nature chemical engineering

Supplementary information https://doi.org/10.1038/s44286-024-00137-y

Process and techno-economic analyses of ethylene production by electrochemical reduction of aqueous alkaline carbonates

In the format provided by the authors and unedited

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Experimental Methods

Fabrication of Cu electrodes*:* A catalyst ink consisting of 50 mg Cu nanoparticles (25 nm, Sigma-Aldrich) in 20 mL isopropyl alcohol (IPA) with 66 μL of 5% Sustainion® ionomer solution (XA-9) was prepared and sonicated for at least 2 h. A 5 × 2.5 cm^2 39BB carbon paper (Sigracet) was allowed to dry on a hotplate at 100°C for 1 h to remove any moisture. The ink was then spray coated with periodic weighing on the carbon paper by an airbrush to obtain the desired catalyst loading of 2 mg/cm^2.

Electrochemical measurements: Carbonate electrolysis was performed in a membrane electrode assembly (MEA) cell fixture (Scribner). 5 cm^2 of the spray coated Cu electrode was used as the cathode and 5 cm^2 of Ni mesh was used as the anode. A 3 × 3 cm^2 bipolar membrane (Fumasep FBM) was used as the ion-exchange membrane. 100 mL of the desired K_2CO_3 concentration (Sigma-Aldrich, ACS grade, ≥ 99%) was used as the catholyte and was continuously circulated at flowrates of 39, 65 and 130 mL/min using a peristaltic pump (Masterflex L/S). The anolyte was 200 mL of 1 M KOH circulated at 130 mL/min. N_2 was used as sweep gas to deliver the gaseous CO_2RR products to the GC (SRI 8610C, SRI Instruments). We used an interdigitated flow pattern (IDF) for all measurements unless mentioned otherwise, where a serpentine flow pattern (SF) was used of 3 × 3 cm^2. The gases were detected, and concentrations measured using a thermal conductivity detector (TCD) and flame ionization detector (FID). Liquid products, if any, were measured using a HPLC (Waters Corporation) with an Aminex HPX-87H column (Bio-Rad). Gaseous products were measured for 2 h at 50 mA cm^{-2} and 1 h for all other current densities with the same catholyte. The faradaic efficiency for each CO_2RR product was calculated as $FE = znF/Q$, where z is the number of electrons required to form the product, n is the number of moles of the product detected by GC/HPLC, F is Faraday's constant (98485 C/mol), and Q is the total amount of charge transferred.

Supplementary Notes

Assumptions for the techno-economic model

A production rate of 2 million metric tons per annum (MMT/yr) of ethylene is chosen as the basis. A stream factor of 0.9 (or 90% operational efficiency) was assumed to account for approximately 3-week downtime and the total operational lifetime for the plant was assumed as 20 years with an additional 2 years for construction. The current state-of-the-art capture efficiency of 75%[1] demonstrated in the Carbon Engineering pilot plant and the sizing/costing model developed by Stolaroff et al.[2] are adopted (after adjusting for inflation to 2022 $) for the pessimistic scenario. We assume a modest improvement to 80% for the base case since this is easily achievable by increasing the concentration KOH absorbent to >1 M as shown by calculations from (see **Supplementary Table 9**) and a capture efficiency value of 90% for the optimistic scenario based on the rapid strides and investments in the research and development of this field.[3] We also assume that this would lead to a substantial decrease (25%) in the CCU capital in the future, for the optimistic scenario. The pessimistic scenario for CAPEX in general is assumed to become more expensive than the current state-of-the-art values to account for the uncertainties involved.

The basis for the pessimistic scenario GO membrane permeability and rejection properties is our experimental results shown in **Supplementary Fig. 2**. For the base case scenario, we assume a rejection of 90% which is consistent with commercially available NF membranes (albeit they do not work at highly alkaline conditions). We have demonstrated experimentally that the GO membrane can be fabricated in larger sizes (~4 ft^2) and stable operation for over 1500 hours under at conditions that are more harsh (same pH, higher temperature, very high solids content) than those in this study.[4] Based on this we expect the membrane to have a life of 1 year for the pessimistic scenario, and a value of 3 and 5 years was assumed for the base and pessimistic case scenarios.

We assume a recent state-of-the-art C_2H_4 FE of 31% and 13% for C_2H_5OH as the pessimistic case (**Supplementary Table 1**) at a current density of 0.3 $A.cm^{-2}$.[5] For the base and optimistic cases, we assume 50% and 70% C_2H_4 FE at current densities of 0.5 and 1 $A.cm^{-2}$, which could be attained by extensive R&D for carbonate-fed electrolyzers in the near/foreseeable future.[6,7] These figures are based on the values that has already been demonstrated experimentally for other CO_2 gas-fed electrolyzer architectures.[6,8] Along similar lines, we assume ethanol FE of 20% for the base case and a modest increase to 25% for the optimistic case. It should also be noted that several of the parameters in the electrolyzer are interdependent - current density directly influences the cell voltage, single pass conversion and faradic efficiency. Therefore, we chose the current density values as a specification and the cell voltages are computed using a simple voltametric model as described below in the Supplementary Information pg. 10. This model was developed to have a consistent methodology for the prediction of the cell voltages as a function of current densities and does not involve rigorous calculation to make accurate predictions at larger current densities. For the pessimistic case, the full cell voltage computed by the model is 5.25 V at 300 $mA.cm^{-2}$. For the base case, we assume that improvements to the CO_2RR catalyst and to the BPM would lower ionic resistances (~2.75 $\Omega\ cm^{-2}$) and hence the overpotentials, so we assume that the cell voltage would be one-third lower than the value predicted by the model at a given current density. For the base case, a cell voltage of 4.26 V at 500 $mA.cm^{-2}$, which is 33% lower than the value predicted by the model (6.38 V). Similarly, for the optimistic case we assume that the cell voltage is 40% lower than the value predicted by the model. This is feasible as catalysts with effective Tafel Slope lower than 100 mV/dec and BPM's that can operate almost at par with the thermodynamic requirement, which have been demonstrated experimentally in the literature.[9,10] Lab-scale experiments have extremely low conversions <0.1%, but the weight hourly space velocity (WHSV) in hour $^{-1}$ tends to be $>10^4$ h^{-1}. In large scale reactors, the WHSV can be reduced (which is desired) by increasing the catalyst loading, which in turn increases conversion (see supplementary note in pg. 14). This is desirable over decreasing the flow rate as it might modify the flow patterns of the electrochemical reactor. This will allow increasing the single-pass carbonate conversion per stack (assuming all cells within the stack perform uniformly with the same conversion) from ~0.1% to ~1%, linear relationship for first-order reactions. We assume this for the pessimistic case, and we aim to achieve an overall carbonate conversion of 20% using the stacks-in-series configuration as described in the results section. For the base case, we assume that breakthroughs in electrolyzer design research will allow the development of much favorable local reaction environments that would significantly increase the single pass conversion. Assuming that these modifications enable 5% single-pass conversion per stack, an overall conversion of 35% as a specification for this case was assumed. Similarly, for the optimistic case we assume a per stack single pass conversion of 10% and an overall conversion of 65% as a specification. It should be noted that the single-pass conversion values refer to the amount of carbonates fed into the electrolyzer that are reduced to CO_2RR products, while the overall conversions are specifications which is the total conversion that can be achieved by connecting multiple stacks in series as demonstrated in the "stack-in-series configuration" in the results and discussion section.

We consider the current market prices sourced from various reports in the literature for oxygen, hydrogen, and ethanol after a 15% discount for the pessimistic case, and assume that the price might increase about 20% for the optimistic case. The economic viability of CO_2 electrolysis processes have been shown to have a direct dependence on the electricity prices in majority of the previous works.[11–13] The average industrial electricity price in the US is 0.08 $/kWh based on February 2024 data (https://www.eia.gov/electricity/monthly/epm_table_grapher.php?t=epmt_5_3). To provide a realistic picture of the current-state-of-the-art costs associated with this technology, we assumed an electricity price of 0.08 $/kWh for the pessimistic case. But the large disparity in these prices around the world ranging from 0.5 $/kWh in Europe to 0.02 $/kWh in the middle east and Asian countries should also be considered.[14] In addition to this, the decreasing trends in renewable energy-sourced electricity prices, especially solar and wind energy are observed, and recent studies forecast electricity costs ~0.03 $/kWh are attainable by 2030 and prices are expected to be under 0.02 $/kWh by 2040.[15,16] We therefore assumed a electricity price of 0.02 $/kWh for the base case scenario and 0.015 $/kWh for the optimistic cases being scaled based on a 90% production capacity (stream factor, not capacity factor). We assume capacity factor of 1 in this study implying uninterrupted electricity supply from the grid, for calculation of electricity prices despite the intermittent nature of renewable electricity supply. But, renewable electricity sourced from geothermal or nuclear energy tend to have very high capacity factors (75% and 93% respectively)[11] and works to deploy small modular nuclear reactors (SMR) to supply electricity to chemical plants are in progress which is promising.

The electrode area per cell for the pessimistic case was assumed to be 1.5 m^2 (~16 ft^2), which was based the PEM water electrolyzer designed by NREL (https://www.nrel.gov/docs/fy20osti/79055.pdf), while the value of 3.5 m^2 (~38 ft^2) was based on the Thyssenkrupp PEM water electrolyzer (https://thyssenkrupp-nucera.com/green-hydrogen-solutions/). While we emphasize the non-transferability of learnings from PEM technology to CO_2RR electrolyzers, these numbers serve as an expedient first approximation. For the CHA zeolitic hollow fiber membranes used downstream for gas separation, in house permeation experiments at 1 bar and 25℃ revealed a very high permselectivity of >80 for H_2 over C_2H_4. The membrane also had a high H_2 flux of 0.26 $kg.m^{-2}.h^{-1}$, which is our pessimistic case assumption. The lifetime for this technology has not been well-established yet, therefore we adopt the figures available in the literature for commercialized hollow fibers that are used in membrane bioreactors which is 5.5 – 10 years.[17] Those conditions are much harsher towards membrane materials than the relatively "clean" gas mixtures produced by the electrolyzers. We hence assume a lifetime of 5.5 years for the pessimistic case and a value of 7 years for the optimistic cases. Similarly for the pervaporation membranes we assume an ethanol/water selectivity of 8500[18] for the pessimistic case, which have also been commercialized. Whereas the lifetime of these membranes is expected to be between 3-5 years, and we assume a lifetime of 5 years[19] for the optimistic scenario.

The primary raw materials for the process include KOH, water, air, and steam. While air is assumed to be free of cost, the cost of water of 0.1 $/m^3 was assumed[1] and steam costs were calculated using ASPEN Plus. We assume that all the KOH can be regenerated in the electrolyzer and assume that the total loss of KOH in the entire process <1%. This is because of the very high solubility of KOH in water (121 g/100 mL, ~21 M). The methodology used for calculating the labor costs is summarized in the sample calculations and is based on the equations provided by Turton et al.[20] The replacement costs are unit specific – for the membrane-based units the replacement costs are based on the membrane lifetime and for the electrolyzer the costs are calculated as the sum of replacing the BPM (assumed lifetime 2.5 years)[21], replenishing the catalysts (5% of the GDE costs incurred every year)[22] and replacing the cell compartment (assumed lifetime 7 years)[13].

Concentration of KOH absorbent for the Carbon Capture Unit

(1) For the Direct air capture (DAC) case, Stolaroff et. al.[2] have shown the calculation of CO_2 flux (J_{CO2}) from air into the absorbent and the mass transfer coefficient (k_{MTC}) estimation using suitable correlations in detail. They are calculated as:

$$k_{MTC} = \frac{K_H^\circ}{\gamma_{CO_2}} \sqrt{\frac{D_l^\circ}{\mu^{1.14}} k_r \gamma_{KOH} [OH^-]}$$ **Supplementary Equation (1)**

$$J_{CO_2} = K_H C_0 \sqrt{D_l k_r \{OH^-\}}$$ **Supplementary Equation (2)**

where K_H refers to the Henry's constant or solubility of CO_2 into the KOH absorbent (=0.6), and K_H° refers to the Henry's constant of CO_2 in pure water (infinite dilution), C_0 refers to the concentration of CO_2 in the feed (air, mol/m^3), {OH^-} refers to the activity of OH^- ions (which is primarily a function of the KOH concentration ([OH^-])). D_l° refers to the diffusivity of CO_2 in solution at infinite dilution (=1.88e-9 m^2/s) where as D_l refers to the diffusivity of CO_2 in the KOH absorbent, γ_i refers to the activity coefficient of species 'i' in the solution (=1), μ refers to the viscosity of the solution, k_r refers to the rate constant of the rate-limiting elementary reaction: $CO_2(aq) + OH^- \rightarrow HCO_3^-$, which is assumed to remain constant with temperature (20℃ for all parameters). The CO_2 capture efficiency (η) can be calculated as:

$$\eta = 1 - \exp\left(-\varepsilon\, SSA\, D \frac{k_{MTC}}{V}\right)$$ **Supplementary Equation (3)**

Where ε refers to the packing efficiency of the absorber (=80%), SSA refers to the specific packing area (=200 m^2/m^3), D refers to the packing depth of the absorber (=6.2 m), V refers to the air velocity (=1.6 m/s). Here we neglect the effect of ionic strength and therefore do not adjust the activity of [OH^-], similar to the second approach provided by Stolaroff et al.[2] We calculate the rate constants using the fitted formula for CO_2 – KOH from Pohorecki et al.[23] While the actual values of capture efficiency may be lower than the values shown in **Supplementary Table 9**, they clearly capture the effect of KOH concentration on capture efficiency and hence pH of the outlet stream.

(2) For the capture of CO_2 from a representative flue gas by chemical absorption, Rastegar et al.[24] have provided regression model equations to compute the CO_2 loading (α; mol CO_2 per mol KOH) as a function of temperature (T; °C), pressure (P; bar) and absorbent concentration (C; mol/L) as seen below:

$$CO_2\ \text{Loading} = 1.05023 - 0.004528\,T + 0.061992\,P - 1.05041\,C - 0.000430\,TP + 0.016246\,TC - 0.072635\,PC - 0.000106\,T^2 + 0.003019\,P^2 + 0.382349\,C^2$$

Supplementary Equation (4)

$$\alpha_{CO_2} = \frac{(P_i - P_f)V_g}{RTz} \cdot \frac{1}{n_{KOH}}$$

Supplementary Equation (5)

$$\eta = \frac{(P_i - P_f)}{P_i} \times 100$$

Supplementary Equation (6)

where η refers to CO_2 removal efficiency (%), P_i is the initial pressure (Pa), P_f is the final pressure (Pa), V_g is the volume of flue gas (cm^3), T is the temperature (K), z is the compressibility factor which is considered to be 0.975 (at pressure of 5 bar and temperature of 25°C)[3], n_{KOH} refers to the number of moles of KOH and R is the universal gas constant (cm^3 Pa/K mol). Fixing the P = 2 bar, T = 45°C and 15% CO_2 gas feed, for R_{CO_2}= 65% we require a [KOH] = 0.56 M, similarly for R_{CO_2}=80%, [KOH] = 0.68 M is required. Detailed calculations and correlations used here can be found in Rastegar et al. and Kucka et al.[24,25] It is to be noted that for the Amine-based flue gas capture technology the energy costs are significantly lower, but the captured CO_2 would exist predominantly as carbamates and not as [bi]carbonates, changing the kinetics, performance & modelling of the electrolyzer[26].

Full cell voltage calculation

The full cell voltage for a carbonate-fed BPM system can be calculated as shown below, which is a simplified version of the model developed by Shin et al.[13] by neglecting the mass transport effects. The primary objective for incorporating the voltametric model in this study was not to make accurate cell voltage predictions but only to capture key trends and to have a consistent methodology to calculate the cell voltage as a function of current density for all cases and the sensitivity analysis. We evaluate the contributions of the resistive losses due to various components as a function of current density in addition to the overpotentials associated with the electrodes and the bipolar membrane. The thermodynamic requirement is constant at 1.08 V ($E^0_{OER} - E^0_{CO2RR}$). Since the state-of-the-art electrolyzer depicted by Lee et al.[5] includes a pH modulating layer (IP), we factor in its contribution. Even in its absence a pH gradient will exist in a BPM electrolyzer due to the large difference between the pH in the cathode and anode compartments. The Nernstian overpotential was calculated assuming a pH difference of 8. The resistive contributions – ionic and contact resistances – of various parts including the bipolar plate, GDL, IP and the corresponding calculations are summarized in the **Supplementary Table 10**. It is to be noted that the pH modulating layers are extremely porous (porosity >85%) hence we assume that this could be approximated as an electrolyte gap. The concentration of salts across the IP decreases drastically as shown by Lee et al.[5] So, we assume a conservative value (50 Ω cm) for the resistivity, which corresponds to a K_2CO_3 concentration of 1 wt%.[27] For the BPM, we assume an ionic resistance of 4 $\Omega\,cm^{-2}$, which is the average ionic resistance in Fumasep BPM between 0.1-1 A cm^{-2} in addition to the thermodynamic requirement of 0.83 V for the WDR at the BPM interface. Voltage across different sections of the electrochemical cell are calculated as $V = IR = (J_{tot}A_{cell}) \times (\rho_{section}d_{section})$. The overpotentials for the electrodes were computed based on a generalized Tafel slope (TS) to reduce complexity, which is assumed to be independent of the variations that might arise due to the variations in local pH or hydration or the reaction's rate-determining step.[13] A reference overpotential (η_{ref}) and current density (j_{ref}) corresponding to this Tafel characteristic were obtained from the literature and the overpotential was calculated as: $\eta = \eta_{ref} +$

$TS \times \log(\frac{j}{j_{ref}})$. For the anode, we assume the same values as Shin et al.[13] for a Ni-mesh and 1 M KOH anolyte, where the reference overpotential was 247 mV at a current density of 10 mA.cm^{-2} and a Tafel Slope of 41 mV.dec^{-1}. For the cathode, we assume a reference overpotential of 920 mV at 200 mA.cm^{-2}.[5] Due to the substantial variation in the Tafel slopes for the cathode seen in literature, we decided to fit the fit value to the full cell voltage presented by Lee et al.[5] and was equal to 1037 mV/dec^{-1}. **Supplementary Figure 11** shows how the selected parameters for the BPM and other parameters used to compute the cell voltages as a function of current density (100 – 1000 mA.cm^{-2}) agree with the literature data. It should be noted that the predictions at higher current densities would be underestimated by the model primarily because – (1) a linear polarization curve for the BPM is assumed and (2) the mass transport effects become significant when current densities approach the limiting current densities. **Supplementary Figure 12** depicts effect of modifying cathodic Tafel slope and the BPM ionic resistance. It can be seen that in order to achieve the cell voltages desired in the optimistic case, effective TS in the range of 100 mV/dec and ionic resistance of BPM < 2.5 Ω cm^{-2} are required. It should be noted that since we assume 150 cells with the stack are connected in series electrical configuration, the voltage applied to the stack should be 150 × 5.26 V = 789 V (for the pessimistic case).

Heat balance across the electrolyzer cell & stack

The overall energy balance for an electrolyzer stack of 150 cells each of 3.5 m^2 electrode area *(base case assumption)*, assuming an open system can be[28]

$$mC_p \frac{dT}{dt} = \sum_r \dot{n}_r H_{in,r} - \sum_p \dot{n}_p H_{out,p} + W_{elec} - \dot{Q}_{heat,gen} - \sum_j r_i \Delta H_{rxn,j}$$ **Supplementary Equation (7)**

where m is the mass of the system [constant, kg], C_p is the specific heat capacity of the system [J kg^{-1} K^{-1}], H_{in} and H_{out} refer to the enthalpy of the inlet and outlet streams respectively [J mol^{-1}], $\dot{n}_r$ and $\dot{n}_p$ are the molar flowrates of the inlet and outlet streams [mol s^{-1}], $\dot{Q}_{heat,gen}$ is the amount of energy lost as heat [W], W is the power added to the stack as I×V×n_{cell} [W], r_j is the rate of reactions for a given specie 'i' and the enthalpy of full reactions (j) per mole of specie 'i' is given by $\Delta H_{rxn,j}$. Since we are operating at steady state, the LHS of the above equation is zero. The molar enthalpy change for the formation of hydrogen, ethylene and ethanol are 0 kJ/mol, -64 kJ/mol, -86.7 kJ/mol respectively[29]. The corresponding rate of formation of these species per stack, obtained from the Aspen Plus simulation, are 4.56 mol/s, 1.27 mol/s and 0.52 mol/s respectively for the base case. Also, since we expect the outlet to also be at 298 K, the inlet and outlet enthalpies are equal to zero. For the base and optimistic cases, we assume that the ionic resistance of the BPM to be 2.5 and 1 Ω cm^{-2} respectively, which is critical to achieve the desired >30% reduction in cell voltages. This would yield $Q_{heat,\,gen} \approx 323$ W cm^{-2} for the entire stack. The resistive losses of energy as a result of joule heating can be calculated as $J_{tot} \times \Delta V \times n_{cell}$. The heat generated due to the resistances of the bipolar plate, the GDL, and the interposer are 0.0005 W/cm^2, 0.4875 W/cm^2, 25.3125 W/cm^2 respectively for the entire stack for the base case. The contact resistance between the bi-polar plate-GDL interface and the interposer-catalyst interface each generates 0.1875 W/cm^2 of heat. The heat generated due to the ionic resistance is 103.13 W/cm^2. Therefore, the total heat generated per stack would be ~193 W/cm^2 using the energy balance equation shown above out of which ~129 W/cm^2 are resistive losses. For the optimistic cases total heat generated per stack would be >900 W/cm^2, the substantial amount of heat generated clearly indicates the need for integrated cooling systems for large scale electrolyzers.

