagram of "CO₂ Capture Modules #3, #4, and #5" process section.

4.4 Captured CO₂ Post-Processing

This process section is labeled in Figure 5 as "Captured CO₂ Post-Processing." The Captured CO₂ stream is first cooled from 500°C to 77°C in order to condense water vapor from the captured CO₂. Liquid water is then separated from CO₂ using a gas-liquid separator. The purified CO₂ stream is compressed to approximately 100 atm using a multi-stage compressor, making it suitable for sequestration, transportation, or other uses. The compression step heats the CO₂ stream to about 220°C, but no recovery of heat from this stream is assumed.

The CO₂ stream produced by this process still contains small amounts of O₂, H₂, CO, N₂, and H₂O, and may require further purification, depending upon its use.

4.5 Electrical Power Production

Electricity may be generated from a number of sources in the process. High-pressure steam (426°C, 20.4 atm) is generated from excess heat generated in the CO₂ capture modules #2, #3, #4, and #5, and this may be used to drive electrical turbines. Low-pressure steam (155°C, 5 atm) is generated from the heat removed from the hot CO₂-Depleted Flue Gas and Captured CO₂ streams, and this may be used to drive a lower-pressure electrical turbine, albeit with less efficiency than a high-pressure turbine.

There are also significant electrical power sinks in the process that cannot be neglected when considering the electrical power production capacity of the CO₂ capture process. These draws include the compressor used to pressurize the CH₄ flow to the Steam-CH₄ Reformer, the compressor used to generate a compressed CO₂ product

stream, and the liquid water pumps used to supply water to the reformer and the process steam generators.

A summary of the electrical power generators and electrical power sinks in the CO₂ capture process is shown in Table 4.

Table 4. Significant Electrical Sources and Loads

Description	Sources (W)	Sinks (W)
H.P. Steam, CO ₂ Module #1	0	
H.P. Steam, CO ₂ Module #2	825	
H.P. Steam, CO ₂ Module #3	690	
H.P. Steam, CO ₂ Module #4	1,044	
H.P. Steam, CO ₂ Module #5	1,072	
L.P. Steam, CO ₂ -Depleted Flue Gas	608	
L.P. Steam, Captured CO ₂	167	
Syngas Expander, Reformer	788	
CH ₄ Compressor, Reformer		-349
CO ₂ Compressor, Post-Processing		-2419
Liquid Water Pumps		-24
Sub-Totals:	5,723	-3,139
NET POWER GENERATION = +2,402 W		

According to the table, the CO₂ capture process is capable of generating approximately 2,400 W, given the material flow rates shown in Tables 1 and 2. No electrical power is generated from the heat removed from CO₂ Module #1 because the excess heat is used to preheat syngas and make-up air streams. Likewise, some excess heat from CO₂ Modules #2 and #3 are used to preheat syngas and make-up air streams, and less heat is available to make steam for power production. For CO₂ Modules #4 and #5, the full amount of excess heat is available to make steam for electricity production, and so the power output for these modules is greater than is obtained from the first three. The other electrical power outputs are produced by extracting heat and mechanical energy from other flow streams.

The ability of the CO₂ capture process to become a net generator of electricity reduces the parasitic energy load of the CO₂ capture plant. If it is further assumed that the electrical power plant burns CH₄ to make steam for electricity production; that the steam produced at the power plant is at the same temperature and pressure as the steam produced in the CO₂ capture plant (426°C, 20.4 atm); that the electrical turbines in the power plant have the same efficiency as those used in the CO₂ capture plant; and that the CO₂ capture process, as described above, is sized to capture 100% of the CO₂ produced by the electrical power plant, then the power output of the electrical power plant is 9,700 W, which results from the combustion of approximately 340 mol/h CH₄. Therefore, the combined power output of the electrical power plant and the CO₂ capture process is $9,700 \text{ W} + 2,402 \text{ W} = 12,102 \text{ W}$.

There is an energy cost associated with operating the CO₂ capture process, however. The CO₂ capture plant consumes 144 mol/h CH₄ in order to perform its purpose. The CH₄ used to drive the CO₂ capture process could instead have been used to make electricity in the power plant at greater efficiency. If the fuel feed rate to the electrical power plant were simply increased by this amount to 484 mol/h, then an additional 4,108 W could be generated, giving a total potential power output of 13,808 W. The difference between what might have been produced by a larger electrical power plant (13,808 W) and what is produced by the combined power plant/CO₂ capture facility (12,102 W) is the energy penalty associated with operating the CO₂ capture process. In this case, the energy penalty is about 1,706 W, which is an energy loss of about 12%. Presented another way, the CO₂ capture process consumes 1,706 W to

capture 306 mol/h CO₂ from the electrical power plant, which is equivalent to an energy penalty of 0.41 GJ/metric ton CO₂ captured.

This energy penalty is less than the U.S. DOE upper limit of 20% for parasitic energy losses [13] for CO₂ capture processes and is comparable to energy penalties calculated for other advanced post-combustion CO₂ capture processes. Merkel et al. [16] examined polymeric membrane-based CO₂ capture processes, and developed a process flow sheet for a two-step countercurrent sweep membrane process having a 16% energy penalty. Lively et al. [17] examined a number of proposed post-combustion CO₂ capture processes, and found that the energy penalty (parasitic load) for best-case amine-based absorption systems varied between 26-41%, and the energy penalty for chilled ammonia processes varied between 1-24%, with the smallest energy penalties associated with full thermal integration of the CO₂ capture plant with the electrical power plant.

