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Production of Carbon Neutral Methanol Using Co-Electrolysis of CO₂ and Steam in Solid Oxide Electrolysis Cell in Tandem with Direct Air Capture

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The increasing concentration of carbon dioxide (CO₂) in the atmosphere since the industrial revolution is a major contributor to climate change. Among the several options to tackle this issue, the removal of CO₂ from the atmosphere and its subsequent use is becoming increasingly attractive. This paper presents a techno-economic feasibility study and quantification of the environmental benefits of combining direct air capture (i.e. capturing CO₂ from the atmosphere) with co-electrolysis of water and the captured CO₂ in a solid oxide electrolyser cell (SOEC). It was found that the fuel methanol could be modelled to be produced in a carbon negative manner but is not profitable due to current SOEC capital cost. The capital cost of the SOEC is expected to be cut in half by 2030. This would allow for profitable production of methanol.

Introduction

Increase in the amount of atmospheric carbon dioxide and the associated increase in global temperatures is a significant threat to society. This temperature increase is already impacting living conditions in some parts of the world, for example droughts in the United States [1]. These impacts are expected to spread and intensify if no action is taken to mitigate them. Carbon capture technology is one of many solutions being considered to combat climate change. A current challenge with carbon capture is lack of economic incentive [2]. This paper sets out to design a model for a plant which would capture carbon dioxide from the air (aka. Direct Air Capture or DAC) and convert it to a fuel. Fuels produced using captured carbon dioxide are better for the environment than those produced using traditional methods because the traditional methods result in “new” carbon dioxide emissions, whereas this method allows for reuse of previously emitted carbon dioxide.

Aspen Plus was used to design a model that contains three key parts, which are: 1) direct air capture process, 2) carbon monoxide and hydrogen production using a Solid Oxide Electrolyzer Cell (SOEC), and 3) methanol production for the fuel considered here. The DAC process is modelled after a process in the pulp and paper industry known as the Kraft process. This modified process allows for absorption of ultra-dilute (400 ppm) carbon dioxide from air. Large fans are used to help pass air through the process and filtered air is released back into the atmosphere. Other inputs to the process are water and electricity with a mixture of water vapor and carbon dioxide being output as the product of the process. Water can be removed as needed using a water knockout drum.

The SOEC is an electrolytic cell which uses electricity to convert water and carbon dioxide to carbon monoxide and hydrogen. Carbon monoxide and hydrogen are key inputs for fuel production. Unconverted carbon dioxide is another input for methanol production. The SOEC allows for a clean production of these inputs. Water undergoes electrolysis to be converted to hydrogen and oxygen. Then, this model takes advantage of the water gas shift reaction equilibrium at high temperatures to shift the equilibrium to convert carbon dioxide to carbon monoxide. Methanol is produced using the products of the SOEC at an elevated pressure and reduced temperature, which are kinetically favourable. Any remaining water in the SOEC product stream is removed before being sent to the methanol reactor. Finally, a series of distillation units are used to obtain a high purity methanol product.

Direct Air Capture Model Development

The DAC design was based on the Carbon Engineering design presented in [3]. The design was altered to become more akin to the pulp and paper Kraft process. This allowed the design to be more accurately simulated using Aspen Plus without any specialized user models. The fluidized bed causticizer in the Carbon Engineering design was replaced by a series of mixing tanks. The fluidized bed calciner was replaced with a rotary kiln which is the more common calciner used in the pulp and paper industry [4]. A schematic of the process is presented in

Figure i. The following paragraphs briefly describe the main DAC components.