Fitting of Experimental Residence Time Distributions

We use the open vessel dispersion model to fit experimental residence time distributions (RTDs)[30]:

$$E(t) = \frac{1}{\sqrt{4\pi t\tau(\mathbf{D}/uL)}} \exp\left[-\frac{(\tau - t)^2}{4t\tau(\mathbf{D}/uL)}\right]$$

Supplementary Equation (8)

Where E(t) represents the RTD of the fluid (s^{-1}) after t seconds since the introduction of the tracer, τ is the mean residence time (s), and **D** refers to the axial diffusion coefficient which characterizes the degree of back mixing, and is calculated as:

$$\mathbf{D} = \frac{u^2 d_c^2}{192\mathfrak{D}} + \mathfrak{D}$$ **Supplementary Equation (9)**

Here, *u* is the average velocity of the fluid in the bulk flow direction calculated as (ϑ.L/$V_{reactor}$, where ϑ is the volumetric flow rate of the feed and $V_{reactor}$ is the volume of the reactor), d_c is the width of the flow channel, and $\mathfrak{D}$ is the diffusivity of carbonate ions in water.

For the parallel CFD study, the Peclet number (Pe) is calculated as:

$$\mathbf{Pe^{-1}} = \frac{\mathfrak{D}}{\overline{V_{CF}}.\,L_{GDL}}$$ **Supplementary Equation (10)**

Where $\overline{V_{CF}}$ refers to the average cross-flow velocity of the fluid across the gas diffusion electrode (GDE) as shown in **Supplementary Figure 18** and 'L' refers to the length of the GDE (350 µm). The τ_{exp} or simply $\bar{t}$ value is measured as:

$$\boldsymbol{\tau_{exp}} = \bar{t} = \frac{\sum_i t_i C_i \Delta t_i}{\sum_i C_i \Delta t_i}$$ **Supplementary Equation (11)**

Where *i* refers to the i^{th} observation, C_i is the concentration of carbonates(tracer) at time t_i and Δt_i is the time difference between two successive observations.

Stacks in series configuration for a CSTR

For these calculations, we assume that the electrolysis cell operates at a high enough flow rates so it can be approximated as a CSTR as seen in the RTD experiments. The cells within each stack are in parallel flow configuration and the cells in the subsequent stacks are connected in series and are of equal volume. We assume an isotropic distribution of current and feed for the electrode areas per cell assumed in this study (3.5 - 10 m^2). We assume an incompressible flow and that the overall CO_2RR follows 1^{st} order kinetics. Let 'n' be the number stacks connected in series, each of volume 'V' and a residence time of τ, 'k' be the effective first order rate constant, C_0 is the concentration of the carbonates to the first stack and the C_i be the concentration at the i^{th} stack, rate of conversion of the reactant 'A' is r_A, molar flow rate of feed F_0 and X_n refers to the total conversion after the n^{th} stack. The performance equation of a CSTR can be written as:

$$\boldsymbol{\tau} = \frac{C_0 V}{F_0} = \frac{\mathrm{C_0 X_A}}{-r_A}$$ **Supplementary Equation (12)**

$$X_i = 1 - \frac{C_i}{C_0}$$ **Supplementary Equation (13)**

$$\tau_i = \frac{C_{i-1} - C_i}{kC_i}$$ **Supplementary Equation (14)**

Residence time of all CSTRs are equal, which implies

$$\frac{C_0}{C_n} = \frac{C_0}{C_1} \times \frac{C_1}{C_2} \times \dots \frac{C_{n-1}}{C_n} = (1 + k\tau)^n$$ **Supplementary Equation (15)**

$$For\ a\ single\ stack, \qquad r' = k\tau = \frac{X_1}{1 - X_1}$$ **Supplementary Equation (16)**

When n identical stacks are in series,

$$X_n = \frac{C_0 - C_n}{C_0} = 1 - \frac{1}{(1 + r')^n}$$ **Supplementary Equation (17)**

For example, when a single stack conversion is 1%, then r' = 0.0101 and when 22 stacks are connected in series $X_{22} = 1 - \frac{1}{(1+0.0101)^{22}} = 19.83\%$

For a first order reaction, decreasing the space velocity (SV = $1/\tau$) by 1 order of magnitude will increase the conversion by 10x as seen in **Supplementary Equation 12**.

Pressure Drop Across a Electrolyzer Stack

Pressure drop across the flow field assuming parallel channels can be approximated using the incompressible flow equation for pipes[31]:

$$\Delta P = f \frac{L}{D_H} \rho \frac{v_{avg}^2}{2} + \sum K_L \rho \frac{v_{avg}^2}{2}$$ **Supplementary Equation (18)**

Where, f is the friction factor, L is the channel length (m), D_H is the hydraulic diameter (m), ρ is the fluid density (kg m^{-3}), v_{avg} is the average velocity (m s^{-1}), K_L is the local resistance to flow. For the base case, we assume a square cell with active area of 3.5 m^2, the dimensions can be assumed to be 1.87 m × 1.87 m with each stack having 150 cells each (N_{cell}). Assuming that the dimensions of the scaled-up electrolyzer stack is proportional to our lab scale setup, a width of the channel ≈0.02 m and a depth of ≈0.01 m would be required assuming there are about 30 channels (N_{ch}). The hydraulic diameter can hence be calculated as

$$D_H = \frac{2(width)(depth)}{(width + depth)} \approx 0.013\ m$$ **Supplementary Equation (19)**

For our base case we would require 2221 stacks (see sample calculation below), and from **Fig. 3b** in the main text we know that 8 stacks needs to be connected in series to achieve the desired conversion (35%). Therefore, the volumetric flow rate per stack ≈16.7 m^3 h^{-1} (Q_{stack}, from Aspen simulation) with the density of ρ =1283 kg m^{-3} and a viscosity of ~1 mPa s at 20°C. The velocity of the flow channel at the entrance of the cell is calculated as

$$v = \frac{Q_{stack}}{N_{cell} N_{ch} A_{ch}} = \frac{\left(\frac{16.7 \times 10^6}{3600} \frac{cm^3}{s}\right)}{150 \times 30 \times 2\ cm \times 1\ cm} = 0.515\ cm\ s^{-1}$$ **Supplementary Equation (S20)**

The Reynolds number at the channel entrance is

$$Re = \frac{D_H v \rho}{\mu} = \frac{0.013 \times 0.00515 \times 1283}{0.001} = 86$$ **Supplementary Equation (S21)**

To calculate the friction factor "f" for rectangular channels

$$Re.f = 55 + 41.5 \exp\left(\frac{-3.4}{width/depth}\right) = 63$$ **Supplementary Equation (S22)**

Therefore, the friction factor f is 0.728, assuming sixty 90° bends and the pressure drop is calculated as

$$\Delta P = 0.728 \frac{1.87\ m}{0.013\ m} 1283 \frac{kg}{m^3} \frac{(0.0051\ m/s)^2}{2} + (0.728 \times 60 \times 30) 1283 \frac{kg}{m^3} \frac{(0.0051\ m/s)^2}{2}$$
$$= 23.61\ Pa$$

To replicate similar flow conditions as observed in our lab scale setup, a channel width ≈0.005 m and a depth of ≈0.001 m would be required assuming there are about 10 channels (N_{ch}). In this case, the velocity through the channel would be 61.9 cm s^{-1} with a Reynold's number of 1324, friction factor of 0.057 and a pressure drop of 0.24 bar per stack. In this case, it would be feasible to connect only a few stacks in series before the pumping costs overtake the revenue possibly generated by increased overall conversion by connecting multiple stacks in series. The inferences from large scale PEM electrolyzers are not transferrable here as discussed in the main text. The pressure drop must be carefully studied, and reasonable constraints need to be developed that agree with experimental observations. Therefore, it is critical that the flow channels are carefully designed to ensure that the pressure drop is kept to a minimum, while maintaining a uniform distribution across the stack and maintaining well-mixed flow characteristics within the cell.

Sample Calculation of MSP under the <u>Base Case</u> Assumptions

1. *<u>CO_2 required to be captured to produce 2 MMT/yr of ethylene</u>*

The estimation of the total CO_2 required to be captured is computed as follows. The parameters used in this sample calculation are summarized in **Supplementary Table 1**. The methodology for the calculation for the economic equations and basis for the assumptions are summarized in the methods section of the manuscript. First, we convert the ethylene production rate to a molar basis:

$$2000000\ \frac{ton\ C_2H_4}{yr} * 1000\frac{kg}{ton} * \frac{1}{28}\frac{kmol}{kg} * \frac{1}{365}\frac{yr}{day} * \frac{1}{86400}\frac{day}{s} = 2.265\ \frac{kmol\ C_2H_4}{s}$$

When the plant runs with a stream factor (or operational efficiency) of 0.9, then rate of ethylene production would be 2.517 kmol C_2H_4/s

Since we recover all the unreacted carbonates in the cathode of the electrolyzer, the single-pass conversion of the carbonates does not affect the CO_2 to be captured per pass. We assume that ethylene and ethanol are the only major products and assume FE_{C2H4} = 50%, FE_{C2H5OH} = 20% (see **Supplementary Table 1**). This yields a molar ethylene selectivity of 0.71 (calculated as FE_{C2H4} /(FE_{C2H5OH} + FE_{C2H4})) and since 2 moles of CO_2 are required to produce 1 mole of ethylene -

$$2.517\ \frac{kmol}{s} * \frac{1}{0.714} * 2\frac{mol\ C_2H_4}{mol\ CO_3^{2-}} = 7.05\ \frac{kmol\ CO_2}{s}$$

At 80% capture efficiency in the CCU, all CO_2 captured are converted into carbonates and at 90% carbonate rejection in the upstream GO membrane, finally gives

$$6.34\ \frac{kmol}{s} * \frac{1}{0.8} * \frac{1}{0.9} = \mathbf{9.79}\ \frac{kmol\ CO_2}{s}$$

2. *Costs associated with the Carbon Capture Unit*

Since each mole of KOH reacts with 2 moles of CO_2, 9.79 * 2 = 19.58 kmol/s of absorbent is required to capture the desired amount of CO_2. Assuming the CO_2 uptake is ~0.13 mol CO_2/mol KOH[32]. The capture rate required to achieve 80% capture efficiency is 25.01 t-CO_2.m^{-2}.yr^{-1} based on equations discussed above with k_{MTC} =0.0024. The area of absorber required assuming a stream factor 0.9

$$9.787\ \frac{kmol}{s} * 44\frac{kg}{kmol} * \frac{1}{1000}\frac{ton}{kg} * \frac{1}{25.01}\ \frac{m^2.yr}{ton} * 365\frac{day}{yr} * 86400\frac{s}{day} * 0.9 = 488{,}693\ m^2$$

The cost per frontal area of the CCU is calculated from Holmes et al.[32] using the early plant cost estimation in 2022 dollars. The fixed capital investment cost associated with the CCU assuming a bare module factor of 1.5 and an additional 15% for contingency and 3% for fees (assumed standard for all units)[66]

$$4668.4\ \frac{USD}{m^2} * (488{,}693\ m^2)^{0.85} * 1.18 * 1.5 = USD\ 565{,}959{,}201$$

The minor difference in the RHS and LHS of all equations is attributed to rounding of the significant digits. The repair and replacement costs for the CCU and all the associated units are considered to be 5% of the fixed capital investment incurred every year. The raw material cost associated with the CCU assuming water cost of 0.1 $/$m^3$ with a loss of 2 mol H_2O/ mol CO_2 (which corresponds to a solution molarity of ~3 M, temperature of 20°C and a humidity of 55%[1]) is $USD\ 146{,}299{,}145$

The cost of utilities for the fans and pumps associated with the CCU at a pressure drop of 125.44 Pa [Calculated as $D*7.4*V^{2.14}$, with an absorber depth of 6.2 m (=D), air velocity of 1.6 m/s (=V)], 60% fan efficiency[32], SF = 0.9.

$$0.9 * 365\,\frac{day}{yr} * 24\,\frac{hr}{day} * \frac{1}{0.6} * 125.44\,\frac{kg}{m.s^2} * 1.6\,\frac{m}{s} * 488{,}693\ m^2 * \frac{1}{1000}\frac{Wh}{kWh} * 0.02\,\frac{USD}{kWh}$$

$$= USD\ 25{,}775{,}139$$

The levelized cost of CO_2 defined as cost per ton of CO_2 produced through this process and can be calculated as CI*CRF/SF, where CRF is the cost recovery factor and CI is the cost intensity defined as capital cost per unit capacity -

$$Levelized\ cost|_{CRF} = \frac{FCILCCU \times CRF + COM_D CCU}{capture\ rate \times CCUarea \times SF}$$ **Supplementary Equation (S23)**

At CRF=7.5%, Levelized cost of CO_2 is 35.5 USD/t-CO_2 and at CRF = 12.5%, is **38.1 USD/t-CO_2.**

3. <u>Costs associated with the Upstream GO Concentration Unit</u>

The flux of carbonates rejected by the membrane concentrator unit at 2 M concentration is calculated based on the total carbonate feed processed 27.97 (9.79* 2.85 (recycle ratio, from Aspen Plus))

$$27.97\ \frac{kmol}{s} * 0.8 * 0.9 = 20.14\ \frac{kmol}{s}$$

From this, assuming a density of 1 g/cc for the retentate and estimated flow rate of water from CCU of 87,046 L/s, the permeating flux of water can be calculated as

$$87{,}046\ \frac{L}{s} - \ 20.14\ \ \frac{kmol}{s} * 1000\,\frac{mol}{kmol} * \frac{1}{2.5}\frac{L}{mol} = 76{,}979\,\frac{L}{s}$$

The required area of the membrane at 50 bar transmembrane pressure and assumed membrane permeance of 2.8 $Lm^{-2}bar^{-1}h^{-1}$ (see **Supplementary Table 1**)is calculated as

$$76{,}979\ \frac{L}{s} * \frac{1}{2.8}\frac{m^2.bar.h}{L} * \frac{1}{50}\frac{1}{bar} * 3600\,\frac{s}{h} = 1{,}979{,}469\ m^2$$

Similarly, the calculations are performed for the recovery unit, with the only change that the concentration of the retentate stream from the recovery unit is $\approx 0.5\ M$. The permeating flux is 62,886 $\frac{L}{s}$ and the required membrane area for the recovery membrane unit is 1,617,072 m^2. The replacement costs were assumed to be equal to the cost of replacing the membrane at end of lifetime but split to incur every year. The capital cost of the GO membrane including the recovery membrane unit (total area = 3,596,541 m^2) at 200 $\frac{USD}{m^2}$, with a membrane life of 3 years is calculated as

$$200\ \frac{USD}{m^2} * (3{,}596{,}541)^{0.85} * \left(\frac{20\ years}{3\ years}\right) * 1.5 * 1.18 = \ USD\ 881{,}889{,}894$$

The utilities cost for the membrane unit including the pumping costs (concentration + recovery units)

$$(87{,}046 + 76{,}979)\ \frac{L}{s} * \frac{1}{1000}\frac{m^3}{L} * 3600\frac{s}{h} * 1000\frac{kg}{m^3} * 9.81\frac{m}{s^2} * 0.9 * 365\frac{day}{yr} * 24\frac{hr}{day} * \frac{1}{3.6 \times 10^6} * 0.02\frac{USD}{kWh} = USD\ 253{,}721$$

4. *Costs associated with the Electrolyzer Unit*

Cell voltage associated with the BPM MEA can be calculated as function of total current density based on the model described earlier. Since we assume a total current density of 500 $mA.cm^{-2}$, the cell voltage predicted by the model after 33% reduction is $4.26\ V$. The detailed basis for the assumptions are summarized in Supplementary Information pg. 4. The total current required for the production of the desired amount of ethylene with 50% FE is calculated as

$$2.51\ \frac{kmol}{s} * 12e^{-} * 96{,}485\ \frac{C}{mol} * 1000\frac{mol}{kmol} * \left(\frac{1}{0.5}\right) = 5{,}827{,}655{,}770\ A$$

This current would produce a total of 2 MMT/yr of ethylene, 1.31 MMT/yr of ethanol, 0.52 MMT/yr of hydrogen in the cathode and 15.23 MMT/yr of oxygen in the anode. The electrode area that would be required can be calculated as

$$5{,}827{,}655{,}770\ A * \frac{1}{0.5}\frac{cm^2}{A} * \frac{1}{10000}\frac{m^2}{cm^2} = 1{,}165{,}531\ m^2$$

The electricity costs associated with the electrolyzer unit is

$$5{,}827{,}655{,}770\ A * 4.26V * \frac{1}{1000}\frac{Wh}{kWh} * 0.02\frac{USD}{kWh} * 0.9 * 365\frac{day}{yr} * 24\frac{hr}{day} = USD\ 3{,}914{,}534{,}287$$

For costing the electrolyzer, we adopt the cost equations for the different components of the electrolyzer whose basis is well-explained by Badgett et al.[22] The cost associated with a 0.0961 m^2 BPM-based electrolyzer is considered with a flowing catholyte layer to account for the cooling plates, whose costs is 1035 USD (10,771/m^2 * 0.0961 m^2, data from reference with no scaling in this range). The breakdown of costs for the components - the cooling plate (catholyte flow plate), the bipolar membrane, frame of the electrolyzer, bipolar plates, and stack assembly of the 0.0961 m^2 electrolyzer are - 194.7, 83.6, 257.7, 29.4, and 239.1 USD respectively. The area of the cathode gas diffusion layer and the anode gas diffusion layers are smaller (=0.0676 m^2) and the associated costs are $ 95.2 and $135.5.

The stack configurations we assume involve a stack with a total of 150 cells per stack and an electrode area per cell of 3.5 m^2, leading to 525 m^2 of electrode area per stack. This would require a total of 1,165,531 m^2 /525 m^2 = 2221 stacks. The number of cells per stack is included within the scaling equation since larger quantity of production would decrease per capita costs of production. The cost associated with a particular component can be calculated

$$\begin{pmatrix} Component \\ cost\ of\ the \\ 0.0961\ m^2 \\ electrolyzer \end{pmatrix} * \left(\frac{Area\ per\ stack}{0.0961}\right)^{0.85} * \begin{pmatrix} Number \\ of\ stacks \\ required \end{pmatrix} = USD\ 3.723 \times 10^9$$

The capital cost associated with the electrolyzer unit is calculated as

$$USD\ 3.723 \times 10^9 * 1.5 * 1.18 = USD\ 6{,}589{,}153{,}281$$

In addition to this, replacement costs of electrolyzer includes replacement of the MEA - BPM every 2.5 years, 5% of catalyst cost every year and cell compartment every 7 years[13]. These costs translate to ~5% of the electrolyzer CAPEX on a per year basis. It is to be noted that when the electrolyzer cells are connected in series electric configuration, the total cell voltage required by the stack would be $V_{cell} \times n_{cell}$. We require highly efficient step-up transformers to achieve the desired voltage. To simplify the calculations, we neglect the losses in power and the computations associated with the voltage modifications.