4.6 Costs

In addition to the 20% upper limit for parasitic energy losses, the U.S. DOE has also established a cost target – operation of a CO₂ capture facility should not increase the cost of electricity from a combined power plant/CO₂ capture facility by more than 30%, as compared to a power plant where no CO₂ capture is performed [3,10].

Evaluation of this criterion for the proposed process is more difficult and will require a great deal more research and development (R&D) before it can be fully addressed. The MECC membranes on which this concept is based are not yet

commercially available, and a reliable cost per unit membrane area can't yet be assigned. Also, the design of the MECC module hardware has not yet been developed.

Some order-of-magnitude calculations can be performed at this stage, however, that will provide some information on the material cost of the membranes. Although the detailed temperature and CO_2+O_2 partial pressure profiles within the membrane modules are unknown at this stage of development, some average membrane conditions can be calculated for each module, thus allowing for a determination of required membrane surface area. Assuming that the membrane modules operate between $500\text{-}700^\circ\text{C}$, the membrane permeance may be calculated by taking the temperature-weighted average of the permeance data shown in Fig. 2 from Ref [1]. The average partial pressure of CO_2 and O_2 in the retentate of each membrane module may be approximated by taking the arithmetical average of the CO_2 and O_2 partial pressures at the inlet and outlet of each membrane module. If it is then assumed that the CO_2 and O_2 partial pressures are zero on the permeate side of the membrane due to the action of the syngas sweep gas, then the sum of the CO_2 and O_2 partial pressures becomes the partial pressure driving force for transport across the membrane. The required membrane surface area within each module is then determined by dividing the total molar capture rate of CO_2 and O_2 (mol/h) in each module by the molar flux rate (mol/h m^2). The molar flux rate is calculated by multiplying the temperature weighted average permeance ($150 \text{ mol/s}\cdot\text{m}^2\cdot\text{Pa}$) times the partial pressure driving force. Table 4 shows membrane surface areas for each module that were calculated using the above algorithm.

Table 5. Calculated Membrane Surface Areas

Module #	CO ₂ +O ₂ Capture Rate (mol/h)	Membrane Permeance (mol/s·m ² ·Pa)	Average CO ₂ +O ₂ Driving Force (Pa)	Membrane Surface Area (m ²)
1	120.7	150 x 10 ⁻¹⁰	14,400	155
2	120.7	150 x 10 ⁻¹⁰	11,600	193
3	120.7	150 x 10 ⁻¹⁰	8,540	262
4	120.7	150 x 10 ⁻¹⁰	5,290	423
5	60.3	150 x 10 ⁻¹⁰	2,730	409
Total membrane surface area (m ²)				1,442

According to Table 5, the total membrane surface area required is 1,442 m². Assuming the structural component of the membrane is composed of porous stainless steel having a thickness of 1.58 mm and a porosity of 0.25, then the mass of stainless steel required for this surface area is 1,442 m² x 0.00158 m x 0.75 * 8,000 kg/m³ = 13,670 kg. Then, if stainless steel costs \$4/kg, then the cost of metal for the membranes is approximately \$54,680.

The membranes will not last for the lifetime of the plant, and the membranes are expected to be replaced at defined time intervals. For the purposes of this calculation, it is assumed that the membrane designs are mature and durable, and will survive at least 20,000 hours in-service before needing replacement. If the power plant has a capacity factor of 0.9, then the CO₂ capture plant will operate 90% of the time, and the membranes will accumulate approximately 8,000 hours of service per year. Over the lifetime of the membranes, the membrane modules will collect about 266 tons CO₂ from the power plant flue gas, according to the process flow sheet described above. At this capture rate, the membrane material costs alone are \$54,680/266 tons CO₂, or \$210/ton CO₂.

The next question to answer is to determine whether this cost is tolerable, or whether it is too high to have any hope of attaining the U.S. DOE goal of not increasing the cost of electricity by 35% or more above the baseline. In the simulated plant, a 35% increase in the cost of electricity translates to an allowance of up to about \$35/ton CO₂ captured, so the total yearly cost of operating the CO₂ capture plant must be less than \$35/ton CO₂ captured in order to be viable. The cost of the membrane materials alone greatly exceeds this limit, and the process will not achieve its cost target.

The membrane costs could be brought within the cost limit if the membrane permeance were increased. Calculations performed in Ref [1] indicate that the theoretical membrane permeance increases monotonically with increasing temperature, and that a permeance of 800×10^{-10} mol/s·m²·Pa or greater could be achieved at a membrane operating temperature of 700°C for the same membrane thickness and porosity. If the membrane permeance were increased to 800×10^{-10} mol/s·m²·Pa, then the membrane material costs would be reduced by a factor of 5 to about \$38/ton CO₂. More work is needed, however, to understand the chemistry of the membranes at higher temperatures, so that membrane performance is not degraded above 650°C. High membrane permeance may only be achieved if the membranes remain stable, and the membrane constituents do not react with each other to make non-conducting by-products. A potential new direction in fashioning such membranes may be to replace the metal support structure with a ceramic material, as is described in Rui et al [6], so that the support structure does not react with the carbonate salt at high temperature. If, in the short term, such advanced membrane types prove to be difficult to make on a large scale or are too expensive, then the maximum operating temperature of the

membranes may need to be lowered to 650°C instead of 700°C, and greater membrane surface area and additional intercooling steps may be required to operate the process. Both changes would result in higher system costs.

