Supporting Information

Techno-economic analysis and life-cycle assessment of the electrochemical conversion process with captured CO_2 in amine-based solvent

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Supplementary Notes

N1. Technoeconomic assumptions

CO_2 capture plants are mainly built for gas-fired power plants and coal-fired power plants. Therefore, we considered the power plant to be included in the economic evaluation boundary for a more reasonable evaluation. And the reference process is a subcritical pulverized coal-fired power plant (Ppc = 550 MW) as reported in a 2010 report by the US Department of Energy (Table S1) [1]. Although a CCS process that consumes less energy has been reported [2] than the corresponding NETL data, USDOE's 2010 report contains detailed information on the carbon capture process, and the information was cited in this paper because it is classic information.

N2. Calculation of electrolyzer capital cost

The electrolyzer cost estimation method is based on the DOE Future central H_2A base case for an alkaline electrolyzer (ver.2019) data [3]. Based on the data, uninstalled capital costs attributed to the stack component were set at \$233 kW^{-1}, typical operating conditions of electrolyzer were set at 3 A cm^{-2} and 1.8 V, and the installation factor was set at 1.2. The installed cost calculated based on that data is \$15,098 m^{-2}. In addition, the maintenance cost is 2.5% of the annual capital investment, and the balance of plant (BoP) cost is assumed to be 35% of the electrolyzer cell system cost.

The area of the electrolyzer was calculated by dividing the amount of current by the current density after calculating the required amount of current using the number of electrons required for the reaction. The installed cost of the electrolyzer in this process was calculated by multiplying the obtained area of the electrolyzer with the installed cost per area of the electrolyzer calculated using the DOE H_2A PEM electrolysis cell data [3]. The calculation results are tabulated in Table S19 and S20.

Calculation of required partial current (A) :

$$3238168\,\frac{kg}{day}\times\frac{day}{86400s}\times 1000\frac{g}{kg}\times\frac{mol}{28.01\,g}\times 2\,e^{-}\times 96480\frac{C}{mol}=\ 258190273\,A$$

Calculation of required total current (A):

$$\frac{Required\ partial\ current\ (A)}{Faradaic\ efficiency}=\frac{258190273\,A}{0.99}=\ 260798256\,A$$

Calculation of required electrolyzer area (m^2)

$$\frac{Required\ total\ current\ (A)}{Current\ density\left(\frac{A}{cm^2}\right)\times\frac{10^4\ cm^2}{m^2}}=\frac{260798256\ A}{0.2}=130399\ m^2$$

Calculation of required power (MW):

$$Required\ total\ current\ (A)\times cell\ voltage\ (V)=\ 260798256\times 2.29\ \times\ 10^{-6}$$

$$=597\ MW$$

Calculated electrolyzer cost (\$)

$$Required\ electrolyzer\ area\ (m^2)\times Reference\ PEM\ electrolyzer\ purchase\ cost\left(\frac{\$}{m^2}\right)$$
$$=130399\times 12582=\ 1{,}640{,}681{,}832$$

N3. Property parameters

1) Pure component properties - Temperature dependent correlation parameters (Table S6-S13)

- Aqueous infinite dilution heat capacity (Parameter: CPAQ0) [4]

$$C_{p,i}^{\infty,aq}\left(\frac{J}{kmol\cdot K}\right)=\ C_1+C_2T(K)+C_3(T(K))^2+\frac{C_{4i}}{T(K)}+\frac{C_{5i}}{T(K)^2}+\frac{C_{6i}}{\sqrt{T(K)}}$$

$$for\ C_{7i}\leq T\leq C_{8i}$$

- Pure component dielectric constant coefficients of nonaqueous solvents (CPDIEC) [5]

$$\varepsilon_i(T)=\ A_i+B_i((1/T)-(1/C_i))$$

- Ideal gas heat capacity (CPIG) [4]

$$C_p^{*,ig}=\ C_{1i}+C_{2i}T+C_{3i}T^2+C_{4i}T^3+C_{5i}T^4+C_{6i}T^5\ for\ C_{7i}\leq T\leq C_{8i}$$

$$C_p^{*,ig}=\ C_{9i}+C_{10i}T^{C_{11i}}\ for\ T<\ C_{7i}$$

- Coefficients for the Jones-Dole correction to liquid mixture viscosity due to the presence of electrolytes (moles) (IONMOB) [4]

$$\Delta\eta_{ca}^{\iota}=A_{ca}\sqrt{C_{ca}^{a}}+B_{ca}\sqrt{C_{ca}^{a}}$$

- Coefficients for the Jones-Dole correction to liquid mixture viscosity due to the presence of electrolytes (volume/mole) (IONMUB) [4]

$$\Delta\eta^{l}_{ca} = A_{ca}\sqrt{C^{a}_{ca}} + B_{ca}\sqrt{C^{a}_{ca}}$$

- Coefficients for the extended antoine vapor pressure equation for a liquid (PLXANT) [6]

$$\ln p_i^{*,l} = C_{1i} + \frac{C_{2i}}{T + C_{3i}} + C_{4i}T + C_{5i}\ln T + C_{6i}T^{C_{7i}} \quad for\ C_{8i} \le T \le C_{9i}$$

- Brelvi-O-connell volume parameter (VLBROC) [4]

$$V^{\infty}_{iA} = fcn\left(V^{BO}_{i}, V^{BO}_{A}, V^{*l}_{A}\right)$$

2) Temperature dependent binary interaction parameters (Table S14-S18)

- Henry's constant (HENRY) [4, 7]

$$\ln\left(H_i/\gamma^{\infty}_{i}\right) = \sum_{A}\omega_A \ln\left(H_{iA}/\gamma^{\infty}_{iA}\right)$$

- Non-random two-liquid (NRTL) [5, 7]

$$\ln\gamma_i = \frac{\sum_{j} x_j\tau_{ji}G_{ji}}{\sum_{k}^{n} x_k G_{ki}} + \sum_{j}^{n}\frac{x_j G_{ij}}{\sum_{k}^{n} x_k G_{kj}}\left(\tau_{ij} - \frac{\sum_{m}^{n} x_m\tau_{mj}G_{mj}}{\sum_{k}^{n} x_k G_{kj}}\right)$$

- Liquid mixture viscosity (MUKIJ, MULIJ) [4]

$$\ln\eta^{l} = \sum_{i=1}^{n} X_i \ln\eta^{*,l} + \sum_{i=1}^{n}\sum_{j>i}^{n} k_{ij}X_iX_j\ln\eta_{ij} + \sum_{i=1}^{n} X_i\left[\sum_{j\neq i}^{n} X_j\left(l_{ij}\ln\eta_{ij}\right)^{1/3}\right]^{3}$$

- Clarke aqueous electrolyte volume (VLCLK) [4]

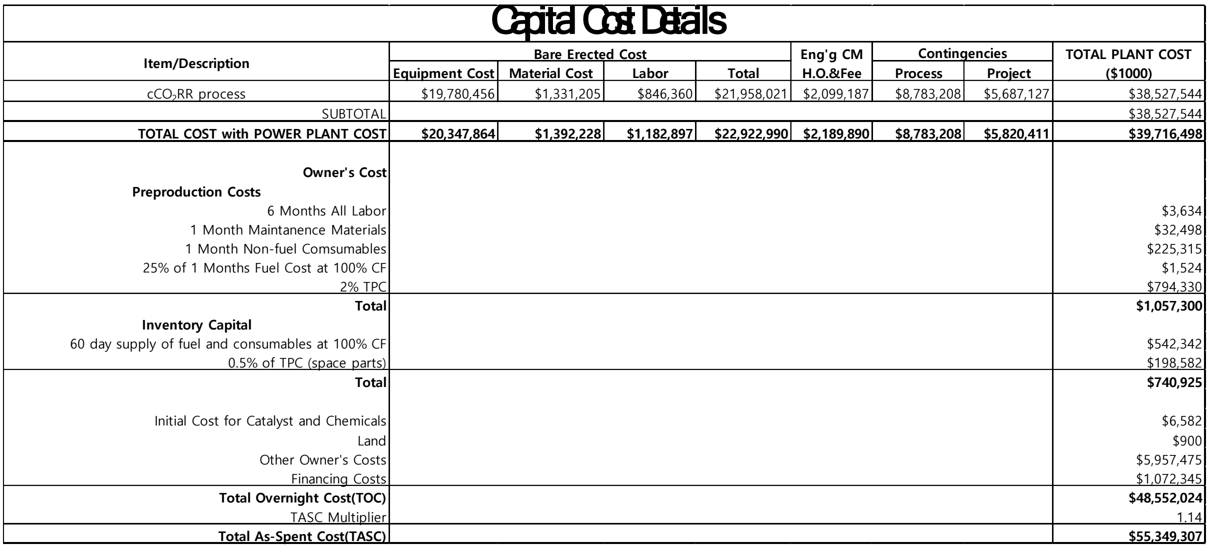
$$V_{ca} = V^{\infty}_{ca} + A_{ca}\frac{\sqrt{x_{solute}}}{1 + \sqrt{x_{solute}}}$$