5. <u>Downstream separation units</u>

(i) Zeolitic hollow fiber membrane

The mass flow rates of hydrogen gas in the binary cathode gas is $5.91 \times 10^4 \frac{kg}{hr}$ and for ethylene is $2.28 \times 10^5 \frac{kg}{hr}$. The mole fraction can be calculated as -

$$\frac{5.91 \times 10^4 \frac{kg}{hr} \Big/ 2\frac{kg}{kmol}}{\left(5.91 \times 10^4 \frac{kg}{hr} \Big/ 2\frac{kg}{kmol} + 2.28 \times 10^5 \frac{kg}{hr} \Big/ 28\frac{kg}{kmol}\right)} = 0.784$$

The area of the hollow fiber membrane that would be required to achieve the desired separation (≈ 99.9%) between H_2 and C_2H_4 at 2 bar feed pressure can be calculated as

$$8.22\ \frac{kmol\ H_2}{s} * 1000\frac{mol}{kmol} * \frac{1}{1100 \times 3.35 \times 10^{-10}}\frac{Pa.m^2.s}{mol} * \frac{1}{200{,}000}\frac{1}{Pa} * \frac{1}{0.784} = 142{,}199\ m^2$$

The capital cost for the membrane separation unit is calculated similar to the other units and 5% of the capital cost is considered as the replacement costs incurred every year (not shown here).

$$20\ \frac{USD}{m^2} * \left(\frac{142{,}199\ m^2}{1\ m^2}\right)^{0.85} * \left(\frac{20\ years}{6\ years}\right) * 1.5 * 1.18 = \ USD\ 3{,}537{,}975$$

The utilities cost associated with the membrane unit

$$5.91 \times 10^4 \frac{kg}{hr} * 9.81\frac{m}{s^2} * 0.9 * 365\frac{day}{yr} * 24\frac{hr}{day} * 0.02\frac{USD}{kWh} = USD\ 203{,}319{,}257$$

(ii) Pervaporation unit (for the optimistic case only)

The flow rate of water to the pervaporation unit is 6698 kg/hr. The area of the zeolite mixed-matrix membrane required to carry out the pervaporation is 80,294 m^2. The capital cost for the membrane separation unit and similar to the other units, cost incurred to replace membranes at the end of their lifetime is considered as the replacement costs split to incur every year.

$$60\ \frac{USD}{m^2} * \left(\frac{96556\ m^2}{1\ m^2}\right)^{0.85} * \left(\frac{20\ years}{5\ years}\right) * 1.5 * 1.18 = \ USD\ 7{,}332{,}412$$

The utilities cost associated with the pervaporation membrane unit

$$6.697 \times 10^3 \frac{kg}{hr} * 9.81 \frac{m}{s^2} * 0.9 * 365 \frac{day}{yr} * 24 \frac{hr}{day} * 0.02 \frac{USD}{kWh} = USD\ 8{,}633{,}801$$

6. *Balance of Plant (BoP)*

The Balance of Plant (BoP) comprises of several components including the Human Machine Interface (HMI) and the associated instrumentation, controls, cooling water supply to maintain the electrolyzer temperature, piping, valves, pumps, safety requirements, the AC to DC rectifier, transformers, heat exchangers, gas liquid separators and evaporators. BoP cost includes all the auxiliary units associated with the process and not the electrolyzer alone. We approximate the BoP costs as 35% of the capital cost of the electrolyzer unit. This could act as an over-estimator for the BoP costs, as seen in the calculations shown in **Supplementary Table 7.**

7. *Process economics*

The number of operating labor required is calculated according to equation 8.3 from Turton et al.[20], for a process with 'P' number of processing steps involving particulate solids (= 0) and 'N' of processing steps not involving particulate solids (= 8 unit processes + (2221/22) electrolyzer stacks, 22 in series)

$$[6.29 + 31.7 * (0)^2 + 0.23 * (8 + 101)]^{0.5} = 5.59$$

The cost of operating labor as 245 shifts per operator (49 weeks per year at 5 shifts a week, assuming 3 weeks of leave per operator), since the plant has to operate 365 days with 3 shifts a day (regardless of the operational efficiency) we require 1095 operating shifts per year: ~4.5 operators per required operator for the plant (1095 operating shifts per year/ 245 shifts per operator); per operator cost is 28.01 USD/hr (US South), for 365 days a year with 0.9 SF (https://www.bls.gov/regions/southwest/news-release/employercostsforemployeecompensation_regions.htm)

$$5.59 * 28.01 \frac{USD}{hr} * (4.5 * 365 * 24) \frac{hr}{yr} = USD\ 6{,}182{,}232$$

The working capital (WC), defined as the financing required for the process initialization and its operation for the initial months as is calculated as $A.C_{RM} + B.C_{FI} + C.C_{OL}$; wherein the coefficients A, B, and C are all equal to 0.1, C_{RM} is cost of raw materials, C_{FCIL} refers to the fixed capital investment and C_{OL} is the cost of labor for the entire plant:

$$0.1 * (USD\ 149{,}406{,}536)_{C_{RM}} + 0.1 * (USD\ 12{,}201{,}581{,}749)_{C_{FI}} + 0.1 * (USD\ 6{,}182{,}232)_{C_{OL}} = USD\ 1{,}235{,}655{,}229$$

The annual cost of manufacturing is computed as $C_M = 0.18C_{FI} + 2.73C_{OL} + 1.23(C_{UT} + C_{WT} + C_{RR} + C_{RM})$. Here C_{UT}, C_{WT} and C_{RM} refer to the costs of utilities, waste treatment, and raw materials respectively.

$$0.18 * (USD\ 12{,}201{,}581{,}749)_{C_{FI}} + 2.73 * (USD\ 6{,}182{,}232)_{C_{OL}} + 1.23 * \left((USD\ 4{,}414{,}245{,}234)_{C_{UT}} + (0)_{C_{WT}} + (USD\ 149{,}406{,}536)_{C_{RM}} + (USD\ 637{,}388{,}431)_{C_{RR}}\right) = USD\ 8{,}608{,}920{,}830$$

The revenue from all products except ethylene are calculated using the values summarized in **Supplementary Table 1** and the minimum selling price (MSP) in USD/kg is calculated as the value for which the Net Present Value is equal to zero at end of the plant life (20 years + 2 years construction) and this was found to be **2.66 USD/kg** of ethylene for the base case. Sample stream table for the flue gas capture scenario is shown in **Supplementary Table 8.**

Scope 1 and scope 2 CO_2 emissions for the process

We use the USEPA's "Simplified GHG Emissions Calculator" for all the calculations and sourcing mission factor values for the 2 MMT/yr CO_2RR ethylene plant corresponding to the optimistic case (https://www.epa.gov/climateleadership/simplified-ghg-emissions-calculator). The primary sources of SCOPE 1 emissions would be from the steam generated in the stationary combustion units for the heating utilities. We assume natural gas as the fuel source to generate steam with an 80% thermal efficiency leads to a CO_2 efficiency factor 66.33 kg/MMBTU. For electricity, when grid electricity is used assuming emission factors corresponding to "ECRT" region [868.6 lb/MWh], where the majority of the chemical industries are concentrated in the US. This can be offset when coupled with renewable sources of electricity, the emission scales down by 8x [~95 lb/MWh]. The mobile sources of CO_2 can be negated by the use of electric or low emitting CNG-run vehicles, and their contribution is insignificant compared to the other sources. The highly flammable nature of the product gases requires large fire suppressants, we assume emission-free substances like HCFCs and inert gases can be employed instead of CO_2. The summary of the calculations is shown in **Supplementary Table 12**.

AEM vs BPM Minimum Selling Price (MSP) Comparison

The upstream concentrator unit is eliminated for the AEM based CO_2 gas-fed electrolyzers case. But for production of gaseous CO_2, the regeneration loop will be required in the CCU, which increases the costs associated with the CCU as shown in **Fig. 4c** in the main text. We assume that 50% of the CO_2 crosses over to the anode through the AEM as carbonates (similar to the assumptions made in previous studies[33,34]) and oxidizes back to CO_2 in the anode which necessitates dedicated separation units for the anode gas separation. We consider a pressure swing adsorption module to carry out this separation and the costs associated with this unit are calculated as shown by Jouny et al.[12] The full cell voltage required for AEM based gas-fed CO_2RR is much lower than the BPM-based carbonate-fed electrolyzers being discussed in this study because of the additional overpotential required for WDR in BPM's.[35] We utilize the voltametric model developed by Shin et al. to calculate the full cell voltages for both the AEM and BPM based cases.[13] The alkaline anolyte is consumed over time in AEM-based systems due to its consumption to form O_2 in the anode and its diffusion to the cathode where it participates in CO_2RR. Recovery of these ions is very energy intensive (detailed calculations performed using the methodology described by Blommaert et al.[34]) and counterbalances the cost reduction achieved by the reduction of cell potential in AEM based systems as seen in **Fig. 4c** in the main text. The alkaline cathodic reaction environment created by the AEM favors CO_2RR over HER, unlike in the case of BPM, where the reaction environment is acidic. This requires additional layers/ coatings to modulate the pH between the PEM and the catalyst layer, but these have negligible impact on the process economics.[35,36] We removed the costs of including these additional layers from the electrolyzer costs and replacing the costs of BPM with the cost of the AEM, whose values are based on the work by Badgett et al.[22] As reported by Vass et al.[37] over 45% of the experimental studies utilize IrO_2 as the catalyst for the anode, whose costs are also calculated based on the work by Badgett et al.[22] For the CEM, we adapt the costs reported by Badgett et al. for the membrane and IrO_2, while the cell voltage is calculated by using the voltametric model presented before, with the contribution of CEM modified to no longer include the WDR overpotential and a negligible ionic resistance of 0.1 Ω cm^{-2}.

Ethanol Recovery from the Liquid Phase Cathode Products Stream

The concentration of ethanol in the cathode product stream is very dilute (~0.5% by weight, **Supplementary Tables 3-5**). This makes its separation very cost intensive and difficult. The two viable options for achieving this separation are through adsorption and distillation-pervaporation.[38] But we consider the latter option because it has been well-studied and industrially implemented to separate such dilute solutions.[38,39] **Supplementary Fig. 1b** demonstrates the Aspen Plus simulation for the cathode liquid separation using the above-mentioned methodology. The two distillation columns are used to recover and purify ethanol to >80 wt% (an arbitrary assumption, to bring it close to the azeotropic composition). The presence of salts affects the ethanol vapor-liquid equilibrium and improves the separation of ethanol due to the "salting-out" effect. It should be noted that CO_2 evolution is not possible in this unit – the thermal decomposition of K_2CO_3 is negligible below 900°C[40] and the highly alkaline conditions in the stream entering the distillation column shift the equilibrium towards carbonates[41]. We used the well-known E-NRTL thermodynamic model available within ASPEN+ to simulate the distillation columns[42–44]. Since the salts are completely removed after the first column, their effect is limited to differences in sensible heat, exclusive to the first column. The first column does not contain a reboiler due to high salt concentrations, to circumvent any issues related to scaling or corrosion. Instead, direct steam is used to recover 99% of the ethanol in this column. We further purify the ethanol stream using a pervaporation membrane unit based on zeolitic membranes, with a water-over-ethanol selectivity of ~10,000[45] and high water flux - assumed as 1.5 $kg.m^{-2}h^{-1}$ at ethanol concentrations >95 wt% and separation factors of ~800 and water flux ~1 $kg.m^{-2}h^{-1}$ at ethanol concentration of 80 wt%[46–48]. We set the desired ethanol outlet concentration as a design specification within the definition of the second RadFrac distillation block in ASPEN+, to achieve which reflux ratio and boil up ratio are varied. 80 wt% was iteratively found to be an optimal outlet concentration for column 2 distillate in terms of energy, capital required for separation and pervaporation-dehydration performance. Due to the energy intensive nature of distillation-pervaporation, a minimum FE of ethanol is required to make this separation economically viable as shown in **Supplementary Fig. 19**.

Using the distillation-pervaporation system is also profitable for the base case scenario, though this is not adopted to maximize profits. But recovering the ethanol in the pessimistic scenario is uneconomical, as the cost of purifying ethanol far exceeds the revenue that could be generated by selling the ethanol (see **Supplementary Table 13**). To address this, we employ a multi-effect vacuum evaporation unit followed by a distillation column scheme as shown in **Supplementary Fig. 1c**, to recover ethanol at lower concentrations <35%[49]. The capital costs, operating costs and utility costs were calculated using Aspen Process Economic Analyzer assuming an electricity cost of 0.02 USD/kWh.

Supplementary Tables

Table 1. Literature review for various techno-economic analyses for CO_2RR technologies

Reference	Year published	Full-process simulations	CCU	CO_2 source	CO_2 capture absorbent	Electrolyzer feed	Electrolyzer design, sizing and costing	Sensitivity Analysis	Trade-off calculations	Scale dependent economics	Product separations
This work	***NA***	***Y***	***Y***	***Air (420 ppm) & flue gas (15 mol%)***	***2.5 M KOH solution***	***2 M K2CO3 solution***	***Y***	***Y***	***Y***	***Y***	***Y***
Alerte et al.[50]	2023	Y	Y	Flue gas (6.4 wt% CO_2)	Monoethanolamine (MEA)	Humidified CO_2 (98.2 wt%)	N	Y	N	Y	Y
Guerra et al.[51]	2023	Y	N	Multiple sources	Suitable absorbents	Gaseous CO_2	Y	N	Y	N	Y
Badgett et al.[22]	2022	N	N	N	NA	Gaseous CO_2	Y	N	Y	Y	N
Ramdin et al.[52]	2021	Y	N	Biogas (40 wt% CO_2)	NA	Gaseous CO_2	N	N	N	N	Y
Shin et al.[13]	2021	N	Y	NA	NA	Gaseous CO_2	N	Y	N	N	Y
Orella et al.[53]	2020	N	N	NA	NA	Gaseous CO_2	Y	Y	N	N	Y
Li et al.[54]	2019	N	Y	Air (420 ppm)	2 M KOH solution	1.5 M K_2CO_3 solution	Y	N	N	N	N
De Luna et al.[11]	2019	N	N	Air (420 ppm)	NA	Gaseous CO_2	N	Y	N	Y	Y
Jouny et al.[12]	2018	Y	N	Air (420 ppm)	NA	Gaseous CO_2	N	Y	N	N	Y
Spurgeon et al.[55]	2018	N	Y	Flue gas	Monoethanolamine (MEA)	Gaseous CO_2	Y	Y	N	N	Y
Verma et al.[56]	2016	N	N	NA	NA	Gaseous CO_2	Y	N	N	N	N

Table 2. Parameters for process design and sensitivity analysis for 2 KMT/yr – 2 MMT/yr C_2H_4 plant scale.

Parameter	Pessimistic	Base Case	Optimistic	Units
Carbon Capture Unit				
Capture efficiency	75	80	90	%
CCU CAPEX	15	0	-25	% Change[1]
Upstream GO Concentrator Membrane Unit				
Threshold CO_3^{-2} concentration	2	2	1.5	M
Membrane permeance	2.1	2.8	3.5	$Lm^{-2}bar^{-1}h^{-1}$
CO_3^{-2} rejection	60	90	95	%
Membrane cost	200	200	165	$\$/m^2$
Membrane Lifetime	1	3	5	years
Electrolysis Unit				
Stack cost CAPEX	10	0	-25	% Change[2]
Overall conversion	20	35	65	%
Current density	0.3	0.5	1	$A.cm^{-2}$
Price of product	-15	0	15	% Change[3]
Electricity cost	0.08	0.02	0.015	$/kWh
C_2H_4 faradaic efficiency	31	50	70	%
C_2H_5OH faradaic efficiency	13	20	25	%
Cell Voltage[4]	5.2	4.3	5.5	V
Electrode area per cell	1.5	3.5	7	m^2/cell
Downstream cathode product gas separation unit				
H_2 permeance	1100	1100	1500	GPU*
C_2H_4 permeance	1.2	1.2	0.8	GPU*
Membrane cost	35	25	20	$\$/m^2$
Membrane Lifetime	5.5	6	7	years
Downstream Pervaporation Unit				
H_2O flux	1	1.5	1.5	$kg.m^{-2}.h$
Separation factor	8500	10000	10000	-
Membrane cost	75	70	60	$\$/m^2$
Membrane Lifetime	3	4	5	years
[1] Baseline CAPEX of CCU is 4668 USD/m^2 and assuming 250 m^2/m^3 packing material cost and 6.2 m packing depth[4]. **Note:** We represent the changes from this absolute value as "% change" for the optimistic and pessimistic cases. For example, the CCU CAPEX for the base case is 4668 $\$/m^2$, and for the pessimistic case a +15% change means the CCU CAPEX is 4668*(1+0.15) = 5368.2				
[2] Base case CAPEX for the carbonate-fed BPM electrolyzer is assumed to be 10,771 $\$/m^2$ and 4945 $\$/m^2$ for CO_2 gas-fed AEM electrolyzer based on calculations by Badgett et al.[22]				
[3] Baseline price of product for ethanol is 1 $/kg[22], for hydrogen 3.9 $/kg[11], and oxygen 0.13 $/kg				
[4] Cell voltage computed using the voltametric model described above and the basis for the assumption are in the methods section of the MS (values rounded to 1 significant digit).				
The detailed description for these assumptions are summarized in the methods section of the manuscript				
* **1 GPU (Gas Permeance Unit) =** 3.35×10^{-10} $mol.m^{-2}.s^{-1}.Pa^{-1}$				

Table 3. Base case stream flow table for all streams S1-S39 in the ASPEN flowsheet shown in Supplementary Figure 1a (Direct Air Capture).

Component	Unit	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	S12
Temperature	**C**	25.0	25.0	25.0	25.0	25.0	25.4	34.8	75.0	75.0	75.3	75.0	75.0
Pressure	**bar**	1.0	1.0	1.0	1.0	1.0	50.0	50.0	50.0	1.0	50.0	50.0	50.0
Total flow	kg/hr	1.E+08	1.E+08	2.E+09	2.E+09	1.E+08	1.E+08	2.E+08	2.E+08	1.E+08	1.E+08	1.E+08	5.E+07
CO2		0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.765	0.765	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.234	0.235	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.877	0.833	0.000	0.000	0.830	0.830	0.831	0.831	0.880	0.880	0.880	0.704
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.086	0.110	0.000	0.000	0.109	0.109	0.108	0.108	0.081	0.081	0.081	0.178
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.037	0.035	0.000	0.000	0.029	0.029	0.029	0.029	0.031	0.031	0.031	0.026
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.000	0.022	0.000	0.000	0.032	0.032	0.031	0.031	0.008	0.008	0.008	0.092

Component	Unit	S13	S14	S15	S16	S17	S18	S19	S20	S21	S22	S23	S24
Temperature	**C**	74.6	25.0	75.0	75.0	75.0	25.0	25.0	25.0	25.0	25.0	25.0	25.0
Pressure	**bar**	1.0	1.0	50.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
Total flow	kg/hr	5.E+07	5.E+07	3.E+07	9.E+07	9.E+07	9.E+07	5.E+07	5.E+07	5.E+07	2.E+06	5.E+07	5.E+07
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.036	1.000	0.000	0.000
H2O		0.704	0.704	0.838	0.894	0.894	0.894	0.894	0.894	0.862	0.000	0.697	0.702
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.004	0.004
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.006	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.178	0.178	0.105	0.073	0.073	0.073	0.073	0.073	0.071	0.000	0.185	0.186
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.026	0.026	0.031	0.031	0.031	0.031	0.031	0.031	0.030	0.000	0.045	0.046
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.092	0.092	0.026	0.002	0.002	0.002	0.002	0.002	0.002	0.000	0.062	0.062

Component	Unit	S24A	S24B	S24C	S24D	S24E	S24F	S24G	S24H	S24I	S24J	S24K	S24L
Temperature	**C**	91.2	91.2	91.2	74.8	64.8	64.8	64.8	64.8	53.9	53.9	0.0	25.0
Pressure	**bar**	0.5	0.5	0.3	0.25	0.14	0.14	0.2	0.15	0.06	0.064	0	0.5
Total flow	kg/hr	4E+07	4E+06	4E+07	4E+07	5E+06	4E+07	4E+07	4E+07	5E+06	3E+07	0E+00	4E+06
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.680	0.968	0.680	0.680	0.990	0.641	0.641	0.641	0.998	0.585	0.000	0.968
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.001	0.032	0.001	0.001	0.010	0.000	0.000	0.000	0.002	0.000	0.000	0.032
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.202	0.000	0.202	0.202	0.000	0.227	0.227	0.227	0.000	0.263	0.000	0.000
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.050	0.000	0.050	0.050	0.000	0.056	0.056	0.056	0.000	0.064	0.000	0.000
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.068	0.000	0.068	0.068	0.000	0.076	0.076	0.076	0.000	0.088	0.000	0.000

Component	Unit	S24M	S24N	S24O	S24P	S24Q	S24R	S24S	S24T	S24U	S24V	S24W	S24X
Temperature	**C**	0	25.0	25.0	0.0	25.0	25.0	25.0	25.0	0.0	90.0	99.1	93.1
Pressure	**bar**	0	0.25	1	0	0.15	1	1	1	0	1	0.98	1
Total flow	kg/hr	0	5E+06	4E+06	0E+00	5E+06	5E+06	5E+06	1E+07	0E+00	6E+06	1E+07	5E+07
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.000	0.990	0.968	0.000	0.998	0.990	0.998	0.987	0.000	1.000	1.000	0.702
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.010	0.032	0.000	0.002	0.010	0.002	0.013	0.000	0.000	0.000	0.000
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.189
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.046
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.063

Component	Unit	S25	S26	S27	S28	S29	S30	S32	S33	S34	S35	S36	S37
Temperature	**C**	25.0	104.7	25.0	25.0	25.0	95.3	28.1	0.0	28.1	25.0	0.0	25
Pressure	**bar**	1	2	2	1	1	0.98	1	0	1	1	0	1
Total flow	kg/hr	3E+05	3E+05	3E+05	7E+04	3E+05	5E+05	9E+07	0E+00	9E+07	5E+07	0E+00	46767755.99
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.000	0.000	0.000	0.000	0.000	0.686	0.800	0.000	0.800	0.894	0.000	0.894
H2		0.206	0.206	0.206	0.996	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000	0.000	0.000	0.314	0.000	0.000	0.000	0.000	0.000	0.000
C2H4		0.794	0.794	0.794	0.004	1.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.000	0.000	0.000	0.000	0.000	0.000	0.130	0.000	0.130	0.073	0.000	0.073
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.000	0.000	0.000	0.000	0.000	0.000	0.038	0.000	0.038	0.031	0.000	0.031
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.000	0.000	0.000	0.000	0.000	0.000	0.032	0.000	0.032	0.002	0.000	0.002

*Streams S33 and S36 are purge streams with no flow.