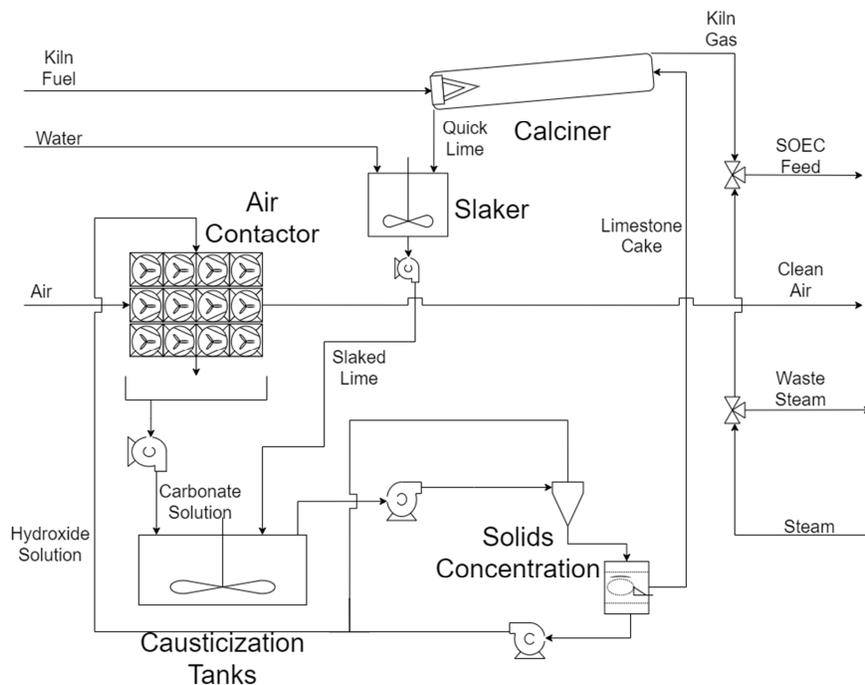


Figure i. Overall schematic of the Direct Air Capture process

Air Contactor

The air contactor used is the same design used by Carbon Engineering [3]. For a CO₂ capture rate of 0.25 megatonnes per year it was found that an inlet area of 11200 m² is required. This corresponds to a power requirement of 2.14 MW for the pumps and fans and a packing volume of 75600 m³. These values were obtained using the optimized parameter values in [5].

Causticization Tanks

The causticization tanks are simple reactor vessels that do not require any heat management as the reaction is spontaneous but dilute enough to not cause temperature spikes. The reactions also occur at atmospheric pressure. The main problem to address with these tanks is the long residence time (about 90 minutes [4]) which increase the tank volume requirements. A floating roof storage vessel was used due to the large storage capacity with propellers to achieve mild agitation. The propellers were then found to have a power draw of 2.55 MW [6].

Solids Concentration

Due to the process changes, the particle size of solids coming out of the causticizer has been drastically reduced. This leads to a lower solids content being sent to the calciner and this needs to be rectified. This is done using an array of hydrocyclones followed by a rotary vacuum filter. The precipitated calcium carbonate is assumed to be uniformly distributed in a geometric particle size distribution ranging from 5 to 50 µm. This range was chosen so that the approximate average particle size is 25 µm as mentioned in [3].

Calciner Kiln

The calciner was sized according to heuristic limits [6]. Excel's solver was used to optimize the design by minimizing the price based on the diameter, length/diameter ratio, and the number of kilns while meeting the flow constraints. The optimal parameters were found to be 3 kilns at a diameter of 3.81 m and a length/diameter ratio of 40, and the corresponding power requirements were 0.78 MW.

Other Units

The slaker was sized in a similar manner to the causticization tanks with an additional headspace of 10%. An axial turbine was used for agitation which had a power consumption of 3.14 kW. The hydrocyclone pump was split into 10 units and the brake power was determined from the Aspen simulation to be 96.4 kW per pump.

Additionally, an air separation unit is included in the model (but not simulated in Aspen) to produce high oxygen content air for oxy-combustion in the calciner. Using the values in [7], the total power draw required was found to be 12.0 MW. Fuel combustion is required due to the high operating temperature of the kiln (900 °C). The combustion

products, composed mainly of CO₂ and water, are combined with the captured CO₂. The water produced from combustion is used in the feed to the SOEC.