N4. Technoeconomic analysis result

Techno-economic evaluation using the results obtained through each cCO_2RR and CO_2RR process simulation under the current and optimistic condition are as follows. The underlying framework was referenced in NETL Cost Estimation Methods [1, 8].

- Total plant cost details

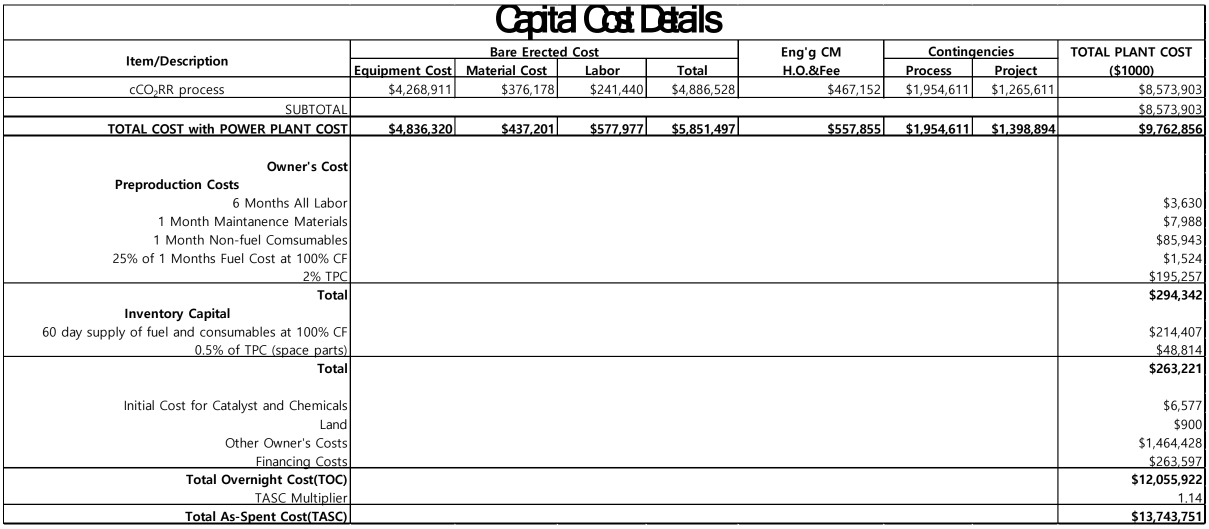
1) cCO_2RR (Current)



Capital Cost Details

Item/Description	Bare Erected Cost				Eng'g CM	Contingencies		TOTAL PLANT COST
	Equipment Cost	Material Cost	Labor	Total	H.O.&Fee	Process	Project	($1000)
cCO_2RR process	$19,780,456	$1,331,205	$846,360	$21,958,021	$2,099,187	$8,783,208	$5,687,127	$38,527,544
SUBTOTAL								$38,527,544
TOTAL COST with POWER PLANT COST	**$20,347,864**	**$1,392,228**	**$1,182,897**	**$22,922,990**	**$2,189,890**	**$8,783,208**	**$5,820,411**	**$39,716,498**
Owner's Cost								
Preproduction Costs								
6 Months All Labor								$3,634
1 Month Maintanence Materials								$32,498
1 Month Non-fuel Comsumables								$225,315
25% of 1 Months Fuel Cost at 100% CF								$1,524
2% TPC								$794,330
Total								**$1,057,300**
Inventory Capital								
60 day supply of fuel and consumables at 100% CF								$542,342
0.5% of TPC (space parts)								$198,582
Total								**$740,925**
Initial Cost for Catalyst and Chemicals								$6,582
Land								$900
Other Owner's Costs								$5,957,475
Financing Costs								$1,072,345
Total Overnight Cost(TOC)								**$48,552,024**
TASC Multiplier								1.14
Total As-Spent Cost(TASC)								**$55,349,307**

2) cCO_2RR (Optimistic)



Capital Cost Details

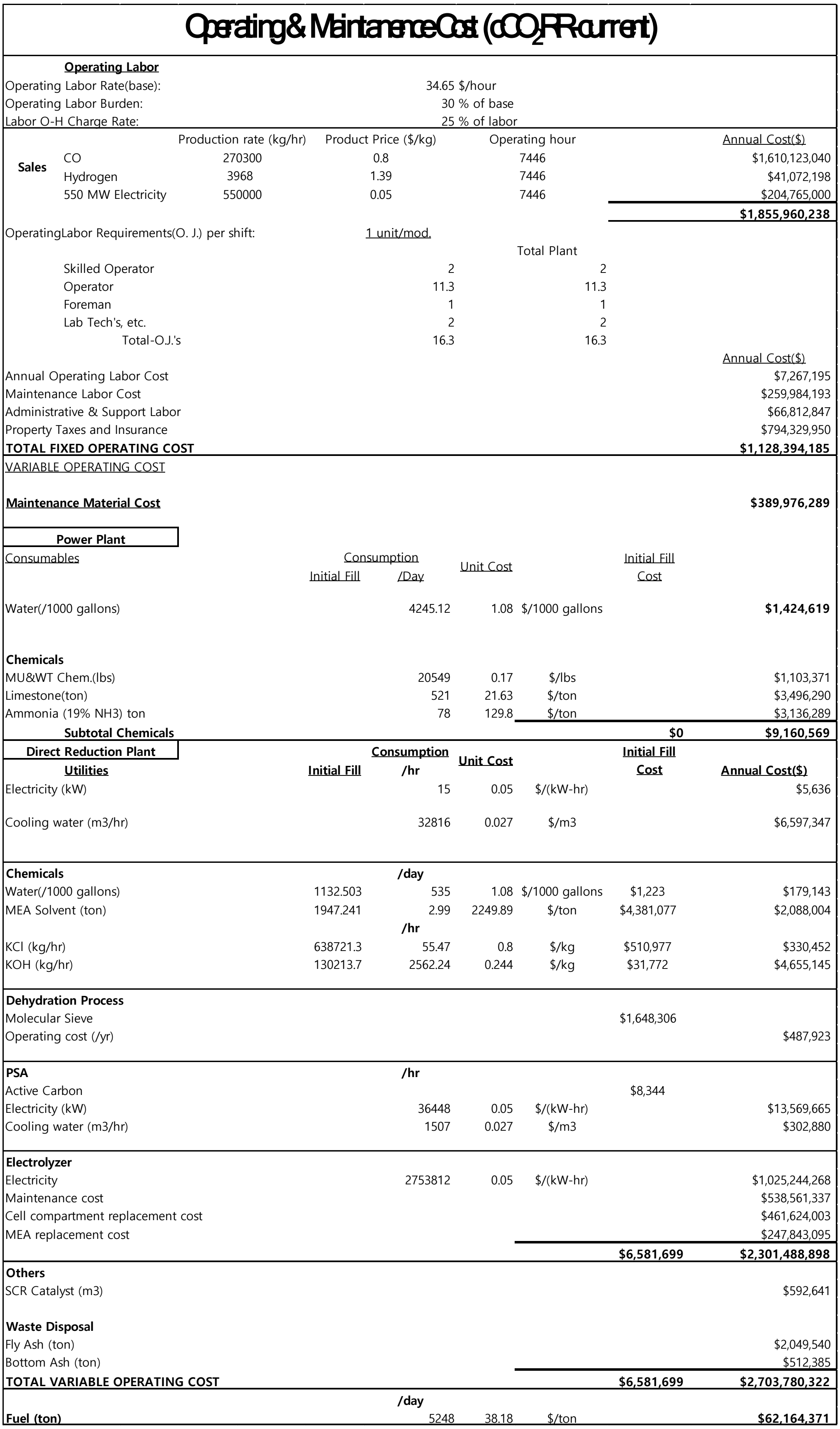
Item/Description	Bare Erected Cost				Eng'g CM	Contingencies		TOTAL PLANT COST
	Equipment Cost	Material Cost	Labor	Total	H.O.&Fee	Process	Project	($1000)
cCO_2RR process	$4,268,911	$376,178	$241,440	$4,886,528	$467,152	$1,954,611	$1,265,611	$8,573,903
SUBTOTAL								$8,573,903
TOTAL COST with POWER PLANT COST	**$4,836,320**	**$437,201**	**$577,977**	**$5,851,497**	**$557,855**	**$1,954,611**	**$1,398,894**	**$9,762,856**
Owner's Cost								
Preproduction Costs								
6 Months All Labor								$3,630
1 Month Maintanence Materials								$7,988
1 Month Non-fuel Comsumables								$85,943
25% of 1 Months Fuel Cost at 100% CF								$1,524
2% TPC								$195,257
Total								**$294,342**
Inventory Capital								
60 day supply of fuel and consumables at 100% CF								$214,407
0.5% of TPC (space parts)								$48,814
Total								**$263,221**
Initial Cost for Catalyst and Chemicals								$6,577
Land								$900
Other Owner's Costs								$1,464,428
Financing Costs								$263,597
Total Overnight Cost(TOC)								**$12,055,922**
TASC Multiplier								1.14
Total As-Spent Cost(TASC)								**$13,743,751**