More likely, multiple factors will need to be adjusted in the membrane design in order to reduce the membrane cost to tolerable levels. Improvements might be made in understanding membrane chemistry, which may allow actual membrane performance to approach the theoretical performance. A cheaper or less dense support material might be used. Thinner membranes might be used, which would also reduce membrane material costs. Using a thinner membrane would likely increase membrane permeance, and this would have a multiplying effect on membrane cost reduction. The actual membrane lifetime in service is unknown, and it is possible that the membranes could survive 40,000 hours, for example, instead of the arbitrary 20,000 hours that was used in the membrane cost calculations, and this would reduce membrane costs by a factor of 2. The partial pressure driving force across the membranes might also be increased in order to decrease membrane area, but this would require adding a compressor to the flue gas stream, and the energy cost associated with this change would be excessive.

5.0 CONCLUSIONS

The proposed combustion-assisted CO₂ capture process, at the conceptual level, appears capable of meeting at least two of the U.S. DOE's requirements for CO₂ capture processes. With recovery of excess process heat for electricity generation, the proposed process has an energy penalty of about 12%, which is less than DOE's expectation that any successful CO₂ capture process should have a parasitic energy

load no greater than 20%. Also, the process is likely capable of capturing at least 90% of the CO₂ generated by the electrical power plant.

DOE's last criterion, that the cost of electricity from the combined power plant/CO₂ capture facility not be increased by more than 35%, will require a great deal more work before it can be fully addressed. The initial cost of the stainless steel for the membranes is calculated to be about \$210/ton CO₂ captured, which greatly exceeds the U.S. DOE cost target. More laboratory and development work is needed to improve membrane performance and reduce membrane costs before the membranes will be ready for deployment in commercial CO₂ capture applications.

ACKNOWLEDGEMENT

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Table 1. Case Descriptions

	Case #1	Case #2	Case #3	Case #4
Feed Type	Methane	Methane	Coal	Coal
Feed Formula $C_xH_yO_z$, (x,y,z)	(1,4,0)	(1,4,0)	(1,0.7,0.1)	(1,0.7,0.1)
Water-Gas Shift Unit Deployed?	No	Yes	No	Yes
Reformer Thermal Efficiency	70%	70%	70%	70%
Molar Conversion of Feedstock in the Reformer	100%	100%	100%	100%
Water-Gas Shift Molar Conversion	---	75%	---	75%
H ₂ Purification Efficiency	---	89%	---	89%
Moles CO ₂ captured per mole feed	2.1	1.4	0.45	~0.3

Table 2

Partial Stream Table, Reformer Section

	Steam-CH ₄ Reformer, Reforming Section		Steam-CH ₄ Reformer, Furnace Section		Output, Syngas mol/h	Output, Furnace Exhaust mol/h
	Input, CH ₄ Stream mol/h	Input, H ₂ O Stream mol/h	Input, CH ₄ Stream mol/h	Input, Air mol/h		
Ar				6		6
O ₂				132		12
CH ₄	100		60			
CO					100	
CO ₂	2		1	0.2	2	61
H ₂					300	
H ₂ O		100		11		131
N ₂	2		1	486	2	487

Table 3

Partial Stream Table, CO₂ Capture Section

	Retentate Input (total)	Retentate Input (total)	Retentate Input (total)	Permeate Input (each) to Modules 1, 2, 3, and 4 (for Module 5, multiply by 0.5)	Permeate Output (each) from Modules 1, 2, 3, and 4 (for Module 5, multiply by 0.5)	Retentate Output (from MECC Module #1)	Retentate Output (from MECC Module #2)	Retentate Output (from MECC Module #3)	Retentate Output (from MECC Module #4)	Retentate Output (from MECC Module #5)
	Flue Gas (mol/h)	Furnace Exhaust (mol/h)	Make-up Air (mol/h)	Syngas Feed (mol/h)	Captured CO ₂ (mol/h)	Flue Gas (mol/h)	Flue Gas (mol/h)	Flue Gas (mol/h)	Flue Gas (mol/h)	Flue Gas (mol/h)
Ar		5	5			10	10	10	10	10
O ₂	79	11	116		2	165	123	82	41	21
CH ₄										
CO				20	0.4					
CO ₂	342	55	0.2	0.3	101	318	238	159	80	40
H ₂				60	1.2					
H ₂ O	427	118	10		59	555	586	586	586	586
N ₂	1827	438	433	0.3	0.3	2699	2747	2747	2747	2747