SOEC Model Development

The modelling of the SOEC process was done in Aspen Plus. Since Aspen Plus cannot model an actual electrolyzer cell, equipment available in Aspen Plus which best represents the actual conditions was selected. A schematic of the SOEC Aspen Plus model is shown in **Figure ii**. RGibbs reactors were used to model the equilibrium, since the different components go to equilibrium inside the cell. An RStoic reactor was used to model the electrochemical reactions, neglecting the formation of ions, and leaving components in their diatomic states since modelling the ions was not necessary. The conversion inside this unit was selected through investigation of the impact on electricity usage compared to hydrogen production. A calculator block was used to predict electricity requirements of the cell as well as additional heat required to account for the endothermic nature of the electrochemical reactions. A separator unit was used to model the electrolyte, which would allow for full separation of oxygen and the other components. In reality, the electrolyte allows for only oxygen ions to pass through. Since the cell is isothermal, hot air is fed to the cell to account for heat consumed by isothermal reactions. Under the condition of this paper, very small conversion occurs in “RWGS” therefore the heat duty of “COOLER” was set to the sum of the heat consumed in “EQUIBOUT” and the heat required for the electrochemical reactions beyond what electricity provides. This air is fed to the portion of the SOEC (the anode side) where only oxygen is present. This provides heating to the SOEC without impacting reactions since the air cannot pass through the electrolyte. This air enters the system at room temperature, thus must be heated. Since the downstream processes require the product at significantly lower temperatures, the final product is fed to a heat exchanger where the cooling fluid is the air that needs to be heated to provide heat to the cell. The Stream “AIROUT2” is used to preheat the warm air stream to reduce the amount of energy required for the furnace “AIRHEAT”. This was done using a heat exchanger.

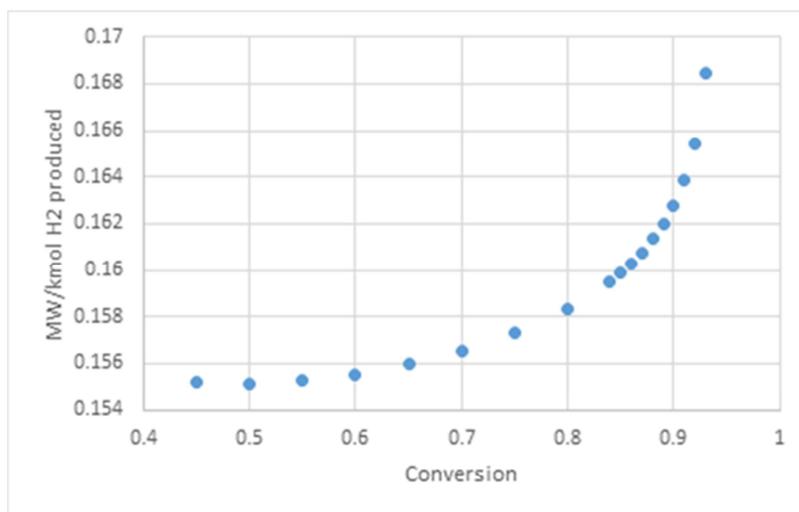


Figure iii. Electricity requirement as a function of conversion in fuel electrode

Cell Parameter Modifications

When using the parameters found in [9] to calculate the cell voltage, it was found that the allowable current density was limited by the concentration overpotential calculations. It was desired to operate at a current density of 5000 A/m^2 since this aligns with literature. The parameters selected for investigation were selected based on their influence on the concentration overpotential. The range of typical values found in literature were used as lower and upper limits. The parameters selected were the porosity, tortuosity, fuel electrode thickness and pore radius. Additionally, the specific ohmic resistance was investigated because when fitting these values to experimental data [9] it was found that the slope of the relationship between current density and cell voltage was notably different than that for the experimental data. Ohmic overpotentials are the most significant contributor to the difference between reversible cell voltage and actual cell voltage, therefore this parameter was also selected for investigation.