3) CO$_2$RR (Current)

Capital Cost Details

Acct No.	Item/Description	Bare Erected Cost				Eng'g CM	Contingencies		TOTAL PLANT COST
		Equipment Cost	Material Cost	Labor	Total	H.O.&Fee	Process	Project	($1000)
1	CO$_2$ Capture System	$155,219	$95,528	$64,060	$314,807	$30,096	$62,961	$81,535	$489,399
2	CO$_2$ Utilization System	$4,148,794	$304,152	$193,152	$4,646,098	$444,167	$1,858,439	$1,203,339	$8,152,044
	SUBTOTAL	$4,304,013	$399,679	$257,213	$4,960,905	$474,263	$1,921,401	$1,284,874	$8,641,442
	TOTAL COST with POWER PLANT COST	**$4,871,422**	**$460,702**	**$593,750**	**$5,925,874**	**$564,966**	**$1,921,401**	**$1,418,158**	**$9,830,396**
	Owner's Cost								
	Preproduction Costs								
	6 Months All Labor								$3,521
	1 Month Maintanence Materials								$8,044
	1 Month Non-fuel Comsumables								$98,809
	25% of 1 Months Fuel Cost at 100% CF								$1,524
	2% TPC								$196,608
	Total								**$308,505**
	Inventory Capital								
	60 day supply of fuel and consumables at 100% CF								$244,680
	0.5% of TPC (space parts)								$49,152
	Total								**$293,832**
	Initial Cost for Catalyst and Chemicals								$4,431
	Land								$900
	Other Owner's Costs								$1,474,559
	Financing Costs								$265,421
	Total Overnight Cost(TOC)								**$12,178,045**
	TASC Multiplier								1.14
	Total As-Spent Cost(TASC)								**$13,882,971**

- Initial and annual operating and maintenance costs - 1) cCO_2RR (Current)



Operating & Maintanence Cost (cCO_2RR-current)

Operating Labor

Operating Labor Rate(base): 34.65 $/hour
Operating Labor Burden: 30 % of base
Labor O-H Charge Rate: 25 % of labor

		Production rate (kg/hr)	Product Price ($/kg)	Operating hour	Annual Cost($)
Sales	CO	270300	0.8	7446	$1,610,123,040
	Hydrogen	3968	1.39	7446	$41,072,198
	550 MW Electricity	550000	0.05	7446	$204,765,000
					$1,855,960,238

OperatingLabor Requirements(O. J.) per shift:	1 unit/mod.	Total Plant
Skilled Operator	2	2
Operator	11.3	11.3
Foreman	1	1
Lab Tech's, etc.	2	2
Total-O.J.'s	16.3	16.3

	Annual Cost($)
Annual Operating Labor Cost	$7,267,195
Maintenance Labor Cost	$259,984,193
Administrative & Support Labor	$66,812,847
Property Taxes and Insurance	$794,329,950
TOTAL FIXED OPERATING COST	**$1,128,394,185**

VARIABLE OPERATING COST

Maintenance Material Cost	**$389,976,289**

Power Plant						
Consumables	Initial Fill	Consumption /Day	Unit Cost		Initial Fill Cost	
Water(/1000 gallons)		4245.12	1.08	$/1000 gallons		**$1,424,619**
Chemicals						
MU&WT Chem.(lbs)		20549	0.17	$/lbs		$1,103,371
Limestone(ton)		521	21.63	$/ton		$3,496,290
Ammonia (19% NH3) ton		78	129.8	$/ton		$3,136,289
Subtotal Chemicals					**$0**	**$9,160,569**
Direct Reduction Plant						
Utilities	**Initial Fill**	**Consumption /hr**	**Unit Cost**		**Initial Fill Cost**	**Annual Cost($)**
Electricity (kW)		15	0.05	$/(kW-hr)		$5,636
Cooling water (m3/hr)		32816	0.027	$/m3		$6,597,347
Chemicals		**/day**				
Water(/1000 gallons)	1132.503	535	1.08	$/1000 gallons	$1,223	$179,143
MEA Solvent (ton)	1947.241	2.99	2249.89	$/ton	$4,381,077	$2,088,004
		/hr				
KCl (kg/hr)	638721.3	55.47	0.8	$/kg	$510,977	$330,452
KOH (kg/hr)	130213.7	2562.24	0.244	$/kg	$31,772	$4,655,145
Dehydration Process						
Molecular Sieve					$1,648,306	
Operating cost (/yr)						$487,923
PSA		**/hr**				
Active Carbon					$8,344	
Electricity (kW)		36448	0.05	$/(kW-hr)		$13,569,665
Cooling water (m3/hr)		1507	0.027	$/m3		$302,880
Electrolyzer						
Electricity		2753812	0.05	$/(kW-hr)		$1,025,244,268
Maintenance cost						$538,561,337
Cell compartment replacement cost						$461,624,003
MEA replacement cost						$247,843,095
					$6,581,699	**$2,301,488,898**
Others						
SCR Catalyst (m3)						$592,641
Waste Disposal						
Fly Ash (ton)						$2,049,540
Bottom Ash (ton)						$512,385
TOTAL VARIABLE OPERATING COST					**$6,581,699**	**$2,703,780,322**
		/day				
Fuel (ton)		5248	38.18	$/ton		**$62,164,371**

2) cCO_2RR (Optimistic)

Operating & Maintanence Cost (cCO_2RR-optimistic)

Operating Labor

Operating Labor Rate(base): 34.65 $/hour
Operating Labor Burden: 30 % of base
Labor O-H Charge Rate: 25 % of labor

		Production rate (kg/hr)	Product Price ($/kg)	Operating hour	Annual Cost($)
Sales	CO	270300	0.8	7446	$1,610,123,040
	Hydrogen	1477	1.39	7446	$15,288,275
	550 MW Electricity	550000	0.05	7446	$204,765,000
					$1,830,176,315