Component	Unit	S38	S39	STEAM
Temperature	C	25	25.0	160
Pressure	bar	1	1	1
Total flow	kg/hr	1E+08	5E+07	6450000
CO2		0.000	0.000	0.000
N2		0.000	0.000	0.000
O2		0.000	0.000	0.000
H2O		0.876	0.702	1.000
H2		0.000	0.000	0.000
HCOOH		0.000	0.000	0.000
CO		0.000	0.000	0.000
CH3OH		0.000	0.000	0.000
C		0.000	0.000	0.000
CH4		0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000
C2H4		0.000	0.000	0.000
C2H6		0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000
H+		0.000	0.000	0.000
K+		0.087	0.189	0.000
KOH(S)		0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000
HCO3-		0.000	0.000	0.000
OH-		0.038	0.046	0.000
HCOO-		0.000	0.000	0.000
CO3--		0.000	0.063	0.000

Table 4. Optimistic case stream flow table for all streams S1-S38 in the ASPEN flowsheet shown in Supplementary Figure 1b (Direct Air Capture).

Component	**Unit**	**S1**	**S2**	**S3**	**S4**	**S5**	**S6**	**S7**	**S8**	**S9**	**S10**	**S11**	**S12**
Temperature	**C**	25.0	25.0	25.0	25.0	25.0	25.4	28.3	75.0	75.0	75.3	75.0	75.0
Pressure	**bar**	1.0	1.0	1.0	1.0	1.0	50.0	50.0	50.0	1.0	50.0	50.0	50.0
Total flow	kg/hr	4.E+04	4.E+04	6.E+05	6.E+05	4.E+04	4.E+04	4.E+04	4.E+04	3.E+04	3.E+04	3.E+04	9.E+03
CO2		0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.765	0.765	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.234	0.235	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.877	0.863	0.000	0.000	0.859	0.859	0.857	0.857	0.893	0.893	0.893	0.746
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.086	0.093	0.000	0.000	0.093	0.093	0.094	0.094	0.074	0.074	0.074	0.156
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.037	0.037	0.000	0.000	0.030	0.030	0.030	0.030	0.031	0.031	0.031	0.027
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.000	0.007	0.000	0.000	0.019	0.019	0.019	0.019	0.003	0.003	0.003	0.072

Component	**Unit**	**S13**	**S14**	**S15**	**S16**	**S17**	**S18**	**S19**	**S20**	**S21**	**S22**	**S23**	**S24**
Temperature	**C**	74.7	25.0	75.0	75.0	75.0	25.0	25.0	25.0	25.0	25.0	25.0	25.0
Pressure	**bar**	1.0	1.0	50.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
Total flow	kg/hr	9.E+03	9.E+03	2.E+03	3.E+04	3.E+04	3.E+04	1.E+04	1.E+04	1.E+04	4.E+02	9.E+03	9.E+03
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.026	1.000	0.000	0.000
H2O		0.746	0.746	0.836	0.898	0.898	0.898	0.898	0.898	0.874	0.000	0.743	0.749
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.005	0.005
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.008	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.156	0.156	0.107	0.071	0.071	0.071	0.071	0.071	0.069	0.000	0.162	0.163
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.027	0.027	0.032	0.030	0.030	0.030	0.030	0.030	0.030	0.000	0.056	0.056
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.072	0.072	0.026	0.001	0.001	0.001	0.001	0.001	0.001	0.000	0.026	0.026

Component	**Unit**	**S24A**	**S24B**	**S24C**	**S24D**	**S24E**	**S24F**	**S24G**	**S24H**	**S24I**	**S24J**	**S24K**	**S24L**
Temperature	C	70.9	99.2	0.0	104.0	104.2	186.3	105.1	105.2	143.9	139.0	130.3	130.3
Pressure	bar	1.0	0.8	0.0	1.0	1.0	1.9	1.9	5.5	5.5	5.5	5.5	5.5
Total flow	kg/hr	9E+03	9E+03	0E+00	9E+03	6E+02	6E+02	6E+02	6E+02	6E+02	6E+02	6E+01	1E+01
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.749	0.749	0.000	0.749	0.927	0.927	0.927	0.927	0.927	0.927	0.200	0.984
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.005	0.005	0.000	0.005	0.073	0.073	0.073	0.073	0.073	0.073	0.800	0.016
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.163	0.163	0.000	0.163	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.056	0.056	0.000	0.056	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.026	0.026	0.000	0.026	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000

Component	Unit	S24N	S24O	S24P	S24Q	S24R	S25	S26	S27	S28	S29	S30	S31
Temperature	**C**	155.4	125.2	100.1	110.4	100.1	25.0	94.8	25.0	25.0	25.0	130.3	75.9
Pressure	**bar**	5.5	5.5	1.0	1.1	1.0	1.0	2.0	2.0	1.0	1.0	5.5	1.1
Total flow	**kg/hr**	6E+02	6E+02	6E+02	9E+03	6E+02	8E+01	8E+01	8E+01	2E+00	7E+01	4E+01	8E+03
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.999	0.999	0.999	0.753	0.999	0.000	0.000	0.000	0.000	0.000	0.001	0.751
H2		0.000	0.000	0.000	0.000	0.000	0.030	0.030	0.030	0.983	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.001	0.001	0.001	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.999	0.000
C2H4		0.000	0.000	0.000	0.000	0.000	0.970	0.970	0.970	0.017	1.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.000	0.000	0.000	0.164	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.166
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.000	0.000	0.000	0.056	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.057
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.000	0.000	0.000	0.026	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.026

Component	Unit	S32	S33	S34	S35	S36	S37	S38
Temperature	**C**	S32	S33	S34	S35	S36	S37	S38
Pressure	**bar**	43.9	0.0	43.9	25.0	0.0	25.0	25.0
Total flow	kg/hr	1	0	1	1	0	1	1
CO2		2.2E+04	0.0E+00	2.2E+04	1.4E+04	0.0E+00	1.4E+04	3.5E+04
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2		0.841	0.000	0.841	0.898	0.000	0.898	0.877
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.000	0.000	0.000	0.000	0.000	0.000	0.000
KOH(S)		0.108	0.000	0.108	0.071	0.000	0.071	0.086
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOO-		0.041	0.000	0.041	0.030	0.000	0.030	0.037
CO3--		0.000	0.000	0.000	0.000	0.000	0.000	0.000

*Streams S33 and S367 are purge streams with no flow.

Table 5. Pessimistic case stream flow table for all streams S1-S45 in the ASPEN flowsheet shown in Supplementary Figure 1c (Direct Air Capture).

Component	Unit	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	S12
Temperature	C	25.0	25.0	25.0	25.0	25.0	25.4	34.8	75.0	75.0	75.3	75.0	75.0
Pressure	bar	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Total flow	kg/hr	3.E+09	3.E+09	5.E+10	5.E+10	3.E+09	3.E+09	3.E+09	3.E+09	4.E+08	3.E+09	8.E+07	3.E+08
CO2		0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.765	0.765	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.234	0.235	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.877	0.853	0.000	0.000	0.849	0.849	0.848	0.848	0.751	0.863	0.674	0.770
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.086	0.099	0.000	0.000	0.098	0.098	0.099	0.099	0.155	0.090	0.199	0.144
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.037	0.036	0.000	0.000	0.030	0.030	0.030	0.030	0.033	0.029	0.034	0.033
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.000	0.012	0.000	0.000	0.023	0.023	0.023	0.023	0.060	0.018	0.092	0.052

Component	Unit	S13	S14	S15	S16	S17	S18	S19	S20	S21	S22	S23	S24
Temperature	**C**	74.6	25.0	75.0	75.0	75.0	25.0	25.0	25.0	25.0	25.0	25.0	25.0
Pressure	**bar**	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Total flow	kg/hr	3.E+09	3.E+09	3.E+09	9.E+08	2.E+09	2.E+09	2.E+09	1.E+09	1.E+09	8.E+07	8.E+07	8.E+08
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.853	0.853	0.853	0.804	0.874	0.874	0.874	0.874	0.874	0.674	0.674	0.804
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.096	0.096	0.096	0.125	0.084	0.084	0.084	0.084	0.084	0.199	0.199	0.125
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.030	0.030	0.030	0.032	0.028	0.028	0.028	0.028	0.028	0.034	0.034	0.032
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.022	0.022	0.022	0.039	0.014	0.014	0.014	0.014	0.014	0.092	0.092	0.039

Component	Unit	S25	S26	S27	S28	S29	S30	S31	S32	S33	S34	S35	S36
Temperature	**C**	25.0	104.7	25.0	25.0	25.0	95.3	28.1	28.1	25.0	0.0	25	25
Pressure	**bar**	50	1	0	1	1	1	1	2	2	1	1	1
Total flow	**kg/hr**	9E+07	1E+09	0E+00	1E+09	1E+09	8E+07	4E+05	4E+05	4E+05	2E+05	3E+05	1E+09
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.002
H2O		0.804	0.859	0.000	0.859	0.859	0.665	0.000	0.000	0.000	0.000	0.000	0.872
H2		0.000	0.000	0.000	0.000	0.000	0.002	0.402	0.402	0.402	0.998	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.000	0.000	0.000
C2H4		0.000	0.000	0.000	0.000	0.000	0.003	0.632	0.632	0.632	0.003	1.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.125	0.092	0.000	0.092	0.092	0.205	0.000	0.000	0.000	0.000	0.000	0.084
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.032	0.030	0.000	0.030	0.030	0.046	0.000	0.000	0.000	0.000	0.000	0.028
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.039	0.019	0.000	0.019	0.019	0.077	0.000	0.000	0.000	0.000	0.000	0.014

Component	Unit	S37	S38	S39	S40	S41	S42	S43	S44	S45	STEAM
Temperature	**C**	160	160	160	160	160	160	25.0	0.0	160	160
Pressure	**bar**	1	1	0	1	1	1	0.98	1	1	1
Total flow	**kg/hr**	2E+06	1E+09	0E+00	1E+09	1E+09	8E+07	4E+05	7E+07	2E+09	6E+06
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		1.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.000	0.874	0.000	0.874	0.874	0.668	0.665	0.668	0.900	1.000
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000	0.000	0.000	0.002	0.335	0.000	0.000	0.000
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.000	0.084	0.000	0.084	0.084	0.206	0.000	0.208	0.070	0.000
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.000	0.028	0.000	0.028	0.028	0.046	0.000	0.046	0.030	0.000
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.000	0.014	0.000	0.014	0.014	0.078	0.000	0.078	0.000	0.000

*Streams S36 and S39 are purge streams with no flow.

Component	**Unit**	**S42A**	**S42B**	**S42C**	**S42D**	**S42E**	**S42F**	**S42G**	**S42H**	**S42I**	**S42J**	**S42K**	**S42L**
Temperature	**C**	91.2	91.2	91.2	74.8	64.8	64.8	64.8	64.8	53.9	53.9	0.0	25.0
Pressure	**bar**	0.5	0.5	0.3	0.3	0.1	0.1	0.2	0.2	0.1	0.1	0.0	0.5
Total flow	kg/hr	7E+07	2E+06	7E+07	7E+07	4E+06	7E+07	7E+07	7E+07	4E+06	7E+07	0E+00	2E+06
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.660	0.970	0.660	0.660	0.984	0.643	0.643	0.643	0.994	0.619	0.000	0.970
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.001	0.030	0.001	0.001	0.016	0.001	0.001	0.001	0.006	0.000	0.000	0.030
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.212	0.000	0.212	0.212	0.000	0.223	0.223	0.223	0.000	0.238	0.000	0.000
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.047	0.000	0.047	0.047	0.000	0.049	0.049	0.049	0.000	0.053	0.000	0.000
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.080	0.000	0.080	0.080	0.000	0.084	0.084	0.084	0.000	0.090	0.000	0.000

Component	Unit	S42M	S42N	S42O	S42P	S42Q	S42R	S42S	S42T	S42U	S42V	S42W	S42X
Temperature	**C**	0.0	25.0	25.0	0.0	25.0	25.0	25.0	25.0	0.0	90.0	99.1	93.1
Pressure	**bar**	0.0	0.3	1.0	0.0	0.2	1.0	1.0	1.0	0.0	1.0	1.0	1.0
Total flow	kg/hr	0E+00	4E+06	2E+06	0E+00	4E+06	4E+06	4E+06	1E+07	0E+00	6E+06	1E+07	7E+07
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.000	0.984	0.970	0.000	0.994	0.984	0.994	0.986	0.000	1.000	1.000	0.668
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.016	0.030	0.000	0.006	0.016	0.006	0.014	0.000	0.000	0.000	0.000
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.208
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.046
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.078

Table 6. Fitted parameters for the residence time distributions using the open-vessel dispersion model, and rigorously calculated values from CFD simulations. τ_{exp} is the experimental mean residence time. Each flow condition is designated by the flow rate (10 or 65 mL/min) and the flow pattern which is serpentine (SF) or interdigitated (IDF). The shaded column corresponds to the results obtained from the CFD simulation. Despite the high R^2 values, the dispersion model derived for an encapsulated channel is not applicable to the MEA electrolyzer architecture as it does not account for the through plane diffusion.

Condition	τ_{exp} (s)	τ_{model} (s)	D/uL (model)	R^2	V_{CF_CFD} (10^{-5} m/s)	Pe_{CFD}
10 SF	18.4	17.3	0.01	0.993	0.61	0.27
10 IDF	17.8	12.8	0.03	0.908	2.66	1.20
65 SF	3.6	2.6	0.15	0.980	4.97	2.24
65 IDF	3.5	1.9	0.34	0.993	19.90	8.97

Table 7. Balance of Plant (BoP) calculations for the electrolyzer unit and the auxiliary units in the entire process for the base case scenario.

System	Subsystem	Unit cost	Unit	Scaling factor	Electrolyzer size (kW)			Cost for the electrolyzer unit (USD)
					200	1000	11194	
Power supply	DC Voltage rectifier (transducer)			1	225	225	225	499,725
	DC Current rectifier (transducer)			1	340	340	340	755,140
	AC-DC rectifier[2]	130	USD/kWh	0.85	11,744	46,126	359,415	798,260,607
	Total				12,309	46,691	359,980	799,515,472
Water circulation	Including separator tanks, circulation & polishing pump, piping, valves & instrumentation, controls	7279	USD/kWh	0.36	48,842	87,082	207,409	460,655,623
	Total				48,842	87,082	207,409	460,655,623
Gas-liquid separators for anode & cathode	GLS unit cost, tubing, valving & instrumentation, controls	1951	USD/kWh	0.46	22,500	47,291	144,187	320,238,889
	Total				22,500	47,291	144,187	320,238,889
Miscellaneous	Valve air supply – N2 or compressed air				2,000	2,000	2,000	4,442,000
	Ventilation and safety requirements				4,000	4,000	4,000	8,884,000
	Total				6,000	6,000	6,000	13,326,000
Cooling systems	Plate heat exchanger, pumps, tubing, valving & instrumentation, dry cooler	3442	USD/kWh	0.31	17,500	28,679	60,190	133,681,026
	Total				17,500	28,679	60,190	133,681,026
Auxiliary units in the entire process	Pumps, heat exchangers and evaporators in the entire process, not associated with the electrolysis unit (Supplementary Table 11)							212,412,700
BOP grand total								$ 1,921,713,413
Electrolyzer CAPEX after scaling								$ 6,589,153,282
BoP as a function of electrolyzer CAPEX								**29%**
[1] All calculations and costs shown here are based on the work by Mayyas et al.[57], excluding the costs specific to water electrolysis.								
[2] The costs for the rectifier were obtained from Mongird et al.[58]								

Table 8. Base case stream flow table for all streams S1-S40 in the ASPEN flowsheet shown in Figure S1a (Flue gas capture).