Table 4. Significant Electrical Sources and Loads

Description	Sources (W)	Sinks (W)
H.P. Steam, CO ₂ Module #2	825	
H.P. Steam, CO ₂ Module #3	690	
H.P. Steam, CO ₂ Module #4	1,044	
H.P. Steam, CO ₂ Module #5	1,154	
L.P. Steam, CO ₂ -Depleted Flue Gas	890	
L.P. Steam, Captured CO ₂	245	
Syngas Expander, Reformer	875	
CH ₄ Compressor, Reformer		-388
CO ₂ Compressor, Post-Processing		-2726
Liquid Water Pumps		-25
Sub-Totals:	5,723	-3,139
NET POWER GENERATION = +2,584 W		

Table 5. Calculated Membrane Surface Areas

Module #	CO ₂ +O ₂ Capture Rate (mol/h)	Membrane Permeance (mol/s·m ² ·Pa)	Average CO ₂ +O ₂ Driving Force (Pa)	Membrane Surface Area (m ²)
1	120.7	150 x 10 ⁻¹⁰	14,400	155
2	120.7	150 x 10 ⁻¹⁰	11,600	193
3	120.7	150 x 10 ⁻¹⁰	8,540	262
4	120.7	150 x 10 ⁻¹⁰	5,290	423
5	60.3	150 x 10 ⁻¹⁰	2,730	409
Total membrane surface area (m ²)				1,442

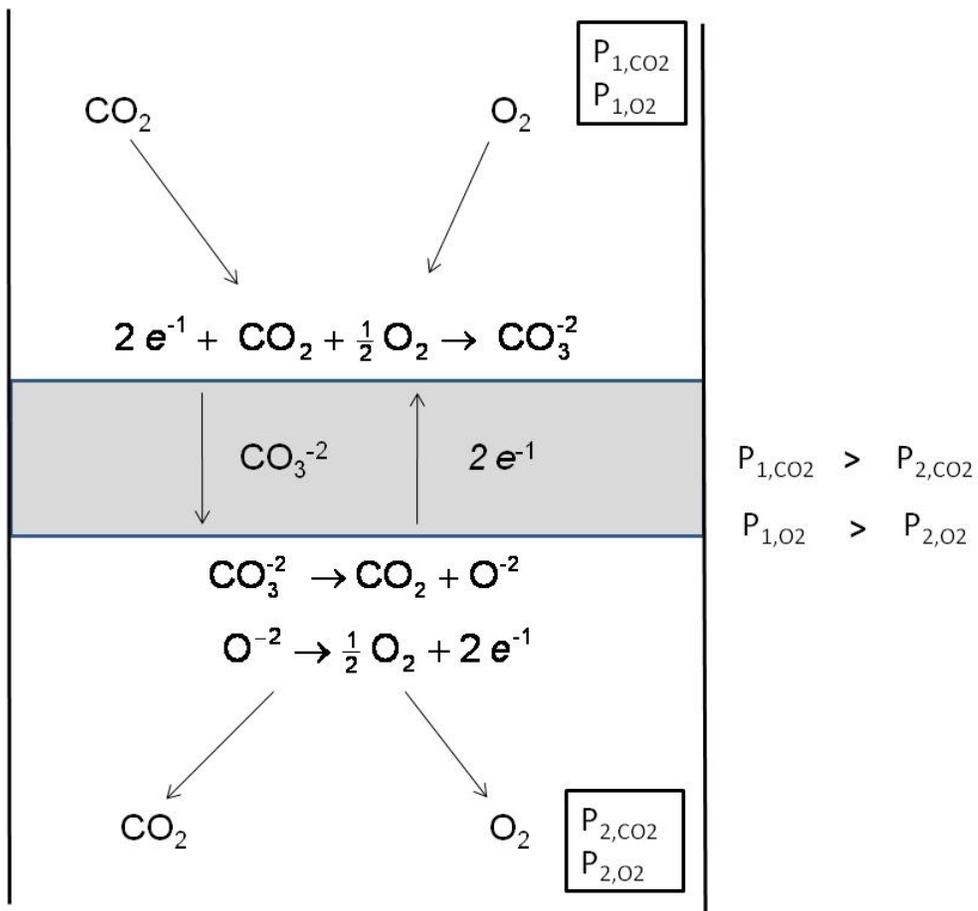


Figure 1: MECC Membrane CO₂ Transport Half-Cell Reactions.