OperatingLabor Requirements(O. J.) per shift:	1 unit/mod.	Total Plant
Skilled Operator	2	2
Operator	11.3	11.3
Foreman	1	1
Lab Tech's, etc.	2	2
Total-O.J.'s	16.3	16.3

	Annual Cost($)
Annual Operating Labor Cost	$7,260,384
Maintenance Labor Cost	$63,907,657
Administrative & Support Labor	$17,792,010
Property Taxes and Insurance	$195,257,126
TOTAL FIXED OPERATING COST	**$284,217,177**

VARIABLE OPERATING COST

Maintenance Material Cost						**$95,861,486**
Power Plant						
Consumables	**Initial Fill**	**Consumption /Day**	**Unit Cost**		**Initial Fill Cost**	
Water(/1000 gallons)		4245.12	1.08	$/1000 gallons		**$1,424,619**
Chemicals						
MU&WT Chem.(lbs)		20549	0.17	$/lbs		$1,103,371
Limestone(ton)		521	21.63	$/ton		$3,496,290
Ammonia (19% NH3) ton		78	129.8	$/ton		$3,136,289
Subtotal Chemicals					**$0**	**$9,160,569**
Direct Reduction Plant		**Consumption**				
Utilities	**Initial Fill**	**/hr**	**Unit Cost**		**Initial Fill Cost**	**Annual Cost($)**
Electricity (kW)		15	0.05	$/(kW-hr)		$5,636
Cooling water (m3/hr)		32905	0.027	$/m3		$6,615,290
Chemicals		**/day**				
Water(/1000 gallons)	1132.496	272	1.08	$/1000 gallons	$1,223	$90,982
MEA Solvent (ton)	1947.394	2.98	2249.89	$/ton	$4,381,423	$2,079,844
		/hr				
KCl (kg/hr)	638716.7	55.25	0.8	$/kg	$510,973	$329,137
KOH (kg/hr)	108519.7	356.23	0.244	$/kg	$26,479	$647,200
Dehydration Process						
Molecular Sieve					$1,648,332	
Operating cost (/yr)						$487,946
PSA		**/hr**				
Active Carbon					$8,075	
Electricity (kW)		31585	0.05	$/(kW-hr)		$11,759,163
Cooling water (m3/hr)		1311	0.027	$/m3		$263,572
Electrolyzer						
Electricity		1721272	0.05	$/(kW-hr)		$640,829,509
Maintenance cost						$112,209,356
Cell compartment replacement cost						$96,179,448
MEA replacement cost						$51,638,155
					$6,576,505	**$923,135,238**
Others						
SCR Catalyst (m3)						$592,641
Waste Disposal						
Fly Ash (ton)						$2,049,540
Bottom Ash (ton)						$512,385
TOTAL VARIABLE OPERATING COST					**$6,576,505**	**$1,031,311,859**
		/day				
Fuel (ton)		5248	38.18	$/ton		**$62,164,371**

3) CO_2RR

Operating & Maintanence Cost (CO_2RR)

Operating Labor						
Operating Labor Rate(base):			34.65 $/hour			
Operating Labor Burden:			30 % of base			
Labor O-H Charge Rate:			25 % of labor			
	Production rate (kg/hr)	Product Price ($/kg)	Operating hour			Annual Cost($)
Sales CO	270300	0.8	7446			$1,610,122,294
Hydrogen	1477	1.39	7446			$15,283,727
550 MW Electricity	550000	0.05	7446			$204,765,000
						$1,830,171,021
OperatingLabor Requirements(O. J.) per shift:		1 unit/mod.				
			Total Plant			
Skilled Operator		2		2		
Operator		11.3		11.3		
Foreman		1		1		
Lab Tech's, etc.		2		2		
Total-O.J.'s		16.3		16.3		
						Annual Cost($)
Annual Operating Labor Cost						$7,041,805
Maintenance Labor Cost						$64,349,774
Administrative & Support Labor						$17,847,895
Property Taxes and Insurance						$196,607,926
TOTAL FIXED OPERATING COST						**$285,847,400**
VARIABLE OPERATING COST						
Maintenance Material Cost						**$96,524,661**
Power Plant						
Consumables	**Consumption**		**Unit Cost**		**Initial Fill**	
	Initial Fill	**/Day**			**Cost**	
Water(/1000 gallons)		4245.12	1.08	$/1000 gallons		**$1,424,619**
Chemicals						
MU&WT Chem.(lbs)		20549.00	0.17	$/lbs		$1,103,371
Limestone(ton)		521.00	21.63	$/ton		$3,496,290
Ammonia (19% NH3) ton		77.88	129.8	$/ton		$3,136,289
Subtotal Chemicals					**$0**	**$9,160,569**
Capture Plant		/hr	Unit Cost			
Utilities	Initial Fill					
Steam, 450psig (kg/hr)		963170	0.0176	$/kg		$126,223,043
Electricity (kW)		320	0.05	$/(kW-hr)		$119,245
Cooling water (m3/hr)		72421	0.027	$/m3		$14,559,562
			Utilities			**$140,901,850**
Chemicals		**/Day**				
Water(/1000 gallons)	1055.457	227.05	1.08	$/1000 gallons	$1,140	$76,077
MEA Solvent (ton)	1964.807	2.60	2249.89	$/ton	$4,420,600	$1,817,982
			Chemicals		**$4,421,740**	**$1,894,059**
CO_2 Utilize Plant		/hr	Unit Cost			
Utilities	Initial Fill					
Electricity (kW)		31265	0.05	$/(kW-hr)		$11,640,012
Cooling water (m3/hr)		5513	0.027	$/m3		$1,108,263
			Utilities			**$12,748,275**
Chemicals	Initial Fill	**/hr**				
Water(/1000 gallons)		10	1.08	$/1000 gallons		$76,828
KOH (kg/hr)	2035.14	20.35	0.8	$/kg	$1,628	$121,229
			Chemicals		**$1,628**	**$198,057**
PSA		**/hr**				
Active Carbon					**$8,075**	
Capture process						
Operating cost (/yr)						**$21,028,534**
Electrolyzer						
Electricity		1719816	0.05	$/(kW-hr)		**$640,287,452**
Maintenance cost						$112,114,442
Cell compartment replacement cost						$96,098,093
MEA replacement cost						$51,594,476
					$8,075	**$921,122,997**
Others						
SCR Catalyst (m3)						$592,641
Waste Disposal						
Fly Ash (ton)						$2,049,540
Bottom Ash (ton)						$512,385
TOTAL VARIABLE OPERATING COST					**$4,431,443**	**$1,185,705,034**
		/day				
Fuel (ton)		5248	38.18	$/ton		**$62,164,371**

Supplementary Tables

Table S1. CO_2 source process technoeconomic assumption parameters [1].

Parameter	**Value**
Year of cost data	2007[a]
Year of publication	2010
Organization	NETL
Region	US
Specific fuel type	Bitcoal
Gross plant power (MWe)	583
Auxiliary load (MWe)	33
Net plant power (MWe)	550
Net plant efficiency (HHV,%)	36.8
Net plant heat rate (BTU/kWh)	9,277
CO_2 emissions w/o capture (kg/MWh)	856
Capital cost w/o capture (USD/kW)	1,996
Total fixed operating costs w/o capture (\$/kW-net)	58.280
Total variable operating costs w/o capture (\$/kWh-net)	0.00515
Fuel cost (\$/kWh-net)	0.01518
Cost of CO_2 avoided (USD/tCO_2)	68
Flue gas flowrate (kmol/hr)	79,300
Absorber inlet pressure (bara)	1.2
Absorber inlet temperature (ºC)	40
Flue gas composition (mol/mol)	
Ar	0.0082
CO_2	0.135
H_2O	0.1517
N_2	0.6808
O_2	0.0243

[a] The year of cost data in the referenced report is 2007, but the cost analysis in this study is based on 2021. (CEPCI: 2007 (525.4)[9], 2021 (699.97)[10])

Table S2. Summary of constants for power law expressions [11].