Component	Unit	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	S12
Temperature	**C**	25.0	25.0	25.0	25.0	25.0	25.4	34.8	75.0	75.0	75.3	75.0	75.0
Pressure	**bar**	1.0	1.0	1.0	1.0	1.0	50.0	50.0	50.0	1.0	50.0	50.0	50.0
Total flow	kg/hr	1.E+08	1.E+08	5.E+06	4.E+06	1.E+08	1.E+08	2.E+08	2.E+08	1.E+08	1.E+08	1.E+08	3.E+07
CO2		0.000	0.000	0.217	0.066	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.783	0.934	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.967	0.930	0.000	0.000	0.927	0.927	0.925	0.925	0.965	0.965	0.965	0.764
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.023	0.044	0.000	0.000	0.043	0.043	0.044	0.044	0.022	0.022	0.022	0.135
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.010	0.010	0.000	0.000	0.005	0.005	0.005	0.005	0.005	0.005	0.005	0.005
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.000	0.017	0.000	0.000	0.025	0.025	0.025	0.025	0.008	0.008	0.008	0.096

Component	Unit	S13	S14	S15	S16	S17	S18	S19	S20	S21	S22	S23	S24
Temperature	**C**	74.6	25.0	75.0	75.0	75.0	25.0	25.0	25.0	25.0	25.0	25.0	25.0
Pressure	**bar**	1.0	1.0	50.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
Total flow	kg/hr	3.E+07	3.E+07	3.E+07	1.E+08	1.E+08	1.E+08	5.E+07	5.E+07	5.E+07	1.E+06	3.E+07	3.E+07
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.024	1.000	0.000	0.000
H2O		0.764	0.764	0.917	0.979	0.979	0.979	0.979	0.979	0.955	0.000	0.759	0.764
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.002	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.004	0.004
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.006	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.135	0.135	0.049	0.014	0.014	0.014	0.014	0.014	0.014	0.000	0.141	0.142
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.005	0.005	0.006	0.005	0.005	0.005	0.005	0.005	0.005	0.000	0.025	0.025
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.096	0.096	0.028	0.002	0.002	0.002	0.002	0.002	0.002	0.000	0.065	0.065

Component	Unit	S24A	S24B	S24C	S24D	S24E	S24F	S24G	S24H	S24I	S24J	S24K	S24L
Temperature	**C**	91.2	91.2	91.2	74.8	64.8	64.8	64.8	64.8	53.9	53.9	0.0	25.0
Pressure	**bar**	0.5	0.5	0.3	0.25	0.13	0.13	0.2	0.15	0.06	0.06	0	0.5
Total flow	kg/hr	2E+07	5E+06	2E+07	2E+07	5E+06	2E+07	2E+07	2E+07	5E+06	1E+07	0E+00	5E+06
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.725	0.981	0.725	0.725	0.996	0.651	0.651	0.651	0.999	0.516	0.000	0.981
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.001	0.019	0.001	0.001	0.004	0.000	0.000	0.000	0.001	0.000	0.000	0.019
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.168	0.000	0.168	0.168	0.000	0.213	0.213	0.213	0.000	0.296	0.000	0.000
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.029	0.000	0.029	0.029	0.000	0.037	0.037	0.037	0.000	0.052	0.000	0.000
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.077	0.000	0.077	0.077	0.000	0.098	0.098	0.098	0.000	0.136	0.000	0.000

Component	Unit	S24M	S24N	S24O	S24P	S24Q	S24R	S24S	S24T	S24U	S24V	S24W	S24X
Temperature	**C**	0	25.0	25.0	0.0	25.0	25.0	25.0	25.0	0.0	90.0	99.1	93.1
Pressure	**bar**	0	0.25	1	0	0.15	1	1	1	0	1	0.98	1
Total flow	kg/hr	0	5E+06	5E+06	0E+00	5E+06	5E+06	5E+06	2E+07	0E+00	6E+06	1E+07	3E+07
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.000	0.996	0.981	0.000	0.999	0.996	0.999	0.993	0.000	1.000	1.000	0.764
H2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.004	0.019	0.000	0.001	0.004	0.001	0.007	0.000	0.000	0.000	0.000
C2H4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.144
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.025
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.066

Component	Unit	S25	S26	S27	S28	S29	S30	S32	S33	S34	S35	S36	S37
Temperature	**C**	25.0	104.7	25.0	25.0	25.0	95.3	28.1	0.0	28.1	25.0	0.0	25
Pressure	**bar**	1	2	2	1	1	0.98	1	0	1	1	0	1
Total flow	kg/hr	2E+05	2E+05	2E+05	4E+04	2E+05	5E+05	8E+07	0E+00	8E+07	5E+07	0E+00	48438179
CO2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
N2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
O2		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H2O		0.000	0.000	0.000	0.000	0.000	0.778	0.898	0.000	0.898	0.979	0.000	0.979
H2		0.206	0.206	0.206	0.996	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCOOH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH3OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CH4		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000	0.000	0.000	0.222	0.000	0.000	0.000	0.000	0.000	0.000
C2H4		0.794	0.794	0.794	0.004	1.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C2H6		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H+		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K+		0.000	0.000	0.000	0.000	0.000	0.000	0.063	0.000	0.063	0.014	0.000	0.014
KOH(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HCO3-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
OH-		0.000	0.000	0.000	0.000	0.000	0.000	0.013	0.000	0.013	0.005	0.000	0.005
HCOO-		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO3--		0.000	0.000	0.000	0.000	0.000	0.000	0.026	0.000	0.026	0.002	0.000	0.002

*Streams S33 and S36 are purge streams with no flow and hence have been excluded.

Component	Unit	S38	S39	STEAM
Temperature	**C**	25	25.0	160
Pressure	**bar**	1	1	1
Total flow	kg/hr	1.24E+08	3E+07	6450000
CO2		0.000	0.000	0.000
N2		0.000	0.000	0.000
O2		0.000	0.000	0.000
H2O		0.967	0.764	1.000
H2		0.000	0.000	0.000
HCOOH		0.000	0.000	0.000
CO		0.000	0.000	0.000
CH3OH		0.000	0.000	0.000
C		0.000	0.000	0.000
CH4		0.000	0.000	0.000
C2H5OH		0.000	0.000	0.000
C2H4		0.000	0.000	0.000
C2H6		0.000	0.000	0.000
C3H7OH		0.000	0.000	0.000
H+		0.000	0.000	0.000
K+		0.023	0.144	0.000
KOH(S)		0.000	0.000	0.000
K2CO3(S)		0.000	0.000	0.000
HCO3-		0.000	0.000	0.000
OH-		0.010	0.025	0.000
HCOO-		0.000	0.000	0.000
CO3--		0.000	0.066	0.000

Table 9. Effect of absorbent concentration on CO_2 capture efficiency for direct air capture scenario. The thermodynamic properties required to compute the capture efficiency are also summarized.

Case	$\mathbf{k_r}$	**[OH$^-$]**	$\boldsymbol{\gamma_i}$	$\mathbf{K_H}$	$\boldsymbol{\mu}^2$	$\mathbf{D_l}$	$\mathbf{k}_{\boldsymbol{spray}}$	$\boldsymbol{\eta}$
	$Lmol^{-1}s^{-1}$	mol L^{-1}	(i = CO_2, KOH)	$\left[\frac{\mathbf{mol/L_{soln}}}{\mathbf{mol/L_{\mathit{air}}}}\right]$	cP	m^2s^{-1}	m/s	%
0.33 M KOH	7301	0.33	1	0.6	0.92	2.07E-9	0.0013	58.2
1.33 M KOH	13455	1.33	1	0.6	1.08	1.72E-9	0.0033	88.6
2.5 M KOH	25496	2.5	1	0.6	1.26	1.44E-9	0.0058	97.6
3 M KOH	32677	3	1	0.6	1.32	1.37E-9	0.0069	98.9

Table 10. The cell voltage distribution for a BPM-MEA electrolyzer system for the pessimistic case scenario.

Section	Properties	Calculation	Voltage (V)
Bipolar plate (×2)	Material: SS316, $\rho_{SS316} =$ 70 $\mu\Omega$ cm	$0.3\,\frac{A}{cm^2} \times 70\ \mu\Omega\ cm \times 0.2 cm$	0.0000084
GDL	Material: 39BB carbon paper, $\rho_{39BB} = 13\ m\Omega$ cm^2 [Data from Fuel Cell Store]	$0.3\,\frac{A}{cm^2} \times 0.013\ \Omega\ cm^2$	0.0039
Contact resistance between BP & GDL	Assuming a value of 0.005 $\Omega\ cm^2$ [28]	$0.3\,\frac{A}{cm^2} \times 0.005\ \Omega\ cm^2$	0.0015
pH modulating layer (IP)	Material: Cellulose Acetate Porosity=85%, $\rho_{IP} = 50\ \Omega$ cm	$0.3\,\frac{A}{cm^2} \times 50\ \Omega\ cm \times 135\ \mu m$	0.2025
Contact resistance between IP & GDL	Assuming a value of 0.005 $\Omega\ cm^2$ [28]	$0.3\,\frac{A}{cm^2} \times 0.005\ \Omega\ cm^2$	0.0025
Ionic resistance across BPM	Assuming a value of 4 Ω cm^2	$0.3\,\frac{A}{cm^2} \times 4\ \Omega\ cm^2$	1.2
Cathodic overpotential	Assuming a TS=1037 mVdec^{-1} (fitted) and η_{ref}= 920 mV at 200 mA cm^{-2}	$1.037\ V \times \log\left(\frac{0.3}{0.2}\right) + 0.92\ V$	1.1026
Anodic overpotential	Assuming a TS=41 mV dec^{-1} η_{ref}=247 mV at 10 mA cm^{-2}	$0.041\ V \times \log\left(\frac{0.3}{0.01}\right) + 0.247\ V$	0.3076
Thermodynamic requirements	SRP for OER, C_2H_4, ethanol formation and WDR at BPM	1.913 V	
Nernstian overpotential due to pH gradient	59mv * (pF difference)	0.472 V	
	Full cell voltage	**5.20**	

Table 11. The costs associated with the auxiliary units like pumps, heat exchangers and evaporation drums for the base case scenario calculated using Aspen Plus Economic analyzer.

Unit operation		
Name	**Installed Cost [USD]**	**Utility Cost [USD/HR]**
HX2	7509900	0
H3	0	10692.82836
G2	5863000	0
H6	2879600	67.91676
P7	5311600	4.336125
H4	0	3305.4288
P2	15802100	12430.225
P5	673400	14.45375
P1	17257200	15205.345
P6	9749300	751.595
H2	423100	89.49504
P4	11940700	809.41
EVAP2	17289800	0
HX1	10409700	0
H1	26864700	177253.1001
D1	14891600	45399.13489
HX5	0	4677.48396
P8	456800	5.7815
G1	4596000	0
EVAP1	8640900	0
P3	51203000	1734.45
P9	650300	11.563
Heating and cooling utilities	**-**	**194,023,951**
Total	**$212,412,700.00**	**$194,296,403**

Table 12. Scope 1 and scope 2 emissions of the BPM-based CO_2RR electrolysis process to produce 2 MMT/yr ethylene for the optimistic case.

Emission source	Parameters	Emission factor	CO_2 emission [ton CO2]	CO_2 emission [ton CO2/ton C2H4]
Steam	6,354,112 MMBTU of low pressure steam and 1,033,821 MMBTU of high pressure steam, values from Aspen Plus simulations	66.33 [kg/MMBTU]	612,552.01	0.306
Refrigerant	Assuming HFC-134a and 15 MW requirement, values from Aspen Plus simulations	1300 [GWP]	523,177	0.261
Process intensification of heating and cooling utilities	Assuming heat integration and process optimization approaches like pinch analysis can reduce total requirement by 40%	NA	NA	0.37
Electricity	195,660 GWh of electricity (full process)	868.6 [lb/MWh] grid electricity	77,089,445.19	38.544
		95 [lb/MWh] Solar powered	8,431,380.72	4.215
Carbon captured in CCU through DAC	Value from Aspen Plus for the optimistic scenario	NA	NA	4.489

Table 13. Comparison of the evaporation-distillation and distillation-pervaporation routes to recover ethanol from the process for the pessimistic case.

Parameter	Value	
	Evaporation-distillation	**Distillation-pervaporation**
Ethanol recovery [%]	90.8	98.6
Ethanol concentration in S30 [wt%]	34.7	99.8
Total Capital Cost [USD]	116,994,000	101,965,000
Total Operating Cost [USD/yr]	375,706,000	2,279,270,000
Total Utilities Cost (incl. electricity at 0.02 $/kWh) and cooling water) [USD/yr]	88,309,677	2,104,300,000
Ethanol selling price [USD/kg]	0.125	0.8
Income from products [USD/yr]	468,923,252	1,457,596,131
NPV at end of plant of life (20 years)	304,782,813	-3,610,779,321

Supplementary Figures

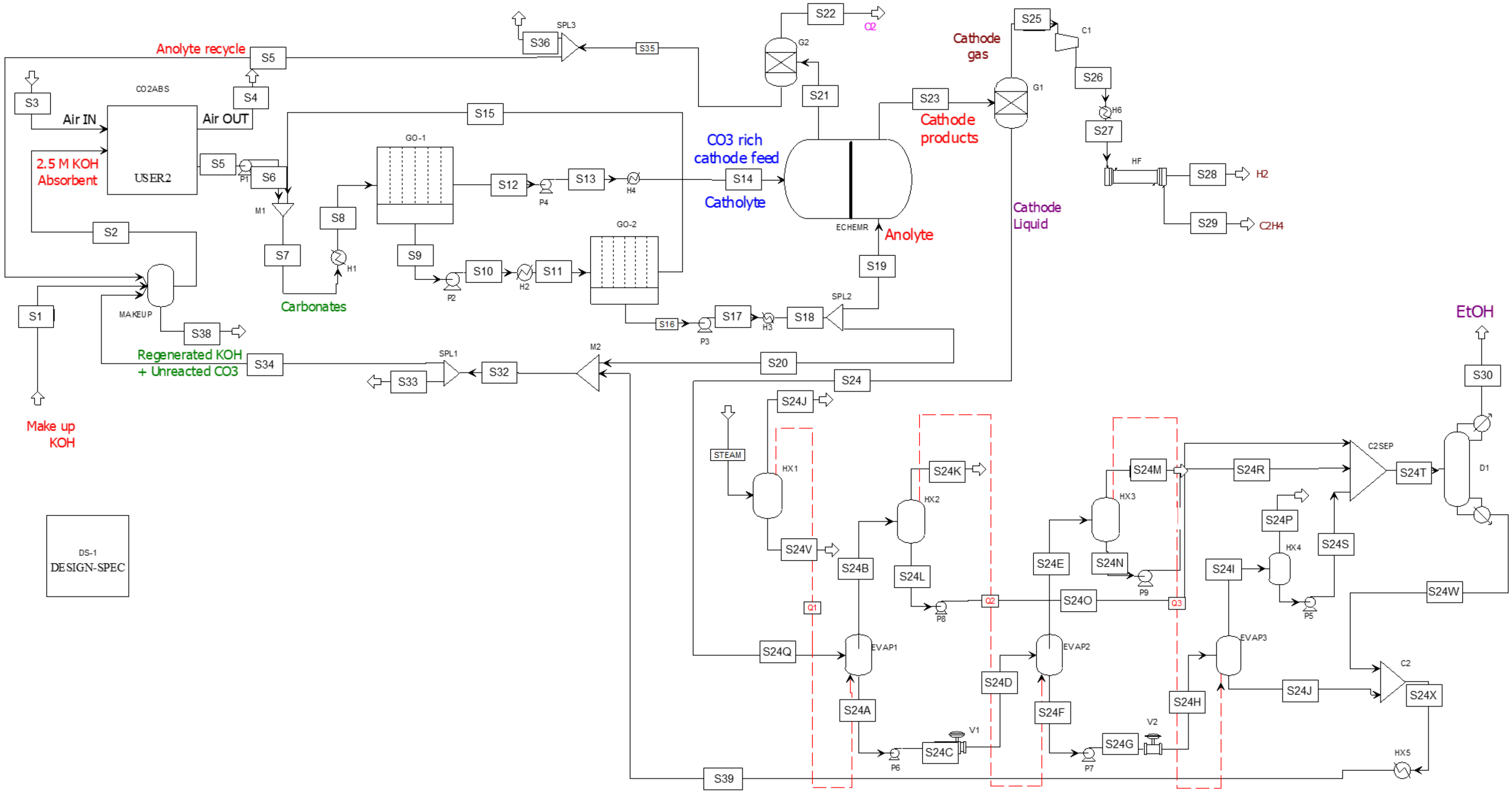


Figure 1(a) ASPEN Plus flowsheet for the **base case**. Recovery of ethanol from the cathode liquid stream is achieved using 3-effect vacuum evaporator and a distillation column to concentrate the recovered ethanol. ~91% ethanol was recovered from the cathode liquid stream (S24) and concentrated to 35% using the distillation column D1. The recycle stream to the CCU (S24X), comprising the bottoms from the column and the concentrate from the 3rd evaporator, was cooled to 298 K using HX5. The salts are recycled back to the make-up tank using stream "S39".

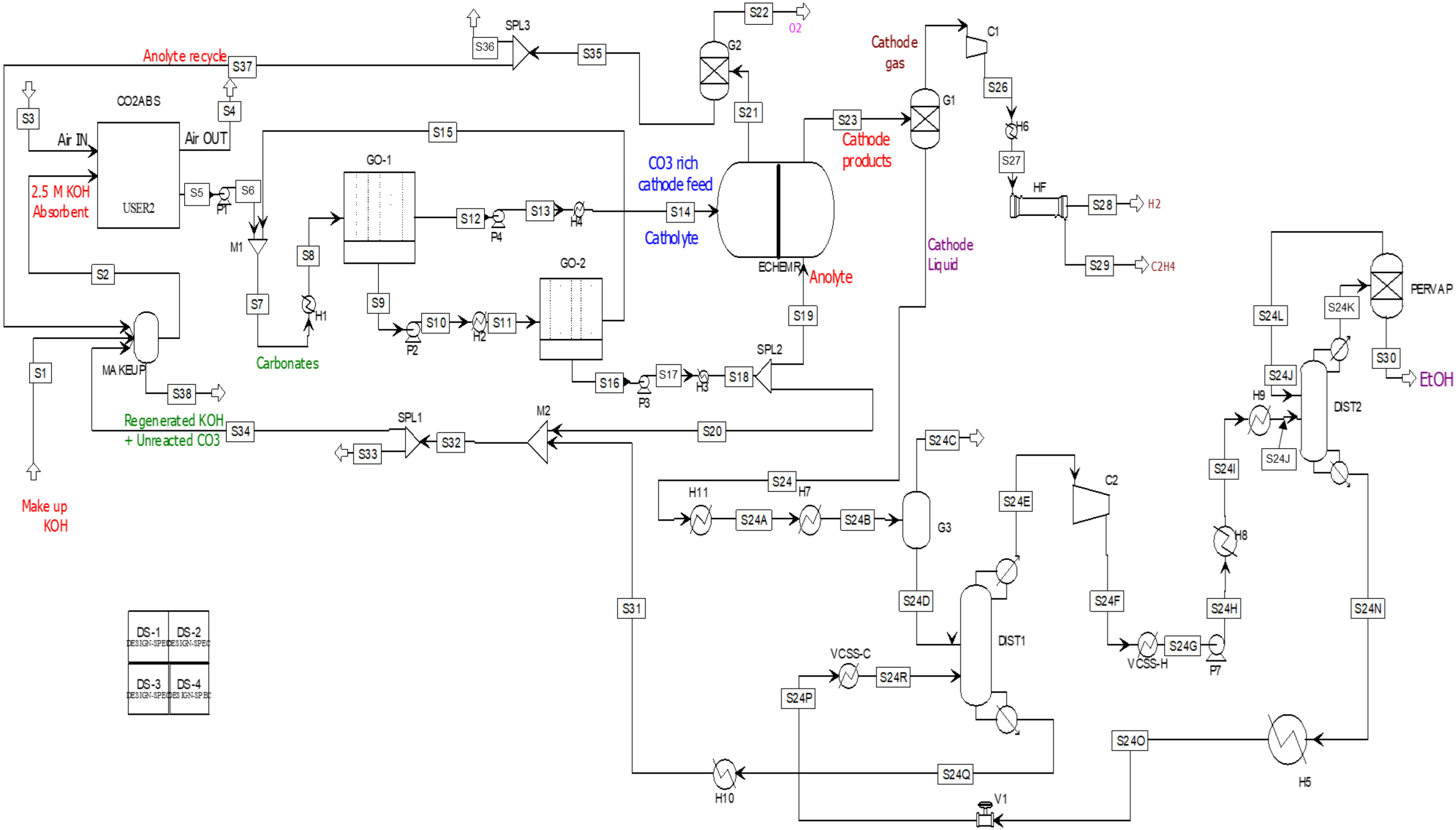


Figure 1(b) ASPEN Plus flowsheet for the **optimistic** case. Recovery of ethanol from the cathode liquid stream using 2 distillation columns – column "DIST1" to maximize recovery of the ethanol and "DIST2" to concentrate the recovered ethanol to 80 wt% (S24K). The pervaporation unit was used downstream of the 2nd column to purify ethanol to concentrations >99% (S30), and the recycle to the CCU (bottoms from "DIST1") was cooled to 303K using H10. Minimal heat integration was carried out to make the overall process feasible (Heat exchangers VCSS-C and VCSS-H).