MATLAB was used to minimize the sum of the squared difference between experimental values from [9] and those predicted with this model. **Table i** shows the chosen model parameters, their original value, the chosen boundaries and the resulting fitted values. In **Table i**, δ_{FE} is the active thickness of the fuel electrode, $r_{ohmic,conc}$ is the specific ohmic resistance of the cell, δ_{el} is the electrolyte thickness, r_p is the pore radius, ϵ is the porosity and τ is the tortuosity. The other parameters used to calculate the cell voltage came from [9]. Any limits in **Table i** for which there is not a reference, a small arbitrary range was selected for variation.

Table i. Determination of Cell Parameters

| Parameter | Units | Original Value [8] | Lower Limit | Upper Limit | Fitted Value |
|------------------|----------------|--------------------|-------------------------|-----------------------|-----------------|
| δ_{FE} | m | $3.2 * 10^{-5}$ | $3.5 * 10^{-5}$ [10] | $3 * 10^{-4}$ [10] | $1.1 * 10^{-4}$ |
| $r_{ohmic,conn}$ | $\Omega * m^2$ | $5.7 * 10^{-6}$ | $1 * 10^{-7}$ | $1 * 10^{-5}$ | $7 * 10^{-7}$ |
| δ_{el} | m | $1.25 * 10^{-5}$ | $1 * 10^{-5}$ | $1.5 * 10^{-5}$ | $1.1 * 10^{-5}$ |
| r_p | m | $1 * 10^{-6}$ | $5 * 10^{-7}$ | $1.5 * 10^{-6}$ | $5 * 10^{-7}$ |
| ϵ | - | 0.3 | 0.3 | 0.5 [10] | 0.4 |
| τ | - | 5 | 3 [11] | 6 [11] | 3.9 |

Feed Ratio Selection

The final operating condition of the SOEC that was investigated for this analysis was the feed to the SOEC which yields the desired inputs for the methanol process. To determine the desired ratio of water to carbon dioxide entering the methanol process, the inputs required for each conversion were found to yield the following relation for the SOEC outputs [12].

$$\frac{\dot{n}_{H_2} - \dot{n}_{CO_2}}{\dot{n}_{CO} + \dot{n}_{CO_2}} = 1.91 \quad [1]$$

Heat Integration of SOEC

An advantage to combining the DAC with the SOEC is that the products from the DAC are at a high temperature ($800\text{ }^\circ\text{C} - 860\text{ }^\circ\text{C}$), which is a good match for the high temperature SOEC. Cooling of the final product stream was required because the downstream processes required the inputs (SOEC outputs, stream SOECOUT in **Figure ii**) to be at a temperature of $70\text{ }^\circ\text{C}$. Additionally, air must be heated as this is the source of heat for the cell to maintain isothermal operation. First, the air entering at $25\text{ }^\circ\text{C}$ was preheated while reducing the SOEC product stream, then this inlet air was further preheated using the air exiting the SOEC at $800\text{ }^\circ\text{C}$. This allowed for minimization of heat provided through burning methane, occurring in the air heater (unit "AIRHEAT" in **Figure ii**).

Methanol Production Model Development

The Aspen Plus model used in this report to model the methanol production process was based on [12]. **Figure iv** shows the entire methanol production Aspen Plus flowsheet with labelled streams and units. The operating conditions of the heat exchangers and distillation units were adjusted due to a change in the inlet flow rate and composition from the original design. The methanol catalytic reactor has an inlet feed containing both carbon monoxide and carbon dioxide. The feed from the SOEC enters a flash distillation unit to remove excess water, which contribute to increase the overall conversion of carbon monoxide and carbon dioxide to methanol. The inlet gaseous stream then enters a