Related species	Reaction direction	$Reaction\ kinetics^a$
HCO_3^-	Forward	$r_{10} = 1.33 \times 10^{17}\, exp\left(-\frac{55.38}{R}\left[\frac{1}{T} - \frac{1}{298.15}\right]\right) a_{CO_2} a_{OH^-}$
HCO_3^-	Reverse	$r_{11} = 6.63 \times 10^{16}\, exp\left(-\frac{107.24}{R}\left[\frac{1}{T} - \frac{1}{298.15}\right]\right) a_{HCO_3^-}$
$MEACOO^-$	Forward	$r_{12} = 3.02 \times 10^{14}\, exp\left(-\frac{41.20}{R}\left[\frac{1}{T} - \frac{1}{298.15}\right]\right) a_{MEA} a_{CO_2}$
$MEACOO^-$	Reverse (absorber)	$r_{13} = 5.52 \times 10^{23}\, exp\left(-\frac{69.05}{R}\left[\frac{1}{T} - \frac{1}{298.15}\right]\right) \frac{a_{MEACOO^-} - a_{H_3O^+}}{a}$
$MEACOO^-$	Reverse (stripper)	$r_{13} = 6.56 \times 10^{27}\, exp\left(-\frac{95.24}{R}\left[\frac{1}{T} - \frac{1}{298.15}\right]\right) \frac{a_{MEACOO^-} - a_{H_3O^+}}{a}$

[a] Reaction rate and pre-exponential factor $(kmol/m^3 \cdot s)$, Activation energy (kJ/mol)

Table S3. Summary of main specifications for CO$_2$RR process.

CO$_2$RR			
CO$_2$ capture rate (%)	90		
MEA concentration (wt%)	30	**STRIPPER**	
Lean CO$_2$ loading (mol/mol)	0.13	Number of Stripper	2
Rich CO$_2$ loading (mol/mol)	0.46	Packing	SULZER Mellapak 250Y
		Calculation type	Rate-based
ABSORBER		Diameter (m)	8.38
Number of Absorber	2	Height (m)	29.41
Packing	SULZER Mellapak 250Y	Section pressure drop (bar)	0.019
Calculation type	Rate-based	Reboiler temp. (°C)	118.5
Diameter (m)	13.26	Reboiler Duty (MJ/hr)	828000
Height (m)	28.96	Condenser Duty (MW)	5.84
Section pressure drop (bar)	0.054		
Lean amine inlet temp (°C)	40	**LEAN AMINE COOLER**	
Rich amine outlet temp (°C)	59	Duty (MW)	130.36
CROSS HEATX			
Duty (MW)	114.42	**ELECTROLYZER**	
Lean amine inlet/outlet temp (°C)	118 / 82	Operating temp. (°C)	25
Rich amine inlet/outlet temp (°C)	59 / 97	Operating press. (bara)	1.01325

Table S4. Summary of main specifications for cCO$_2$RR process.

cCO$_2$RR			
CO$_2$ capture rate (%)	90	**CROSS HEATX**	
MEA concentration (wt%)	30	Duty (MW)	46.65
Lean CO$_2$ loading (mol/mol)	0.13	Lean amine inlet/outlet temp (°C)	25 / 39
Rich CO$_2$ loading (mol/mol)	0.46	Rich amine inlet/outlet temp (°C)	59 / 44
ABSORBER			
Number of Absorber	2	**ELECTROLYZER**	
Packing	SULZER Mellapak 250Y	Operating temp. (°C)	25
Calculation type	Rate-based	Operating press. (bara)	1.01325
Diameter (m)	13.4		
Height (m)	28.96		
Section pressure drop (bar)	0.051		
Lean amine inlet temp (°C)	38	**RICH AMINE COOLER**	
Rich amine outlet temp (°C)	59	Duty (MW)	56.67

Table S5. Pure component properties - Scalar parameter. [4]

Parameters	Units	$MEAH^+$	$MEACOO^-$	CO_2	H_3O^+	HCO_3^-
DGAQFM [a]	J/kmol	-1.8962e+08	-4.9299e+08	-3.8598e+08	-2.37129e+08	-5.8677e+08
DHAQFM [b]	J/kmol	-3.3164e+08	-7.0747e+08	-4.138e+08	-2.8583e+08	-6.9199e+08
DGFORM [c]	J/kmol			-3.9437e+08		
DHFORM [d]	J/kmol			-3.9351e+08		

Parameters	Units	OH^-	CO_3^{2-}	MEA	H_2O
DGAQFM	J/kmol	-1.57244e+08	-5.2781e+08		
DHAQFM	J/kmol	-2.29994e+08	-6.7714e+08		
DGFORM	J/kmol			-1.033e+08	-2.28572e+08
DHFORM	J/kmol	-1.4351e+08		-2.067e+08	-2.41818e+08
PCSFTM [e]				2.9029	1.0656
PCSFTU [f]	K			306.2	366.51
PCSFTV [g]				3.1067	3.0007
PCSFAU [h]	K			2369	2500.7
PCSFAV [i]				0.01903	0.034868
DHVLB [j]	J/kmol			5.02254e+07	

[a] : Aqueous phase free energy of formation at infinite dilution and 25 deg C. For ionic species and molecular solutes in electrolyte systems (DGAQFM)

[b] : Aqueous phase heat of formation at infinite dilution and 25 deg C. For ionic species and molecular splutes in electrolyte systems (DHAQFM)

[c] : Standard free energy of formation for ideal gas at 25 deg C (DGFORM)

[d] : Standard enthalpy of formation for ideal gas at 25 deg C (DHFORM)

[e] : PC-SAFT model parameter - segment number (m)

[f] : PC-SAFT model parameter - segment energy parameter (ε/k)

[g] : PC-SAFT model parameter - segment diameter (σ)

[h] : PC-SAFT model parameter - association energy (ε^{AB}/k)

[I] : PC-SAFT model parameter - effective association volume (κ^{AB})

[j] : Enthalpy of vaporization at TB (DHVLB)

Table S6. Parameters for aqueous infinite dilution heat capacity (CPAQ0) [4].

Component	Prop.unit	CPAQ0-	CPAQ0-	CPAQ0-	CPAQ0-	CPAQ0-	CPAQ0-	CPAQ0-	CPAQ0-
CO_2	J/kmol·K	132231	0	0	0	0	0	0	1000
H_3O^+	J/kmol·K	75291	0	0	0	0	0	0	2000
OH^-	J/kmol·K	-148500	0	0	0	0	0	0	2000
$MEAH^+$	J/kmol·K	210223	0	0	0	0	0	0	1000
$MEACOO^-$	J/kmol·K	-0.04978	0	0	0	0	0	0	1000
HCO_3^-	J/kmol·K	-29260	0	0	0	0	0	0	2000
CO^{2-}_3	J/kmol·K	-397100	0	0	0	0	0	0	2000

Table S7. Parameters for dielectric constant (CPDIEC) [5].

Component	Temp.unit	A_i	B_i	C_i
MEA	K	37.72	0	298.15

Table S8. Parameters for ideal gas heat capacity (CPIG) [4].

Component	Temp.uni	Prop.unit	CPIG-1	CPIG-2	CPIG-3	CPIG-4	CPIG-5	CPIG-6	CPIG-7	CPIG-8	CPIG-9	CPIG-10	CPIG-11
MEA	K	J/kmol·K	13207.4	281.577	-0.1513066	3.128692e-05	0	0	283	1000	33256	119.82113	1.0636971
H_2O	K	J/kmol·K	33738.112	-7.0175634	0.027296105	-1.6646536e-05	4.2976136e-09	-4.169608e-13	200	3000	33256	1.8978e-20	9.2846
CO_2	K	J/kmol·K	19795.19	73.436472	-0.056019384	1.715332e-05	0	0	300	1088.6	29099	0.71876	1.6368

Table S9. Parameters for Jones-Dole correction to liquid mixture viscosity (IONMOB) [4].

Component	Temp.unit	Prop.unit	l_1
$MEAH^+$	K	kmol	5
$MEACOO^-$	K	kmol	5

Table S10. Parameters for Jones-Dole correction to liquid mixture viscosity (IONMUB) [4].