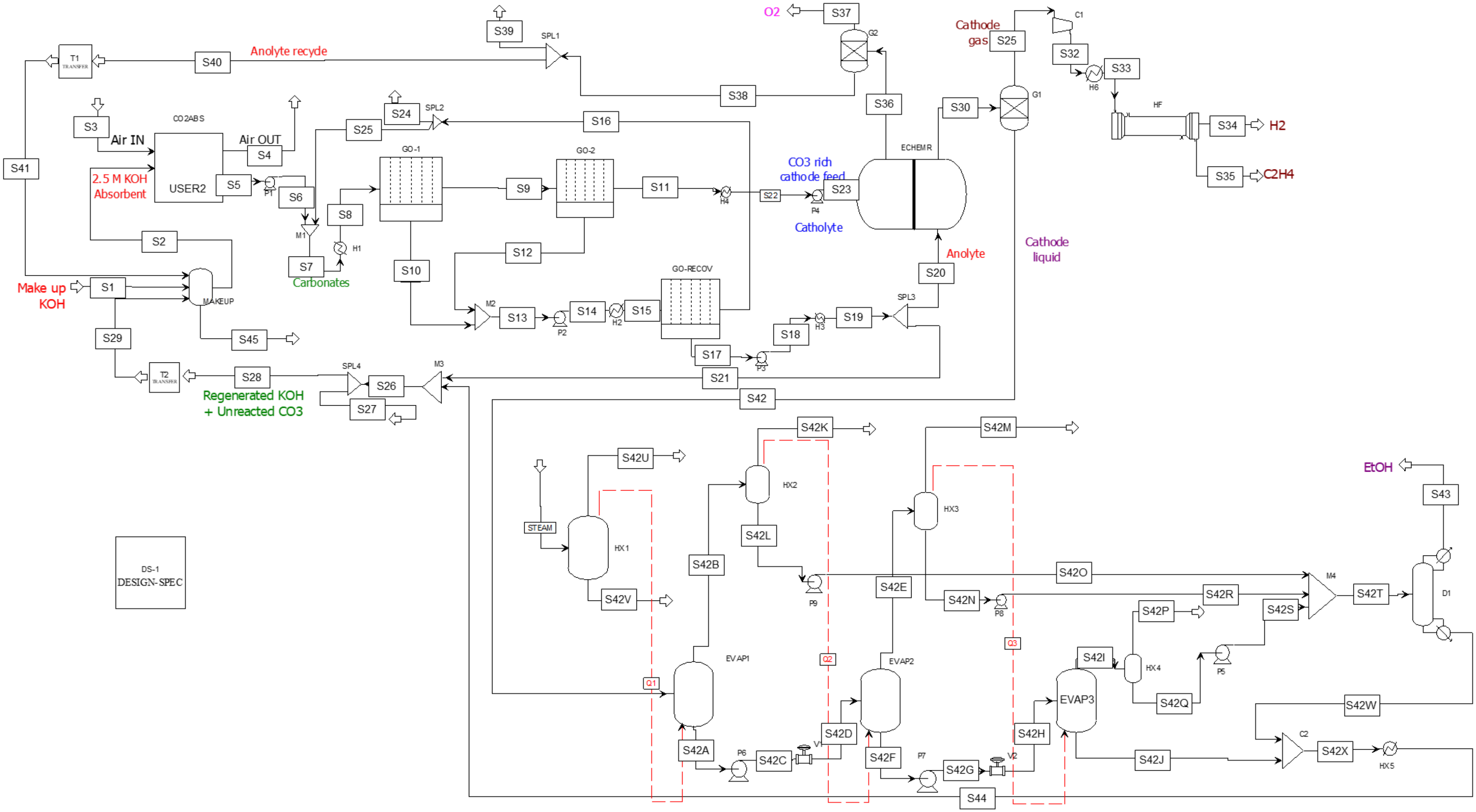


Figure S1(c) ASPEN Plus flowsheet for the **pessimistic** case. The separation of ethanol is carried out using a 3-effect vacuum evaporator followed by a distillation unit, the salts are recycled back to the make-up tank using stream "S44". We employ a 2-stage GO membrane for concentrating the carbonates upstream due to the low carbonate rejection assumption (60%) for the membranes.

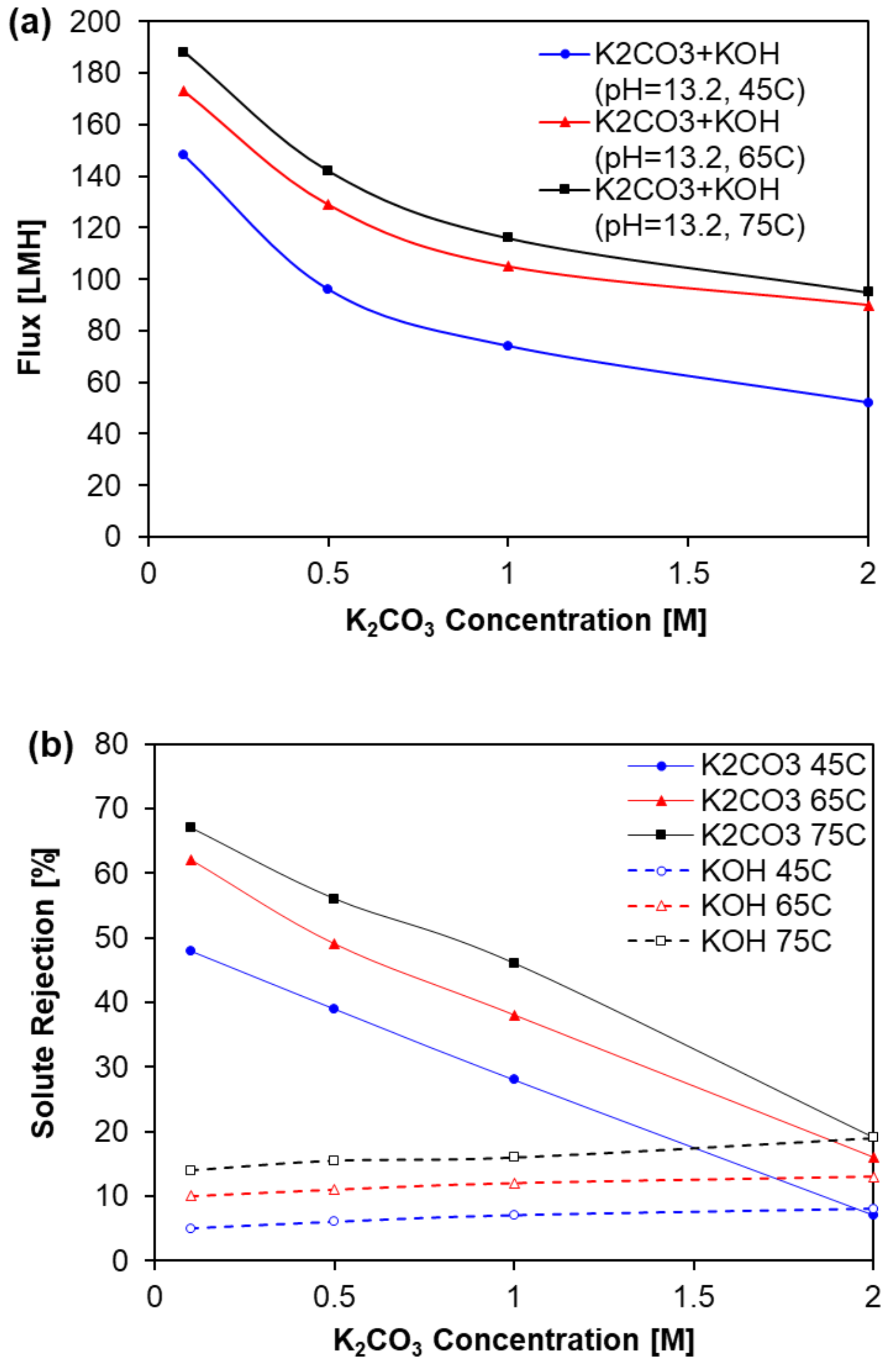


Figure 2. (a) Permeate flux and **(b)** K_2CO_3 and KOH salt rejection, as a function of inlet K_2CO_3 concentration using a graphene oxide (GO) membrane operating at a transmembrane pressure of 50 bar and at three different temperatures from 45, 65 and 75℃.

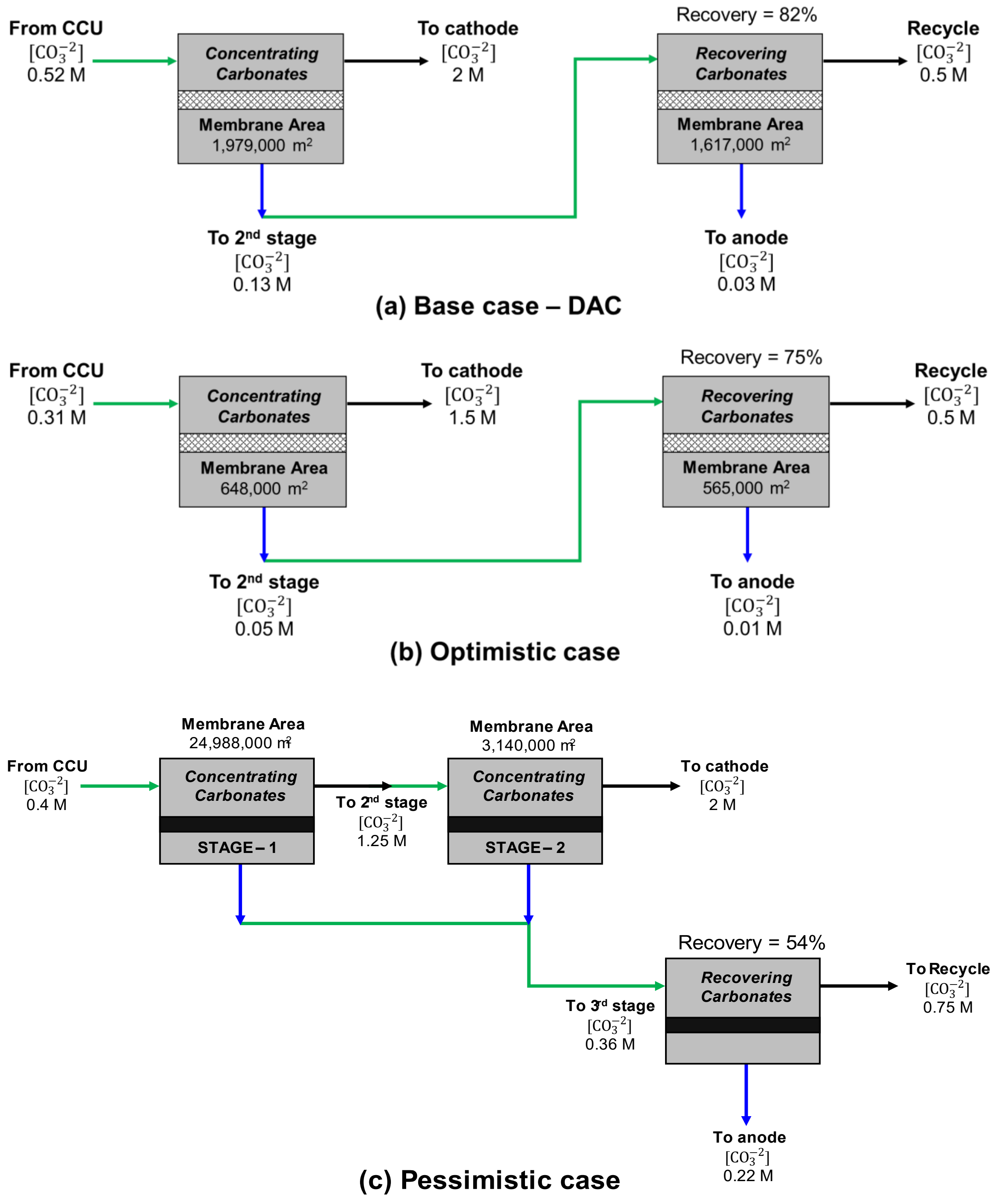


Figure 3. (a-c) ASPEN simulation results showing the carbonate concentration after each GO membrane stage, and the corresponding membrane area required for each stage. Membrane operation is at 75°C at 50 bar. All results shown here are for the DAC case. The measured concentration dependences of the rejection and flux from **Supplementary Fig. S2** are used in the membrane model to simulate the pessimistic case.

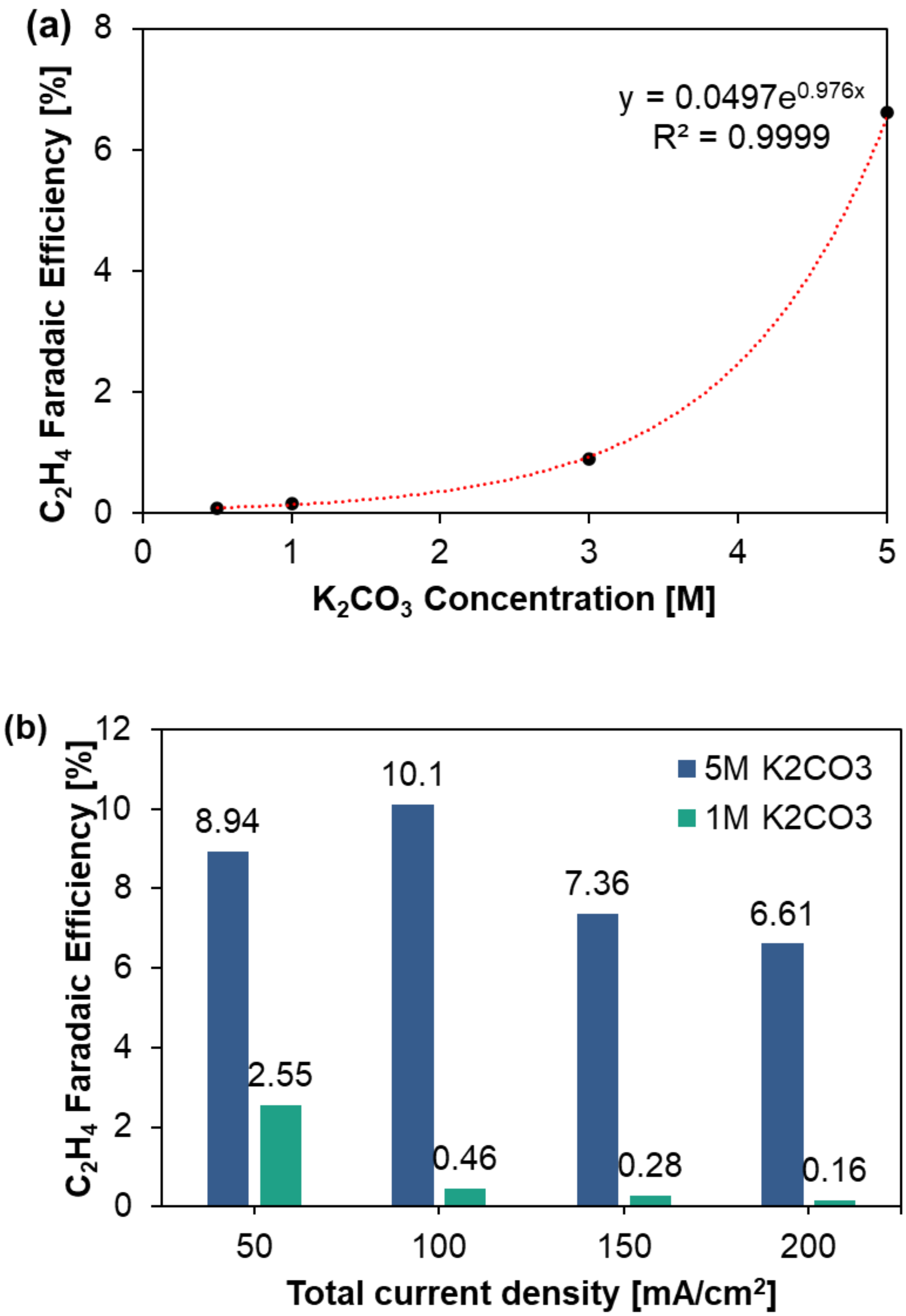


Figure 4. (a) Ethylene faradaic efficiency as a function of molar concentration of K_2CO_3 obtained from our experimental MEA measurements at 130 mL/min feed flow rate, IDF flow geometry at 200 $mAcm^{-2}$. **(b)** Ethylene faradaic efficiency as a function of current density at 1 M, 5 M concentration of K_2CO_3 obtained from our experimental measurements at 130 mL/min catholyte flow rate, 298 K. Data source: Song et al.[59]

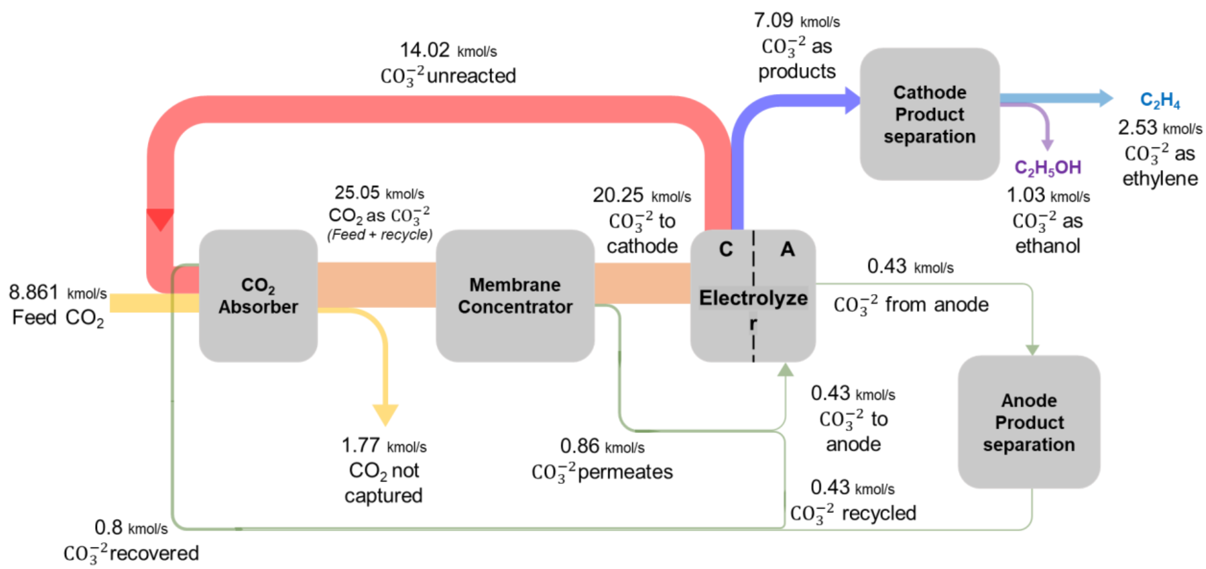


Figure 5. Carbon utilization Sankey diagram of the process, for 2 MMT/yr of ethylene production corresponding to the base case scenario in **Supplementary Table 1**.

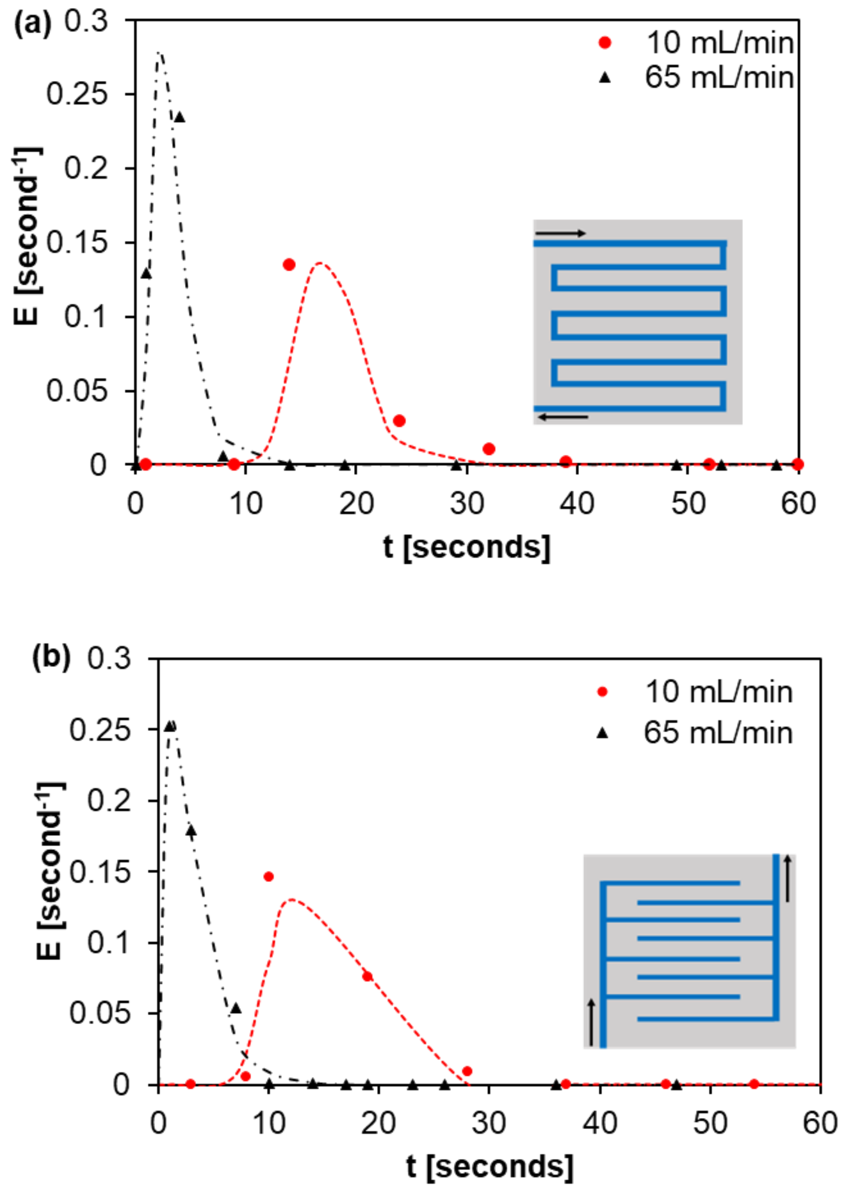


Figure 6. Exit age distributions *E(t)* for the laboratory-scale BPM MEA cell at 10 and 65 mL/min flowrates for **(a)** SF and **(b)** IDF geometries. Experimental data are shown as solid symbols. Fits to the open-vessel dispersion model (see **Supplementary Table 6**) are shown as solid lines. The insets show top views of SF and IDF flow plate geometries.