multistage compressor to obtain the required operating temperature and pressure of approximately 232°C and 71 atm, respectively. The methanol reactor was simulated as a packed-bed reactor. The product of the reactor then passes through a multistage cooling system before it is separated in a flash distillation unit, the material exits the cooling system at 40°C. The steam that is generated from this cooling unit is used in R-HEAT1 and R-HEAT2 to reheat the unreacted material. Some of the remaining steam, exiting COOLER1 in stream COOL4, is then added to the carbon capture product. The distillate of the first flash distillation column is recycled, back to the reactor, to increase the overall conversion of the process and the bottoms product of DIST1 is further refined. The main components of the recycle stream are hydrogen gas, carbon monoxide and carbon dioxide. The bottoms product passes through a second flash distillation unit to remove more unreacted material from the product stream. Finally, the stream enters a tray distillation column to remove the remaining water and increase the purity of the product. The product stream, PROD, exits the top of the tray column.

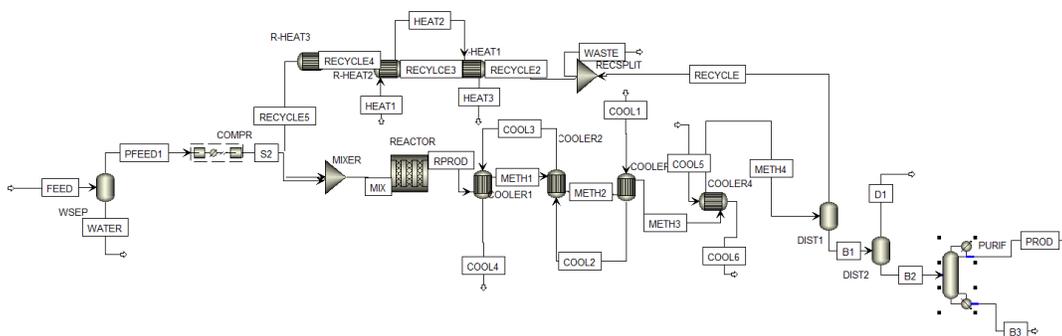


Figure iv. Methanol production process Aspen model [12]

Results

This section reviews the key process results. Some operating parameters must be modified within the carbon capture and SOEC sub-processes in order to provide an optimal feed from the SOEC. The most significant of these parameters is the SOEC electricity demand. This is of particular importance to the economic analysis since the costing of the SOEC unit was linearly related to its capacity. **Table ii** shows the required SOEC capacity for each design case. This table indicates that electricity demand above 200 MW is necessary here, the basis for the size of the process being an amount of CO₂ captured through the DAC of 0.25 megatonnes per year. Recall that the cases are based on water conversion in the fuel electrode, with values of 80% for Case 1 and 85% for Case 2.

Table ii. SOEC Electricity Demand by Case

| Case 1 (MW) | Case 2 (MW) |
|-------------|-------------|
| 203 | 208 |

Since the reaction kinetics of methanol are composed of reversible reactions, excess water must be removed from the feed stream to prevent methanol from being converted back into carbon dioxide. This would result in a lower final product output and lower

purity. The water separation unit, labelled WSEP in **Figure iv**, removed at least 90.7% of the water in the inlet stream, increasing the amount of methanol produced in the reactor. Note that 5.32% of carbon dioxide, dissolved in the water, is also removed in the WSEP.

The mole fractions of the final product are shown in **Table iii**, along with the molar and mass flow rate, for both SOEC cases.

Table iv. Methanol product flow rate by Case

| Species | Case 1 (mol%) | Case 2 (mol%) |
|---|---------------|---------------|
| H ₂ O | 0.28 | 0.32 |
| CH ₄ O | 94.9 | 95.3 |
| CO ₂ | 4.76 | 4.38 |
| CO | 0.01 | 0.01 |
| H ₂ | 0 | 0 |
| N ₂ | 0 | 0 |
| Total Molar Flow Rate (kmol/h) | 842 | 856 |
| Total Mass Flow Rate (tonne/day) | 658 | 668 |

The methanol production process, for case 2, produces approximately 856 kmol/h of methanol, accounting for 96.8% of the methanol exiting the reactor. The remaining 3.2% was contained in the various waste streams of the process. This product stream also accounts for 76.2% of the entire carbon input to the system. This indicates that for this plant design, more than 75% of the captured CO₂ is effectively utilized to produce methanol.