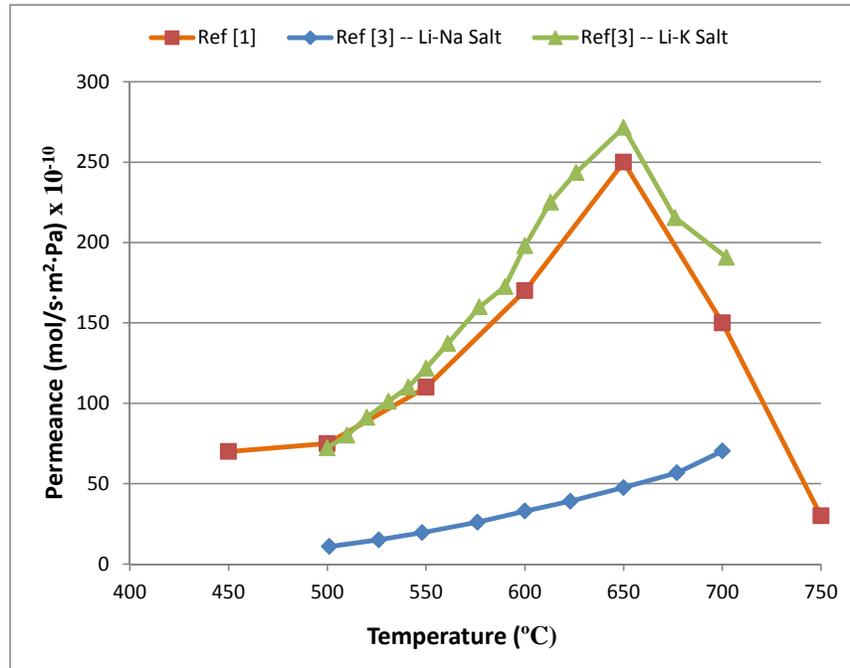


Fig. 2. MECC membrane permeance versus temperature

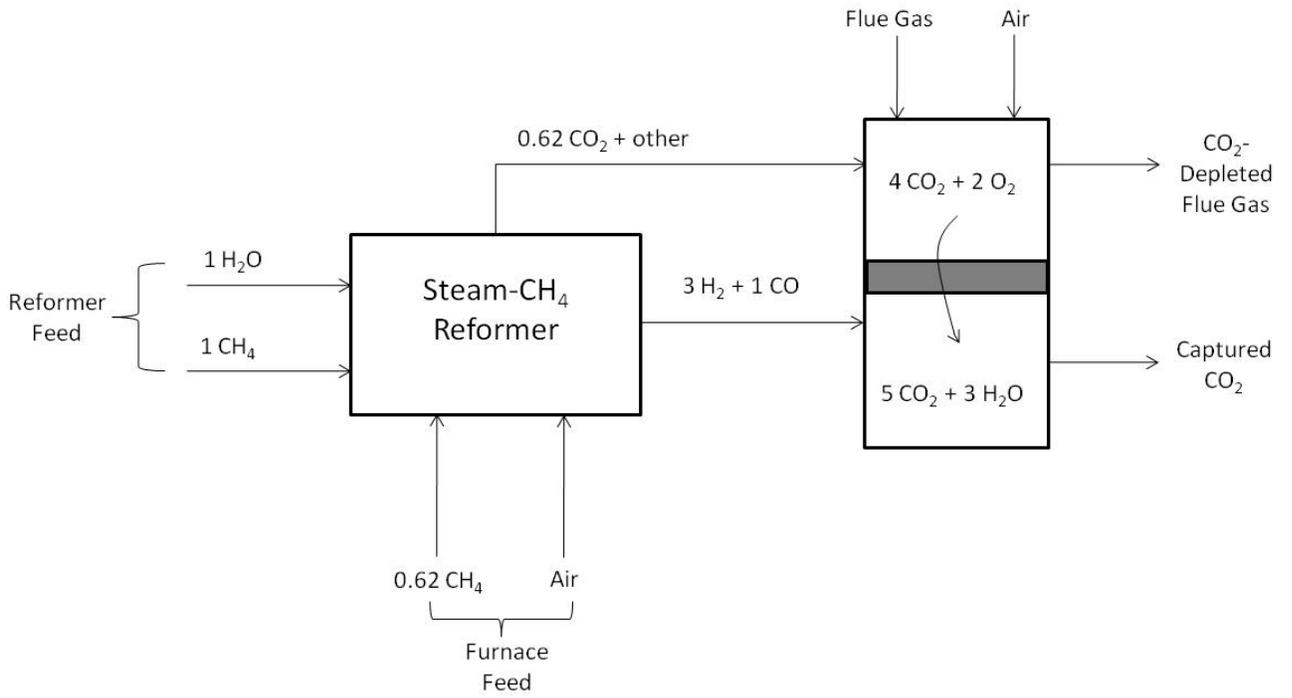


Figure 3. Simple schematic of Syngas Combustion-Assisted CO₂ Capture System.