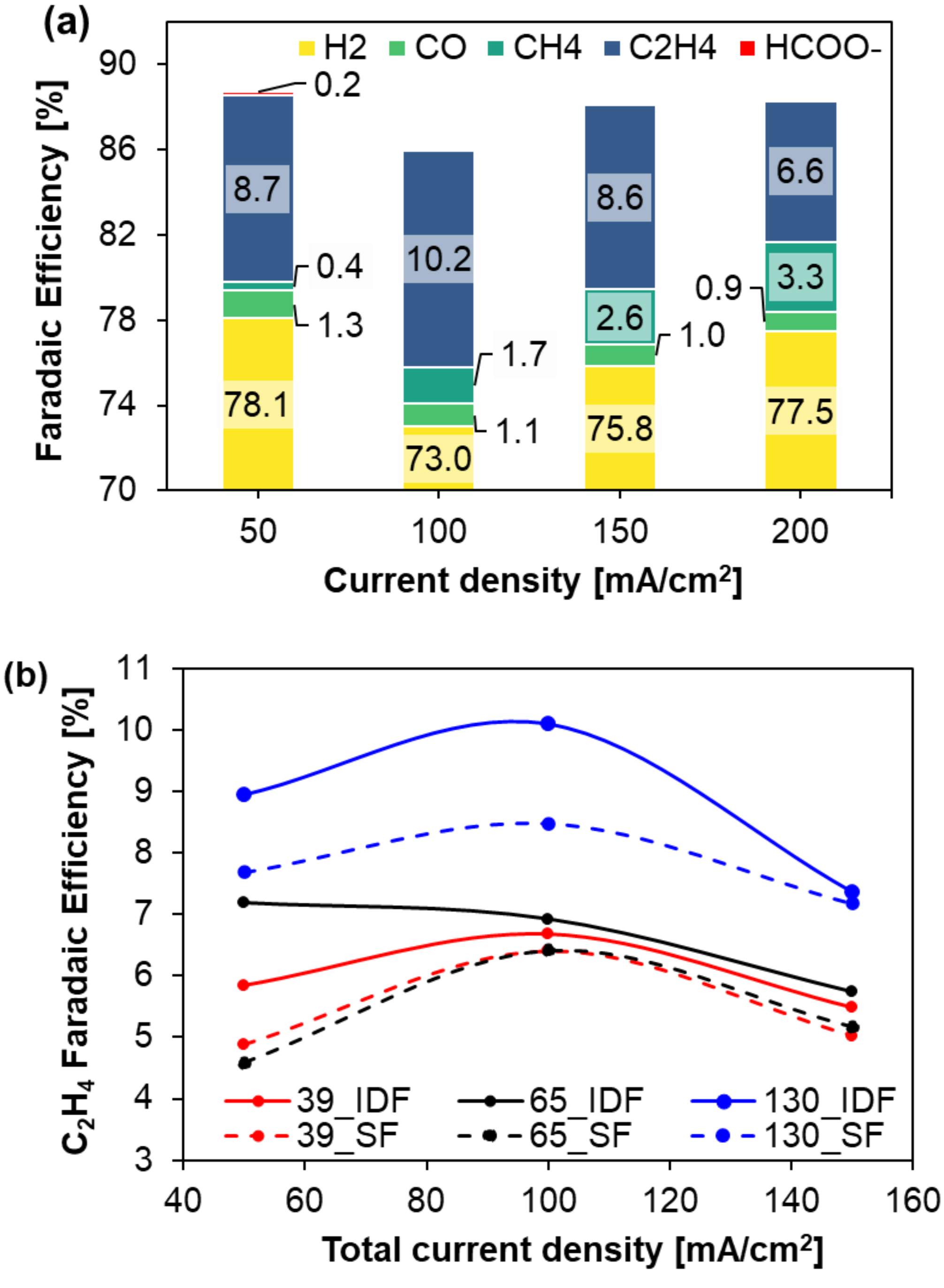


Figure 7. (a) Faradaic efficiencies of products observed at 65 mL/min, 5 M K_2CO_3 feed flow in IDF with a Cu catalyst (Data source: Song et al.[59]) **(b)** C_2H_4 faradaic efficiencies as a function of current density (50, 100, 150 mA cm^{-2}) and feed flow rate (39, 65, 130 mL/min) with Cu catalyst and 5 M K_2CO_3 solution at 298 K. **IDF:** Interdigitated flow pattern, **SF:** Serpentine flow pattern.

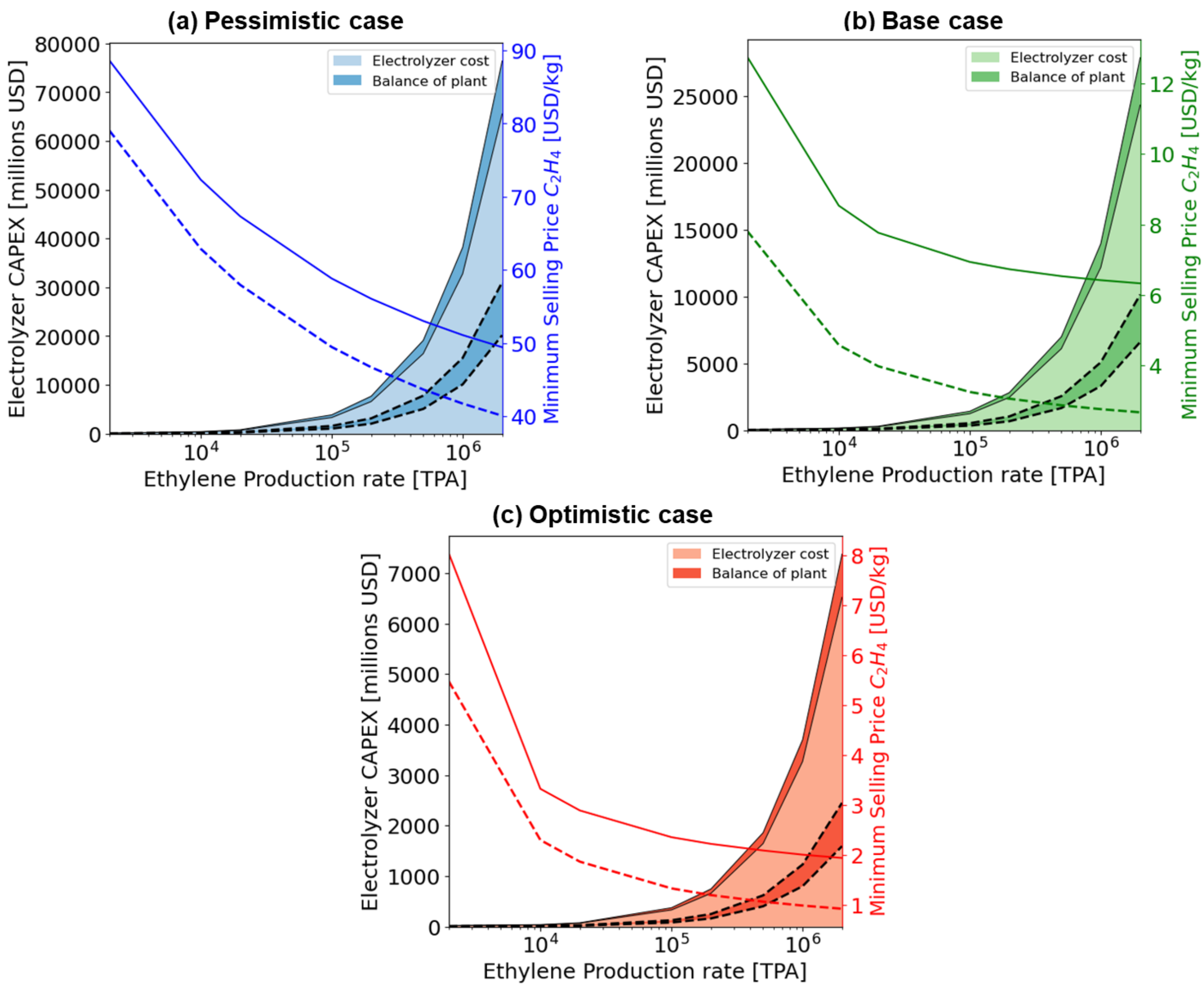


Figure 8. The comparison of capital cost of electrolyzer assuming linear scaling (solid lines) and a non-linear scaling (=0.85, dashed lines) and the corresponding full-plant ethylene MSP for: **(a)** Pessimistic **(b)** Base case **(c)** Optimistic summarized in **Supplementary Table 1**. It should be noted that the Balance of Plant costs are the same for both the scaled and unscaled scenarios. As shown in **Supplementary Table 7**, the BoP costs are computed as 35% of the scaled electrolyzer CAPEX.

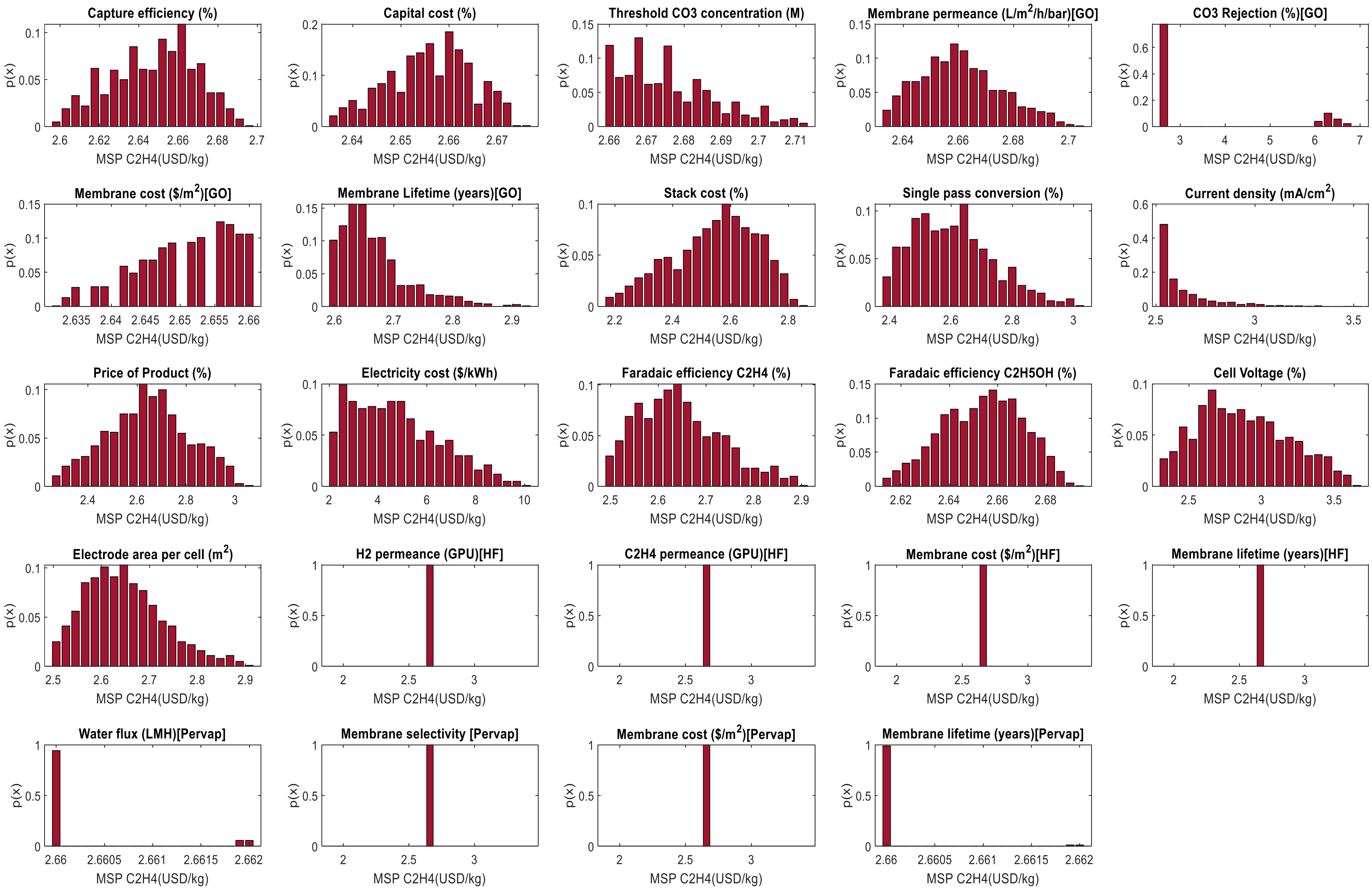


Figure 9. The MSP of ethylene when varying a given parameter, as simulated using the OAT Monte Carlo algorithm.

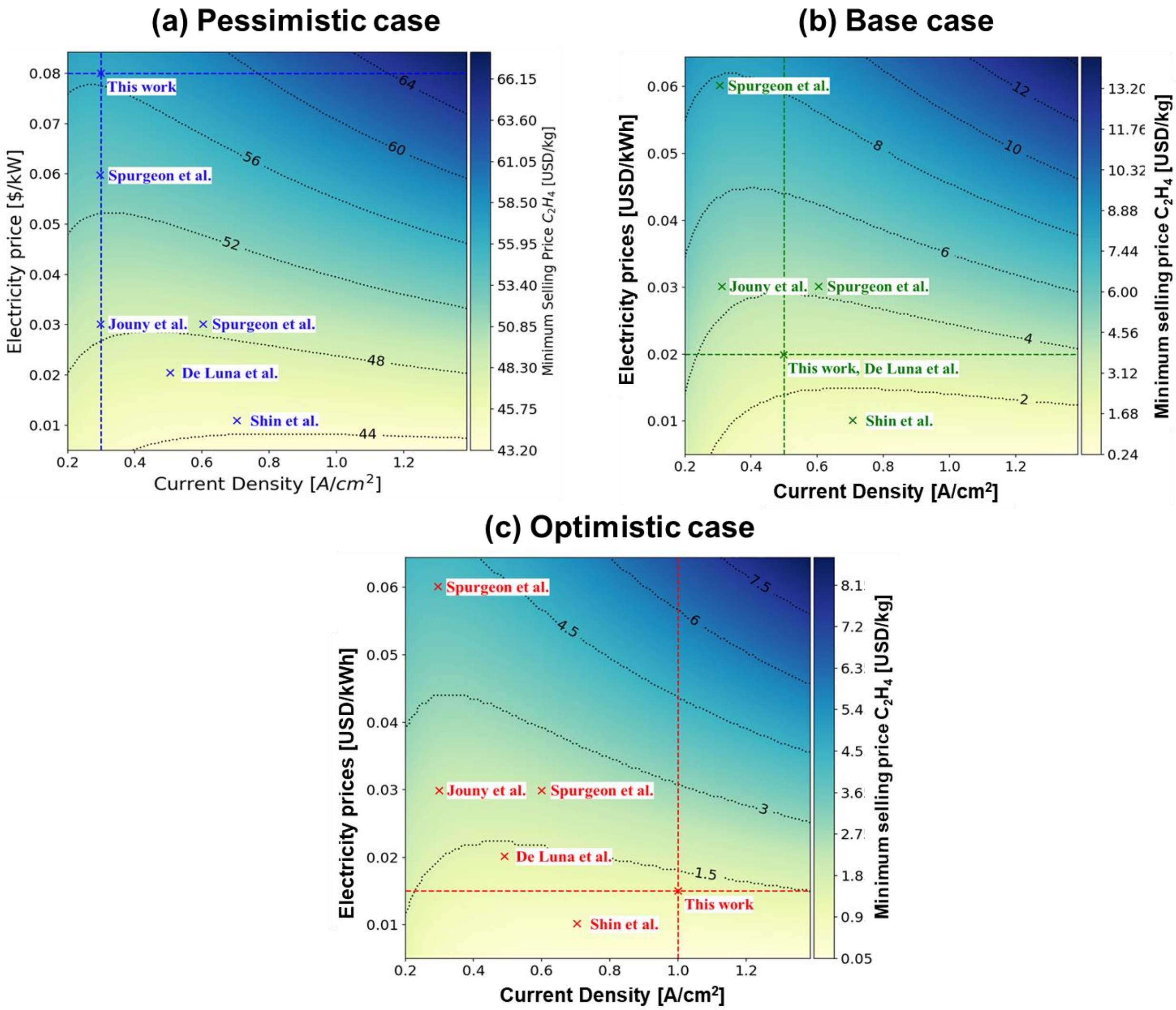


Figure 10. The variation of ethylene MSP as a function of electricity prices and current density for **(a)** Pessimistic **(b)** Base case **(c)** Optimistic scenarios. Various values of electricity prices assumed in the literature are also embedded in the plot for comparison.

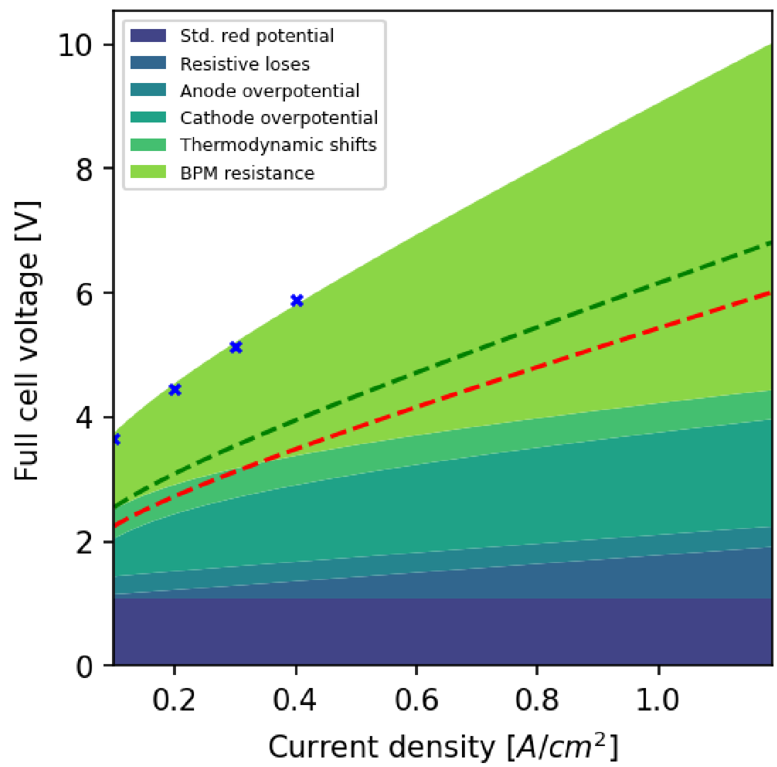


Figure 11. Full cell voltage breakdown predicted by the simplified voltametric model with the overlay of literature data. The voltages are predicted for a carbonate-fed BPM based electrolysis cell with 135 µm pH-modulating layer. The inset points depicted by blue 'x' marks are the experimentally obtained full cell voltages for a lab-scale electrolyzer of similar configuration by Lee et al.[5] The dotted line represent the cell voltage as a function of current density for base case and optimistic case (red, 30% lower than the model prediction).