Component	Temp.unit	Prop.unit	b_1
$MEAH^+$	K	cum/kmol	0.332352
$MEACOO^-$	K	cum/kmol	0.168106
HCO_3^-	K	cum/kmol	0.0946944
CO^{2-}_3	K	cum/kmol	0.564118

Table S11. Parameters for extended antoine vapor pressure for a liquid (PLXANT) [6]**.**

Component	Temp.unit	Prop.unit	PLXANT-1	PLXANT-2	PLXANT-3
MEA	℃	bar	161.267	-13492	0

PLXANT-4	PLXANT-5	PLXANT-6	PLXANT-7	PLXANT-8	PLXANT-9
0	-21.914	1.3779e-05	2	9.85	364.85

Table S12. Parameters for brelvi-O-connell volume (VLBROC) [4].

Component	Temp.unit	Prop.unit	VLBROC-1	VLBROC-2
CO_2	K	cum/kmol	0.174771	-0.000338
H_2O	K	cum/kmol	0.0464	0

Table S13. Parameters for henry's constant (HENRY) [4, 7]

Component	Component	Tem	Prop	a_{iA}	b_{iA}	c_{iA}	d_{iA}	T_L	T_H
CO_2	H_2O	K	N/sqm	100.65	-6147.7	-10.191	-0.01	273.15	473.15
N_2	H_2O	K	N/sqm	176.507	-8432.8	-21.558	-0.0084362	273	346
O_2	H_2O	K	N/sqm	155.921	-7775.1	18.3974	-0.0094435	274	348
H_2	H_2O	K	N/sqm	191.579	-6993.5	-26.312	0.0150431	274	339
CO_2	MEA	K	N/sqm	20.3143	-896.5	0	0	0	2000
AR	H_2O	K	N/sqm	180.991	-8137.1	-23.255	0.00306357	274	347
CO	H_2O	K	N/sqm	183.288	-8296.8	-23.337	0	273	353

Table S14. Parameters for NRTL [5, 7]

Component	Component	Temp	a_{ij}	a_{ji}	b_{ij}	b_{ji}	c_{ij}	T_L	T_H
H_2O	MEA	K	0.1559	1.5201	110.8	-910.3	0.3	273.15	1273.15
CO_2	H_2O	K	0	0	0	0	0.2	0	1000

Table S15. Parameters for liquid mixture viscosity (MUKIJ) [4].

Component	Component	Temp	a_{ij}	b_{ij}	c_{ij}	d_{ij}	e_{ij}	T_{ref}
MEA	H_2O	K	0.704111	0	0	0	0	298.15

Table S16. Parameters for liquid mixture viscosity (MULIJ) [4].

Component i	Component j	Temp Units	a_{ij}	a_{ji}	b_{ij}	b_{ji}	c_{ij}	c_{ji}	d_{ij}	d_{ji}	e_{ij}	e_{ji}	T_{ref}
MEA	H_2O	K	-1.7418	1.7418	0	0	0	0	0	0	0	0	298.15

Table S17. Parameters for clarke aqueous electrolyte volume (VLCLK) [4].

Component i	Component j	Prop Units	V_{ca}^{∞}	A_{ca}
$MEAH^+$	OH^-	cum/kmol	0	0
$MEAH^+$	HCO_3^-	cum/kmol	0.0712212	0
$MEAH^+$	$MEACOO^-$	cum/kmol	0.122196	0
$MEAH^+$	CO_3^{2-}	cum/kmol	0.0997818	0

Table S18. Global economic assumptions [8].

Global Economic Assumptions	
Discounted Rate	10%
Income Tax Rate	34% Federal, 6% State (Effective tax rate: 38%)
Capital Depreciation Type	MACRS (7 years)
Investment Tax Credit	0%
Tas Holiday	0 years
Capital Expenditure Period	Coal Plant: 5 years
Operational Period	30 years
Economic Analysis Period	35 years
Distribution of Total Overnight Capital	5-year period: 10%, 30%, 25%, 20%, 15%
Cash Reserves	8.33% of Annual cost of manufacture
Inventory	1.92% of Annual sales of liquid /solid product
Accounts Receivable	8.33% of Annual sales of all products
Accounts Payable	8.33% of Annual feedstock cost
Start Up Capacity	1st year: 50%, 2nd year: 75%
Year of cost data	2021

Table S19. Electrolyzer parameters adopted from the DOE H_2A project for capital cost calculation [3].

Parameter	Value
Reference PEM electrolyzer for H_2 production	50,000 kg/day
Reference PEM electrolyzer total uninstalled capital : Future central (2016$ k W^{-1})	$233
Reference PEM electrolyzer stack current density (mA cm^{-2})	3000
Reference PEM electrolyzer cell voltage (V)	1.8
Maintenance cost (% of capital per year) [12]	2.5
Cell compartment replacement cost (% of capital cost, every 7 year) [13]	15
MEA replacement cost (% of stack cost) [14]	5
Installation factor (% of uninstalled capital cost) [12]	12
Reference PEM electrolyzer purchase cost ($ m^{-2})	12,582
Reference PEM electrolyzer installed cost ($ m^{-2})	14,092

Table S20. Electrolyzer capital cost calculation.

Parameter	**Value**
Product production rate (kg/day)	3238168
Required partial current (A)	258190273
Required total current (A)	260798256
Required electrolyzer area (m^2)	130399
Required power (MW)	597
Reference PEM electrolyzer purchase cost ($/$m^2$) [3]	12582
Calculated electrolyzer cost ($) [12]	1,640,681,832
Calculated electrolyzer stack cost ($, 65% of total cost) [12]	1,066,443,191
Calculated electrolyzer BoP cost ($, 35% of total cost) [12]	883,444,063
Calculated electricity cost ($/year)	222,347,987

Table S21. Materials & utilities unit prices.

Parameter		Value
Utility cost [9]	Steam, 450 psig (\$/kg)	0.0176
	Steam, 150 psig (\$/kg)	0.0153
	Steam, 50 psig (\$/kg)	0.0132
	Electricity (\$/kWh)	0.05
	Cooling water (\$/m^3)	0.027
	Process water (\$/m^3)	0.27
	Boiler-feed water (\$/m^3)	0.56
	Refrigeration - 150F (\$/GJ)	33.2
	Refrigeration - 90F (\$/GJ)	23.3
	Refrigeration - 30F (\$/GJ)	13.17
	Refrigeration, 10F (\$/GJ)	6.47
	Chilled water, 40F (\$/GJ)	5
Material cost	Water (\$/1000 gallons) [1]	1.08
	MU & WT Chem (\$/lbs) [1]	0.17
	Limestone (\$/ton) [1]	21.63
	Carbon (Mercury Removal) (\$/lb) [1]	1.05
	MEA solvent (\$/ton) [1]	2249.89
	NaOH (\$/tons) [1]	433.68
	H_2SO_4 (\$/tons) [1]	138.78
	Activated carbon (\$/lb) [1]	1.05
	Ammonia (19% NH_3) (\$/ton) [1]	129.8
	KCl (\$/kg) [15]	0.8
	KOH (\$/kg) [16]	0.244
	Fuel (\$/ton) [1]	38.18
Product market price	CO (\$/kg) [13]	0.8
	Hydrogen (\$/kg) [17]	1.39

Table S22. Life cycle assessment assumptions.

Software	Simapro (Version 9.3, Pre-consultants, the Netherlands)
System boundary	Gate-to-gate
Data library	Ecoinvent v3.8
Assessment method	ReCiPe 2016 (H) v1.1
Functional unit	1.0 kg of CO

Table S23. Life cycle inventory obtained from Ecoinvent v3.8 [18] (Software: Simapro v9.3).