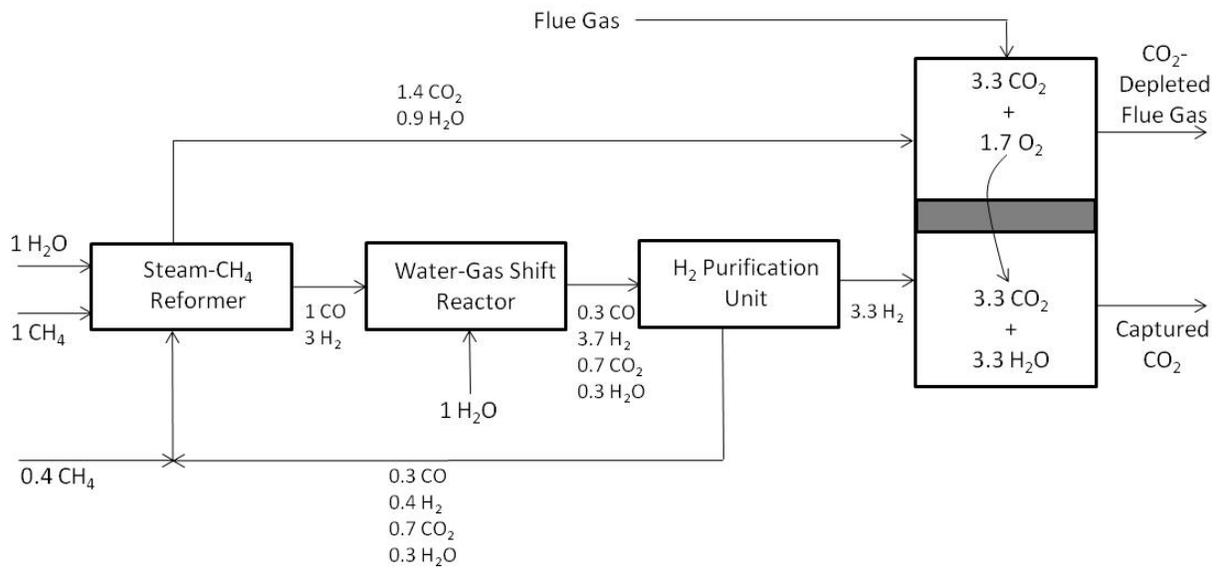


Fig. 4. Block flow diagram of Case #2.

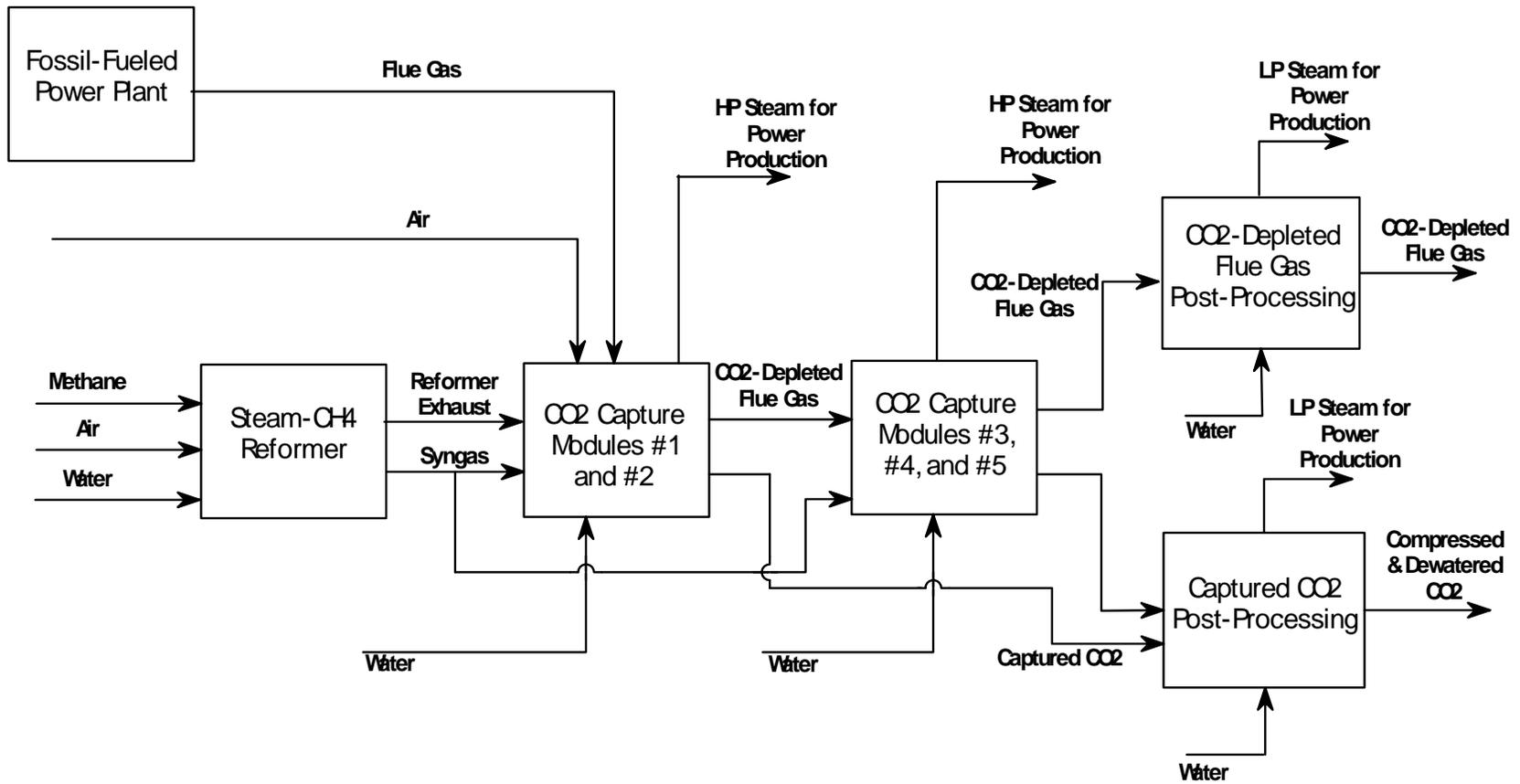


Figure 5. Block flow diagram of proposed combined power plant/CO₂ capture facility.