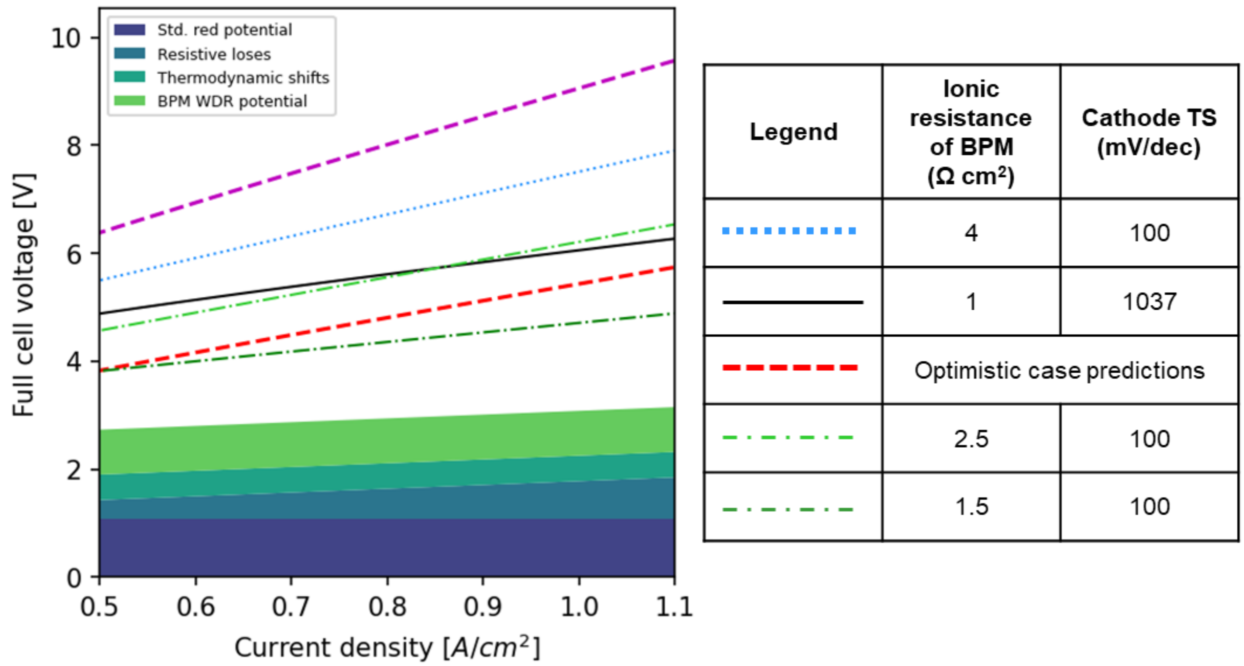


Figure 12. Variation of full cell voltage at higher current densities at different values of generalized cathodic Tafel slopes (TS) and ionic resistance of the BPM. Legends are shown in the inset table above. It can be clearly seen that the BPM ionic resistance is critical for reduction the full cell voltage of carbonate fed electrolyzers.

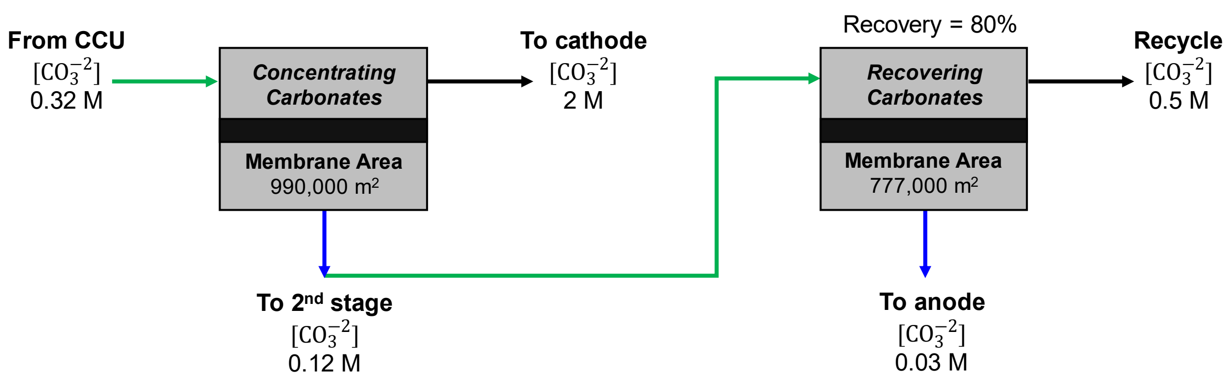


Figure 13. ASPEN simulation results showing the carbonate concentration after each GO membrane stage, and the corresponding membrane area required for each stage for the base case simulation for the flue gas capture. Membrane operation is at 65°C at 50 bar.

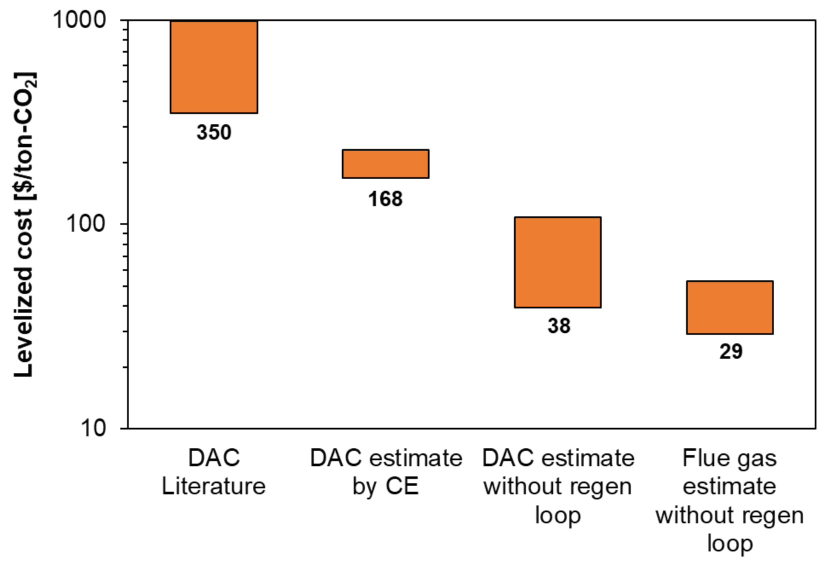


Figure 14. Variation of levelized cost of capturing CO_2 for DAC (literature), DAC (estimates by Carbon Engineering), DAC (our estimate for CO_2 capture as aqueous carbonate without need for the regeneration loop), and flue gas capture (our estimate for CO_2 capture as aqueous carbonate without need for the regeneration loop). The upper and lower bounds of the levelized cost in this figure for the last 3 cases correspond to cost recovery factors of 12.5% for the pessimistic and optimistic cases respectively & assuming a scaling factor of 0.85

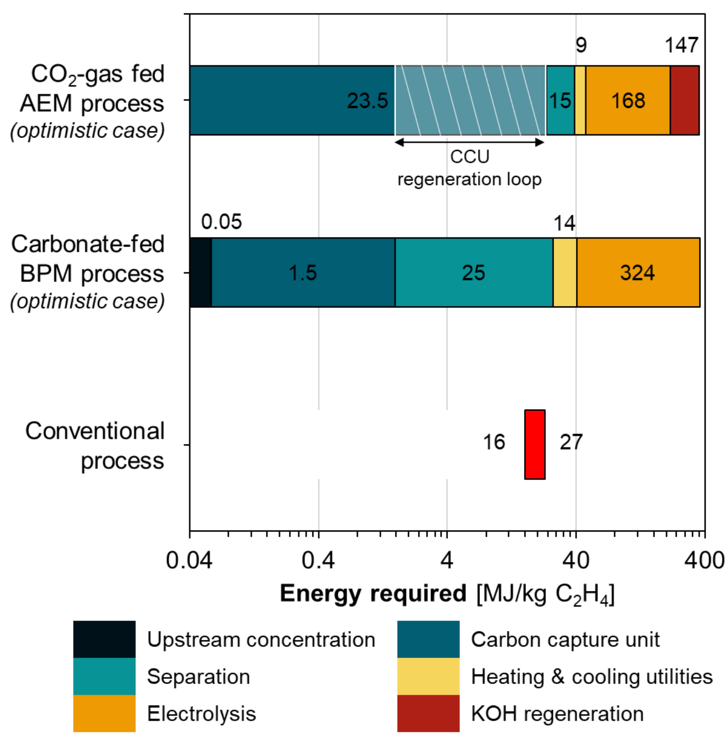


Figure 15. The energy requirement for the conventional process compared against the carbonate-fed BPM process and the CO_2 gas-fed electrolysis process.

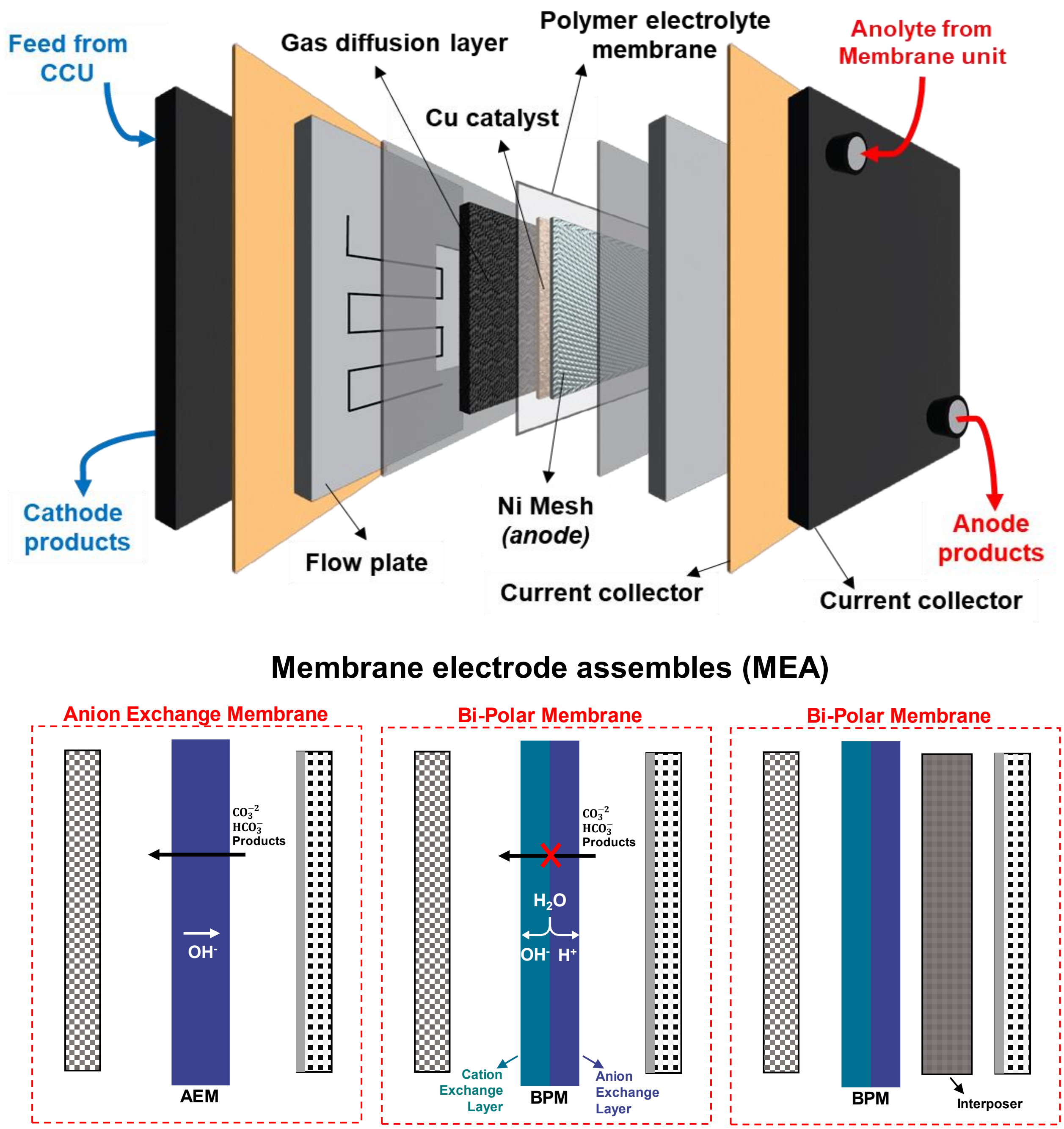


Figure 16. MEA-based architecture used in this work. The inset image depicts flow of species in the MEA with either an AEM or BPM solid polymer electrolyte membrane**.**

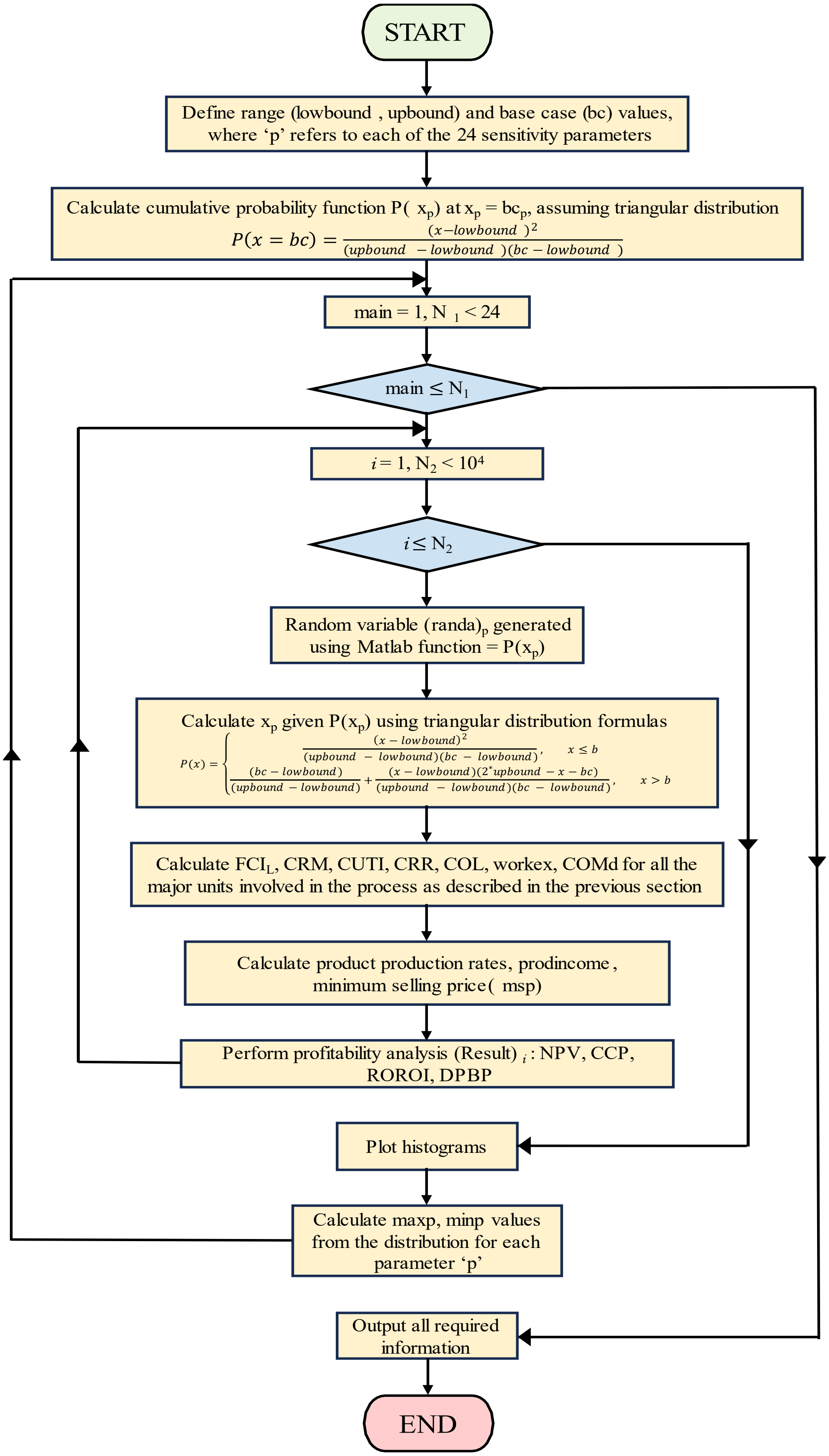
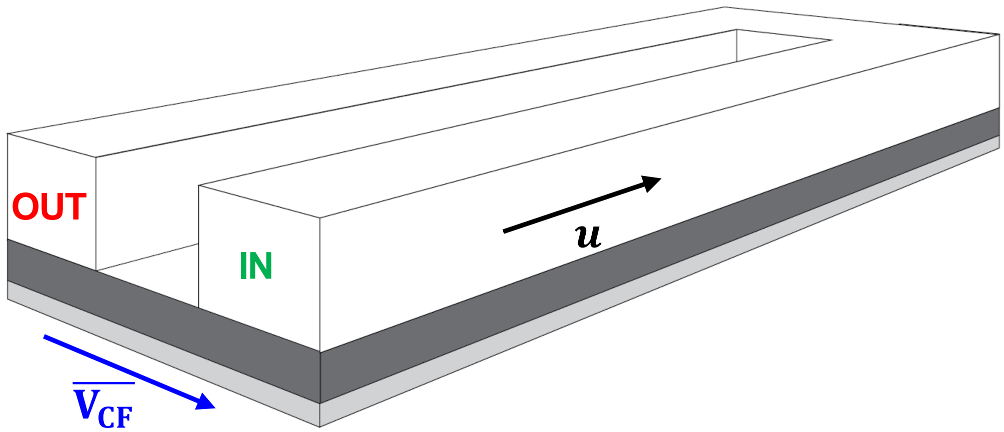
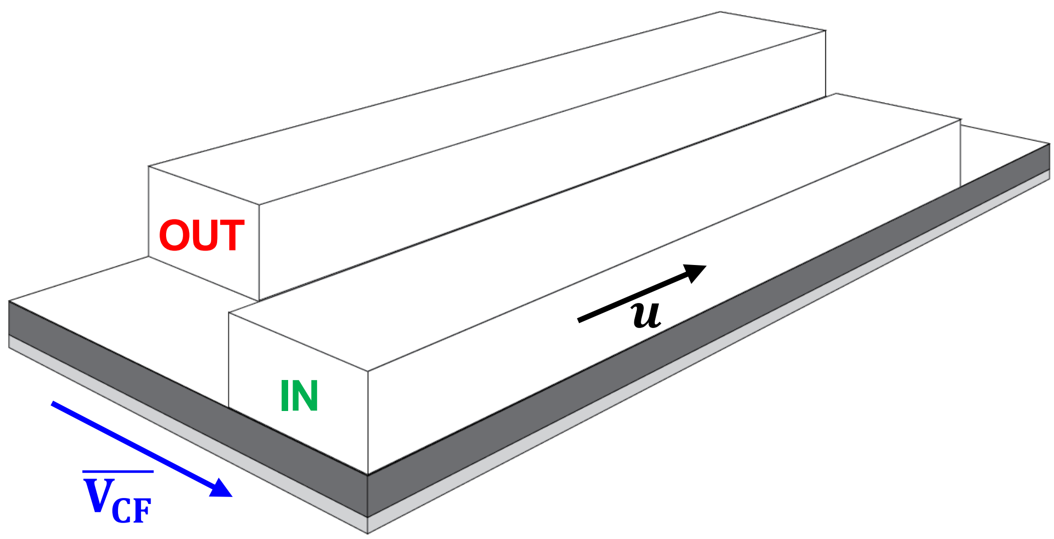


Figure 17. Algorithm used for the One-At-a-Time Monte Carlo Sensitivity Analysis



(a)



(b)

Figure 18. Computational domains for unit pair of **(a)** serpentine flow field and **(b)** interdigitated flow field channels, showing the GDL (darker gray layer) and CL (lighter gray layer).

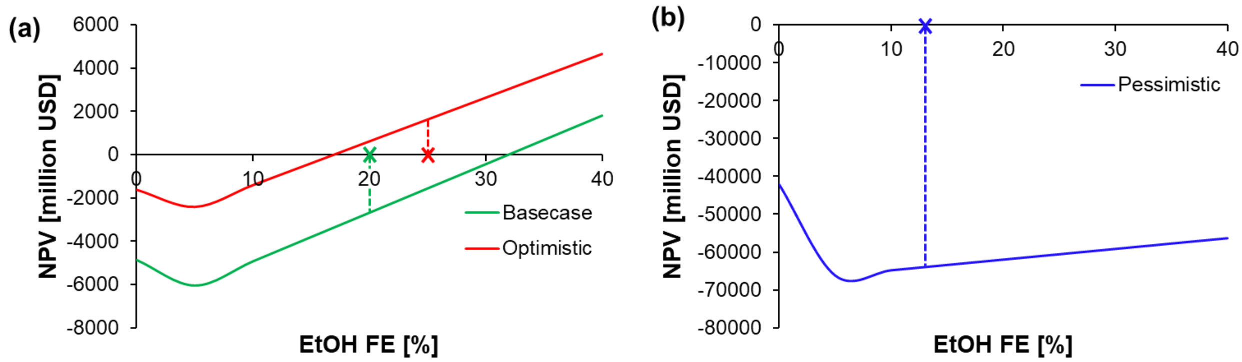


Figure 19. The NPV (in million USD) for the ethanol separation unit comprising of the distillation column and the pervaporation membrane units as a function of the ethanol FE at base case assumptions summarized in Supplementary Table 1.

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