The final point of interest for the methanol production process is the purity of the product. To increase the purity of methanol in this process a distillation column unit was used to remove the excess water in the product stream. The product stream exiting the distillate of the column contained a purity of the 95.26 mole%. The only impurity of consequence is carbon dioxide, 4.38 mol% of the product.

Environmental Analysis

The environmental analysis for this project focuses on net carbon removed (or added) to the atmosphere. The purpose of this design is to produce a valuable product while reducing carbon emissions. To determine if the product is produced in a carbon negative manner the fuel output was compared to the amount of carbon released during the production process. This analysis considers only the mass balance of carbon through the system as shown in **Figure v**. It was assumed that all electrical loads are met through non-emitting power sources.

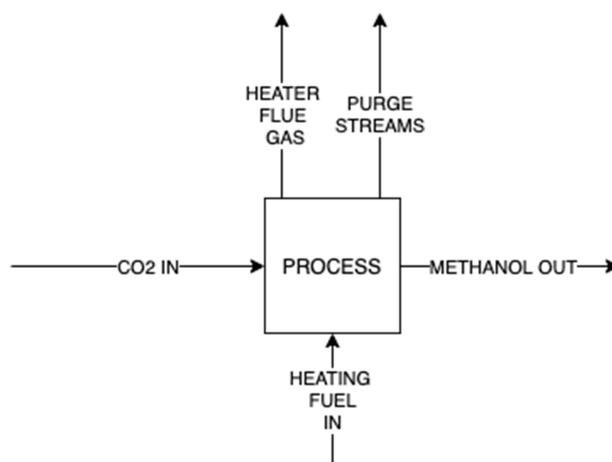


Figure v. Carbon Mass Balance

The results of this carbon balance are summarized in **Table v**. Process emissions are the sum of the heater flue gas and purge streams.

Table vi. Methanol Carbon Balance

| | Case 1 | Case 2 |
|---|---------------|---------------|
| Fuel Out (kmol/h) | 842 | 856 |
| CO₂ Emissions (kmol/h) | | |
| Heater Flue Gas | 174 | 156 |
| Purge Stream | 245 | 240 |
| Total | 419 | 396 |
| Atmospheric CO₂ Captured (kmol/h) | 591 | 591 |
| kmol Carbon Removed per kmol Fuel | 0.20 | 0.23 |
| gCO₂/kgMeOH Removed From Atmosphere | 275 | 316 |

The results for the methanol process show that more carbon is captured in the product stream than is emitted for both cases. This suggests that methanol could be produced in a carbon negative manner. Case 2 removes 11% more CO₂ than case 1 and therefore the SOEC unit should be operated at 85% conversion in order to maximize carbon captured. For case 2, although 591 kmol/h of CO₂ is captured in the DAC, 396 kmol/h of CO₂ is emitted in the process. This means that the net CO₂ captured is 195 kmol/h which is only ~33% of the CO₂ processed in the DAC. A major source of emissions in the process comes from the SOEC preheater; a possibility could be to increase the air separation unit throughput and use oxy-combustion for the SOEC preheater from which the CO₂ could be captured and directed into the SOEC feed.

Cost Analysis

The purpose of this cost analysis is to determine a preliminary estimate of the project's economic feasibility. Capital costs were estimated using a combination of

industry knowledge, engineering heuristics and literature values for chemical engineering equipment. Literature values used were largely based on the Ulrich and Vasudevan method [6]. Although the case study was done for Ontario, Canada, all reported values are in 2019 USD. We used an exchange rate of 1.33 CAD/USD.