Materials	Name of data set	Database
Water	Water, process and cooling, unspecified natural origin	Ecoinvent v3.8
Water	Water, cooling, salt, ocean	Ecoinvent v3.8
Nitrogen	Nitrogen, atmospheric	Ecoinvent v3.8
Oxygen	Oxygen	Ecoinvent v3.8
Argon	Argon	Ecoinvent v3.8
Monoethanolamine	Monoethanolamine | market for | APOS, S	Ecoinvent v3.8
Potassium hydroxide	Potassium hydroxide [GLO]| market for | APOS, S	Ecoinvent v3.8
Carbon dioxide	Carbon dioxide, in chemical industry [GLO]| market for carbon dioxide, in chemical industry | APOS, S	Ecoinvent v3.8
Steam	Heat, from steam, in chemical industry [RER]| market for heat, from steam, in chemical industry | APOS, S	Ecoinvent v3.8
Electricity	Electricity, high voltage [RoW]| electricity production, wind, >3MW turbine, onshore | APOS, S	Ecoinvent v3.8
Electricity	Electricity, high voltage [RoW]| electricity production, natural gas, combined cycle power plant | APOS, S	Ecoinvent v3.8
Electricity	Electricity, high voltage [RoW]| heat and power co-generation, biogas, gas engine | APOS, S	Ecoinvent v3.8
Electricity	Electricity, high voltage [RoW]| treatment of coal gas, in power plant | APOS, S	Ecoinvent v3.8
Electricity	Electricity, high voltage [RoW]| electricity production, hydro, run-of-river | APOS, S	Ecoinvent v3.8
Electricity	Electricity, high voltage [RoW]| electricity production, nuclear, pressure water reactor | APOS, S	Ecoinvent v3.8
Electricity	Electricity, high voltage [RoW]| heat and power co-generation, hard coal | APOS, S	Ecoinvent v3.8
Electricity	Electricity, high voltage [RoW]| electricity production, solar thermal parabolic trough, 50 MW | APOS, S	Ecoinvent v3.8

Table S24. Characterization result of impact categories for cCO_2RR process in current case.

Impact category	Unit	Total	Direct emissions	Indirect emission			
				MEA	Potassium chloride	Potassium hydroxide	Electricity (Coal gas)
Global warming	kg CO_2 eq	3.80E+00	1.75E-01	1.35E-03	9.74E-05	6.34E-03	3.62E+00
Ozone formation, Human health	kg NO_x eq	2.39E-03	0	2.33E-06	2.81E-07	1.71E-05	2.38E-03
Terrestrial acidification	kg SO_2 eq	1.09E-03	0	3.78E-06	3.70E-07	2.46E-05	1.06E-03
Freshwater eutrophication	kg P eq	4.14E-05	0	4.10E-07	3.46E-08	2.88E-06	3.81E-05
Marine ecotoxicity	kg 1,4-DCB	6.80E-03	4.38E-06	5.31E-05	4.19E-06	4.75E-04	6.26E-03
Human carcinogenic toxicity	kg 1,4-DCB	2.19E-02	0	5.69E-05	4.06E-06	4.31E-04	2.14E-02
Fossil resource scarcity	kg oil eq	1.55E+00	0	6.11E-04	2.26E-05	1.73E-03	1.55E+00
Water consumption	m^3	7.52E-03	8.55E-04	2.05E-05	8.25E-07	6.16E-05	6.58E-03

Table S25. Characterization result of impact categories for cCO_2RR process in optimistic case.

Impact category	Unit	Total	Direct emissions	Indirect emission			
				MEA	Potassium chloride	Potassium hydroxide	Electricity (Coal gas)
Global warming	kg CO_2 eq	2.86E+00	1.75E-01	1.35E-03	9.74E-05	6.34E-03	2.68E+00
Ozone formation, Human health	kg NO_x eq	1.78E-03	0	2.33E-06	2.81E-07	1.71E-05	1.76E-03
Terrestrial acidification	kg SO_2 eq	7.95E-04	0	3.78E-06	3.70E-07	2.46E-05	7.67E-04
Freshwater eutrophication	kg P eq	2.89E-05	0	4.10E-07	3.46E-08	2.88E-06	2.56E-05
Marine ecotoxicity	kg 1,4-DCB	4.75E-03	4.38E-06	5.31E-05	4.19E-06	4.75E-04	4.21E-03
Human carcinogenic toxicity	kg 1,4-DCB	1.60E-02	0	5.69E-05	4.06E-06	4.31E-04	1.55E-02
Fossil resource scarcity	kg oil eq	1.15E+00	0	6.11E-04	2.26E-05	1.73E-03	1.14E+00
Water consumption	m^3	5.81E-03	8.55E-04	2.05E-05	8.25E-07	6.16E-05	4.87E-03

Table S26. Characterization result of impact categories for CO$_2$RR process.

Impact category	Unit	Total	Direct emissions	Indirect emission		
				MEA	Potassium hydroxide	Electricity (Coal gas)
Global warming	kg CO$_2$ eq	2.99E+00	1.76E-01	1.19E-03	9.27E-05	2.81E+00
Ozone formation, Human health	kg NO$_x$ eq	1.85E-03	0	2.04E-06	2.49E-07	1.85E-03
Terrestrial acidification	kg SO$_2$ eq	8.25E-04	0	3.32E-06	3.59E-07	8.21E-04
Freshwater eutrophication	kg P eq	3.00E-05	0	3.59E-07	4.21E-08	2.96E-05
Marine ecotoxicity	kg 1,4-DCB	4.92E-03	3.83E-06	4.65E-05	6.94E-06	4.87E-03
Human carcinogenic toxicity	kg 1,4-DCB	1.67E-02	0	4.99E-05	6.29E-06	1.66E-02
Fossil resource scarcity	kg oil eq	1.20E+00	0	5.35E-04	2.53E-05	1.20E+00
Water consumption	m^3	6.07E-03	9.36E-04	1.80E-05	9.00E-07	5.12E-03

Table S27. Normalization result of impact categories for cCO$_2$RR process in current case.

Impact category	Total	Direct emissions	Indirect emission			
			MEA	Potassium chloride	Potassium hydroxide	Electricity (Coal gas)
Global warming	4.75E-04	2.18E-05	1.69E-07	1.22E-08	7.93E-07	4.53E-04
Ozone formation, Human health	1.16E-04	0	1.13E-07	1.37E-08	8.29E-07	1.15E-04
Terrestrial acidification	2.65E-05	0	9.23E-08	9.03E-09	5.99E-07	2.58E-05
Freshwater eutrophication	6.37E-05	0	6.31E-07	5.33E-08	4.44E-06	5.86E-05
Marine ecotoxicity	1.56E-04	1.01E-07	1.22E-06	9.64E-08	1.09E-05	1.44E-04
Human carcinogenic toxicity	2.12E-03	0	5.53E-06	3.95E-07	4.18E-05	2.08E-03
Fossil resource scarcity	1.58E-03	0	6.23E-07	2.30E-08	1.77E-06	1.58E-03
Water consumption	2.82E-05	3.20E-06	7.69E-08	3.09E-09	2.31E-07	2.47E-05

Table S28. Normalization result of impact categories for cCO_2RR process in optimistic case.

Impact category	Total	Direct emissions	Indirect emission			
			MEA	Potassium chloride	Potassium hydroxide	Electricity (Coal gas)
Global warming	3.58E-04	2.18E-05	1.69E-07	1.22E-08	7.93E-07	3.35E-04
Ozone formation, Human health	8.64E-05	0	1.13E-07	1.37E-08	8.29E-07	8.54E-05
Terrestrial acidification	1.98E-05	0	9.23E-08	9.03E-09	5.99E-07	1.91E-05
Freshwater eutrophication	4.85E-05	0	6.31E-07	5.33E-08	4.44E-06	4.34E-05
Marine ecotoxicity	1.19E-04	1.01E-07	1.22E-06	9.64E-08	1.09E-05	1.07E-04
Human carcinogenic toxicity	1.58E-03	0	5.53E-06	3.95E-07	4.18E-05	1.54E-03
Fossil resource scarcity	1.17E-03	0	6.23E-07	2.30E-08	1.77E-06	1.17E-03
Water consumption	2.18E-05	3.20E-06	7.69E-08	3.09E-09	2.31E-07	1.83E-05

Table S29. Normalization result of impact categories for CO_2RR process.