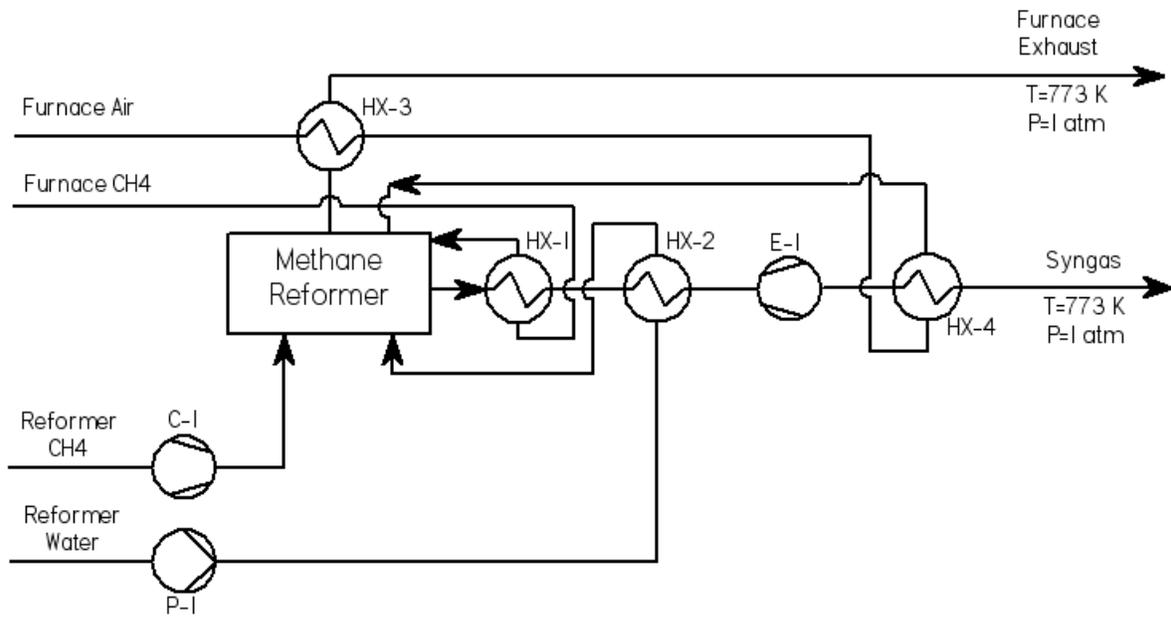


Figure 6. Detailed block flow diagram of "Steam-CH₄ Reformer" process section.