The following assumptions were used to develop this economic analysis.

- A factor of 1.18 on equipment costs accounts for piping and installation.
- Maintenance and labor costs are 2% of the equipment capital costs per year.
- Plant life is 25 years with 15% downtime for maintenance.
- Interest rate is 10%.
- Price of methanol is \$0.61/kg.
- Cost of electricity is \$10.19/MWh based on bulk Ontario electricity prices [13]
- Cost of cooling water is \$1/m³.
- Cost of steam is \$3/GJ. [14]
- Cost of methane is \$1.11/kg. [15]
- A 25-year straight line depreciation was used for equipment costs.

Table vii shows the installed cost of equipment before considering factors such as maintenance and interest payments. It can be seen that the SOEC unit accounts for ~68% of the overall capital cost. The cost per kW of the SOEC was estimated at 2000 euros [13]. Based on this analysis the capital cost of purchasing SOEC equipment will have the largest impact on the project's economic feasibility. This illustrates the significant impact future technology advancements or price reductions could have on the economics of this design.

Table vii. Capital Cost of *DAC/SOEC/Methanol Production Process*

| Process | Capital Cost (Million \$) | |
|---------------------------|---------------------------|------------|
| | Case 1 | Case 2 |
| Direct Air Carbon Capture | 229 | 229 |
| SOEC | 568 | 578 |
| Methanol Production | 40 | 40 |
| Total | 837 | 847 |

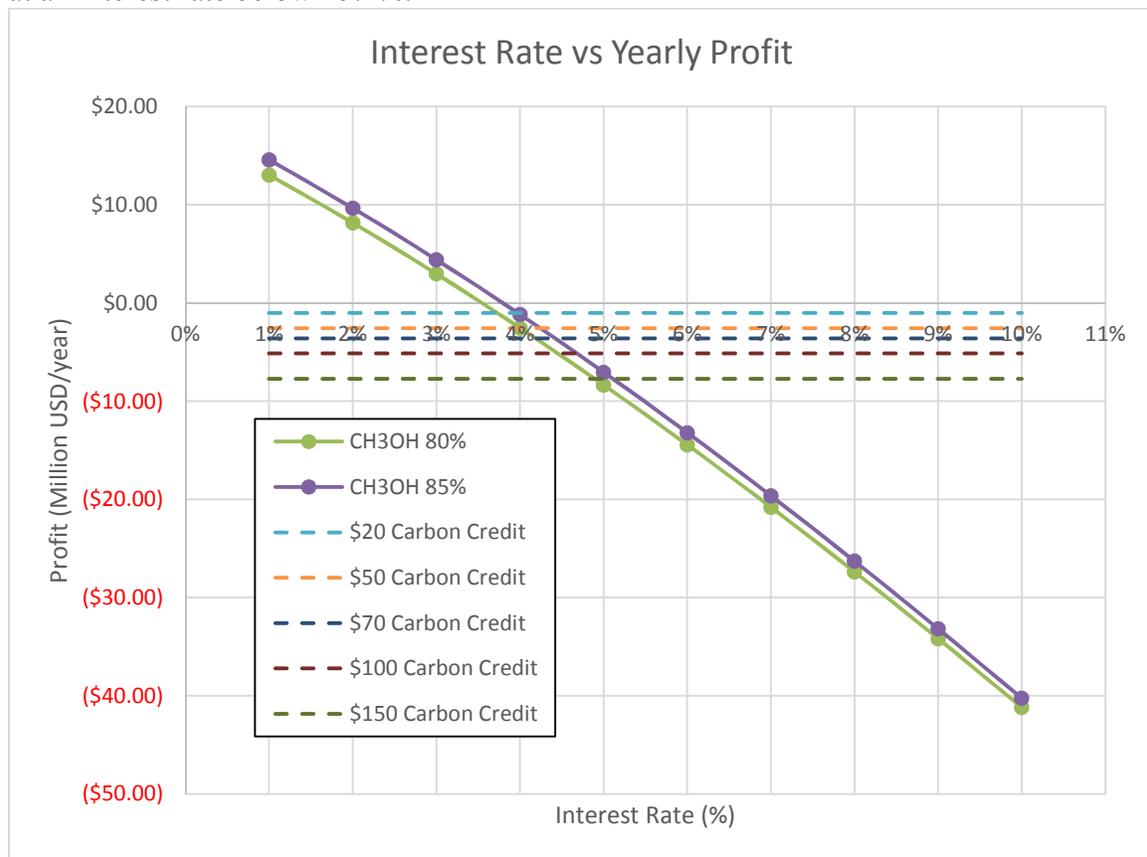
Table viii shows the annual cost and revenue of the overall process. This table indicates that a large portion of the annual cost of operation is due to the high capital cost of the plant and equipment. This finding is contrary to initial expectations that electricity prices would dictate the project's financial feasibility.