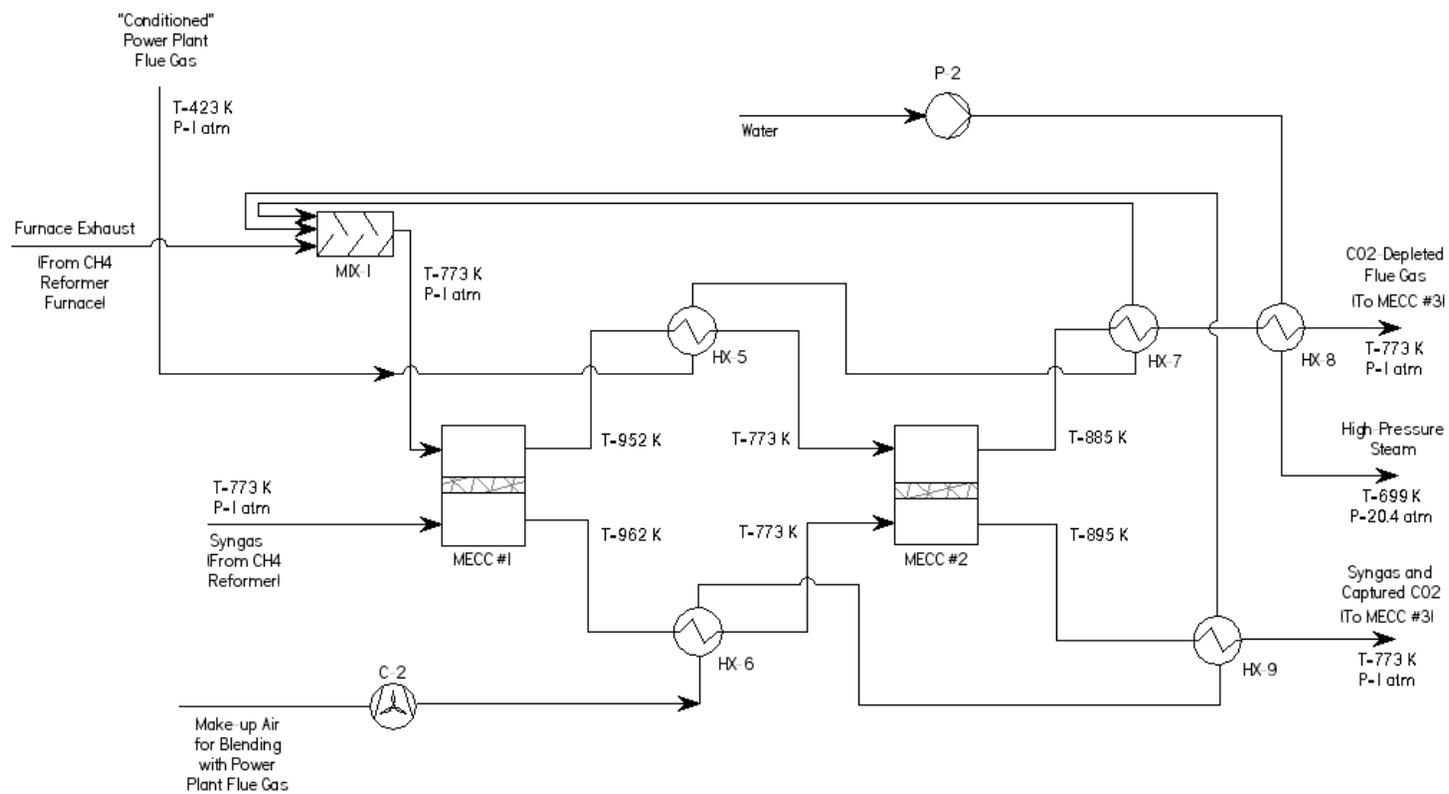


Figure 7. Detailed block flow diagram of "CO₂ Capture Modules #1 and #2" process section.

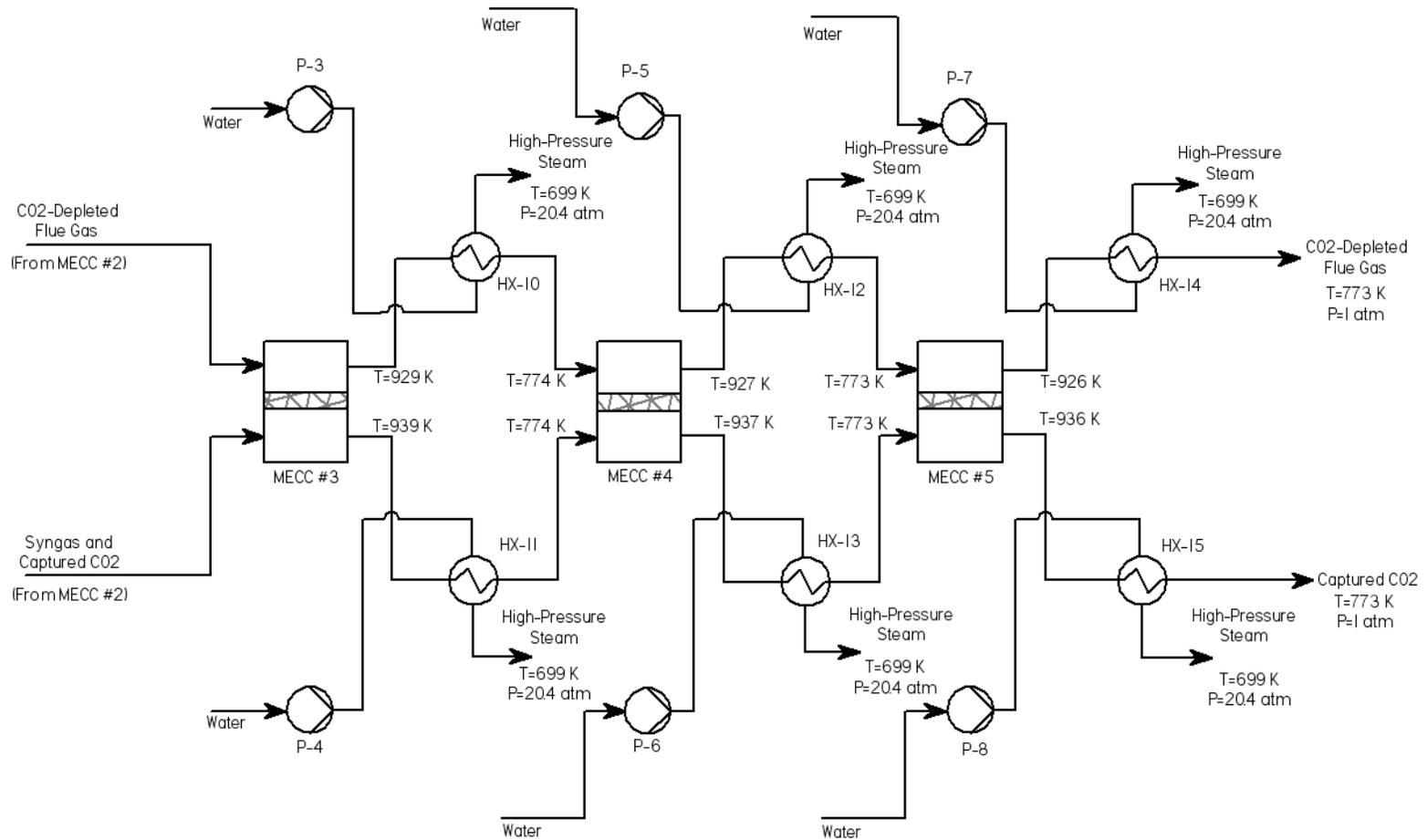


Figure 8. Detailed block flow diagram of "CO₂ Capture Modules #3, #4, and #5" process section.