Table viii suggests that the 85% conversion case for methanol is preferable, in which case, it is expected that the process will cost \$40M/year to operate. Given that this is a net loss, reductions in the project's cost are required improve the financials. These cost reductions could be realized as technologies like SOEC become more mature or if a low enough interest rate could be obtained.

Table viii. Annual Cost and Revenue of Methanol Production Processes at an Interest Rate of 10%

| | Case 1 | Case 2 |
|---|--------|--------|
| Annual Revenue (Million \$/year) | 123 | 125 |
| Annual Cost (Million \$/year) | | |
| Equipment | 109 | 110 |
| Utility | 55 | 55 |
| Total | 164 | 165 |
| Annual Profit (Million \$/year) | -41 | -40 |

A sensitivity analysis was done on the project's interest rate to determine the rate required before the process becomes profitable. This analysis is shown in **Figure vi**. This figure also shows the maximum interest rate for the process to be profitable given different levels of carbon credits (\$/tonne-CO₂). For example, at an 85% conversion rate, the process would be profitable without carbon credits if the interest rate is less than ~3.75% whereas with a carbon credit of \$150/tonne-CO₂, the process would be profitable at an interest rate below ~5.2%.

**Figure vi.** Profit vs. Interest Rate Sensitivity Plot

A similar sensitivity analysis can be done on the cost of electricity. The cost of electricity used for this analysis was chosen based on the Ontario wholesale electricity costs for 2019. Since the project is expected to consume power in the range of 200 MW it is reasonable to assume that power will be purchased in bulk quantities. This could potentially involve a power purchasing agreement between the direct air carbon capture and utilization facility and an independent power producer. For these reasons it is desirable to analyze at what electricity price the process would become profitable. This analysis was done with a set interest rate of 10% and it was found that the profit is negative even with no electricity costs. This suggests that while reducing power costs does benefit the project financials, it is the capital cost and interest rate which control the economic feasibility.

Conclusions

In conclusion, this project has determined that direct air carbon capture in tandem with SOEC and methanol production could be used as a way of producing carbon negative fuels but more work needs to be done to improve the economics. The best design scenario investigated in this report is the case of methanol production with 85% conversion in the SOEC unit (case 2). A higher SOEC conversion would cause exponentially higher electricity demand so 85% was chosen as the maximum conversion for consideration. The SOEC electricity demand for this scenario is expected to be 208 MW.

The economic feasibility of this design depends on the chosen interest rate, the future costs of SOEC and direct air carbon capture technologies as well as potential carbon taxes. It should be noted that the SOEC equipment costs account for approximately 66% of the project's capital cost and has a large impact on the cost analysis. As this technology becomes less expensive the economic feasibility of the process could improve significantly. Additionally, a sensitivity analysis was done on the assumed interest rate of 10% and it was found that at interest rates below 4% the project is profitable.

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