Impact category	Total	Direct emissions	Indirect emission		
			MEA	Potassium hydroxide	Electricity (Coal gas)
Global warming	3.74E-04	2.20E-05	1.48E-07	1.16E-08	3.52E-04
Ozone formation, Human health	8.98E-05	0	9.91E-08	1.21E-08	8.97E-05
Terrestrial acidification	2.01E-05	0	8.09E-08	8.76E-09	2.00E-05
Freshwater eutrophication	4.62E-05	0	5.53E-07	6.48E-08	4.56E-05
Marine ecotoxicity	1.13E-04	8.81E-08	1.07E-06	1.60E-07	1.12E-04
Human carcinogenic toxicity	1.62E-03	0	4.84E-06	6.11E-07	1.61E-03
Fossil resource scarcity	1.23E-03	0	5.46E-07	2.58E-08	1.23E-03
Water consumption	2.28E-05	3.51E-06	6.74E-08	3.37E-09	1.92E-05

Table S30. Environmental impacts of various electricity sources.

Electricity source	Global warming impact		
	cCO_2RR (Current)	cCO_2RR (Optimistic)	CO_2RR
Coal gas	3.803	2.861	2.991
Natural gas	5.086	3.833	4.012
Hard coal	10.341	7.721	8.097
Wind	0.456	0.407	0.413
Nuclear	0.206	0.222	0.219
Hydro	0.189	0.210	0.206
Solar	0.574	0.495	0.505
Biogas	1.843	1.433	1.491

Supplementary figures

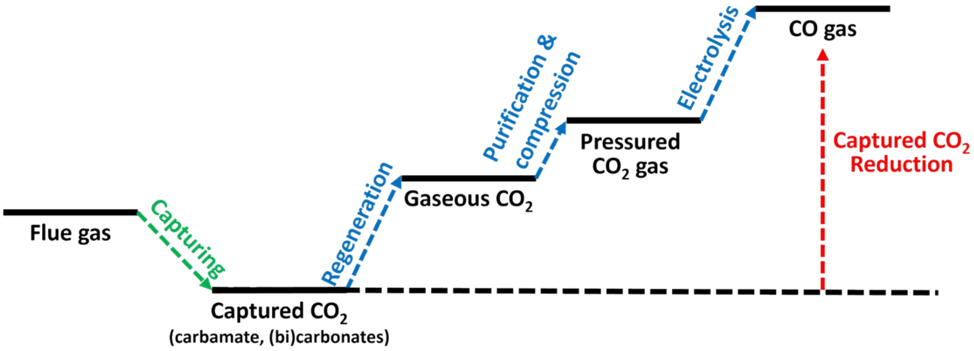


Fig. S1. Energy diagram between gaseous CO$_2$ and carbamate reduction reactions.

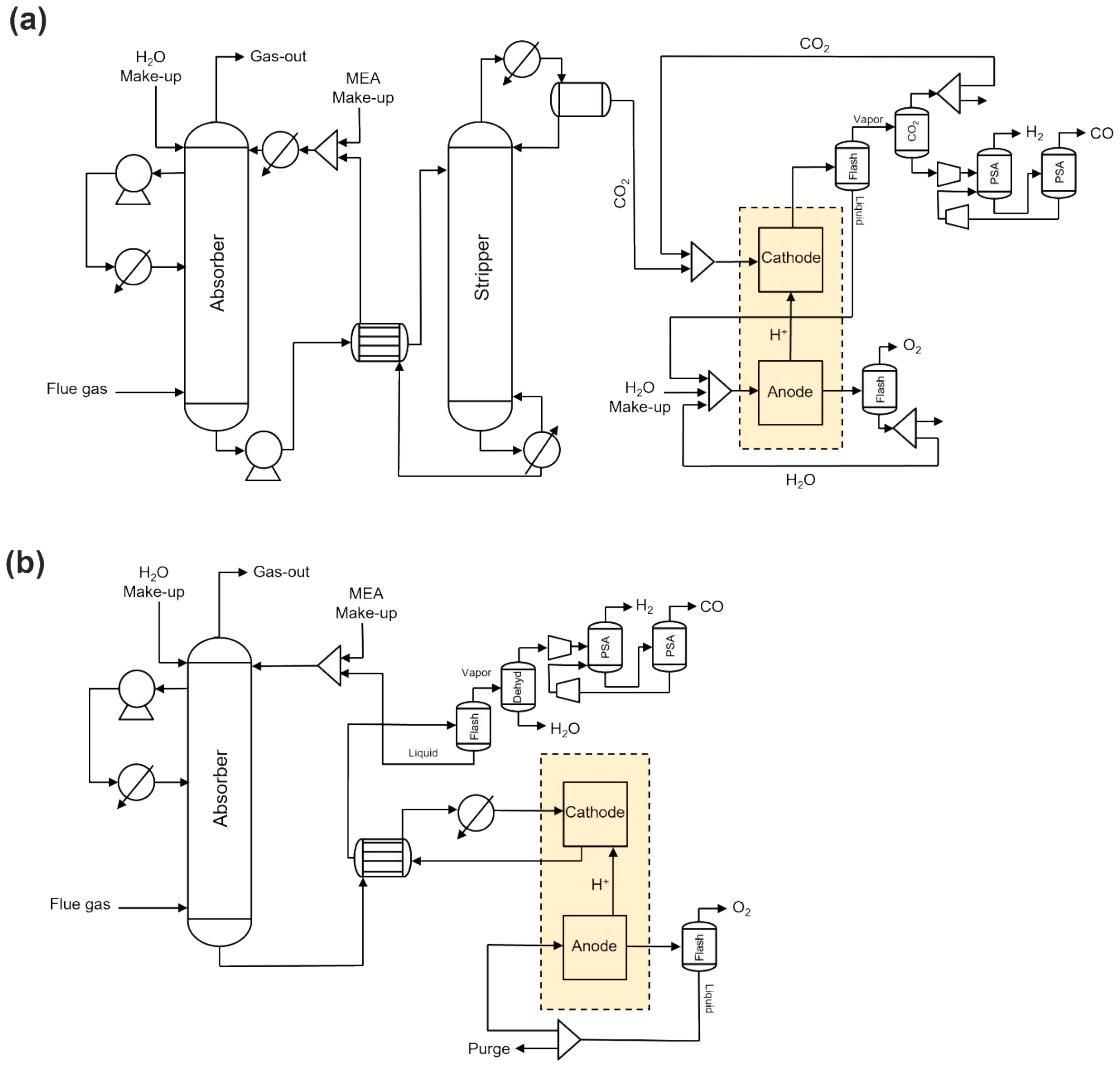


Fig. S2. PFD of two CCU technology (a) CO_2RR process and (b) cCO_2RR process.

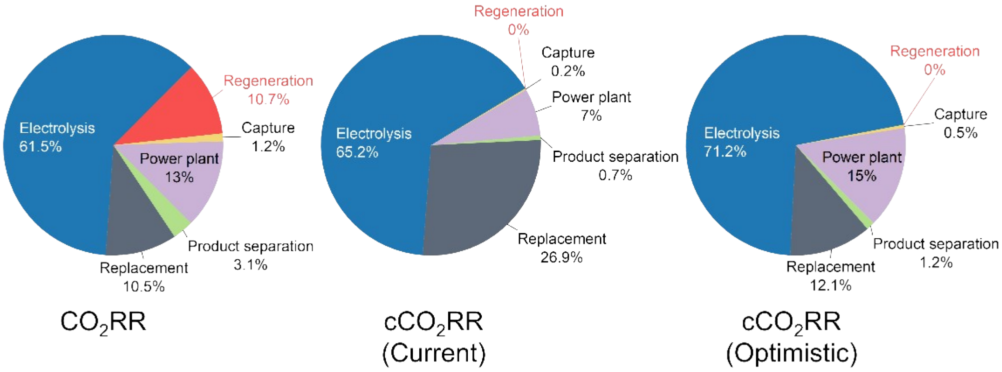


Fig. S3. Pie chart showing the percentage of annual operating cost by unit process under different electrochemical performances scenarios.

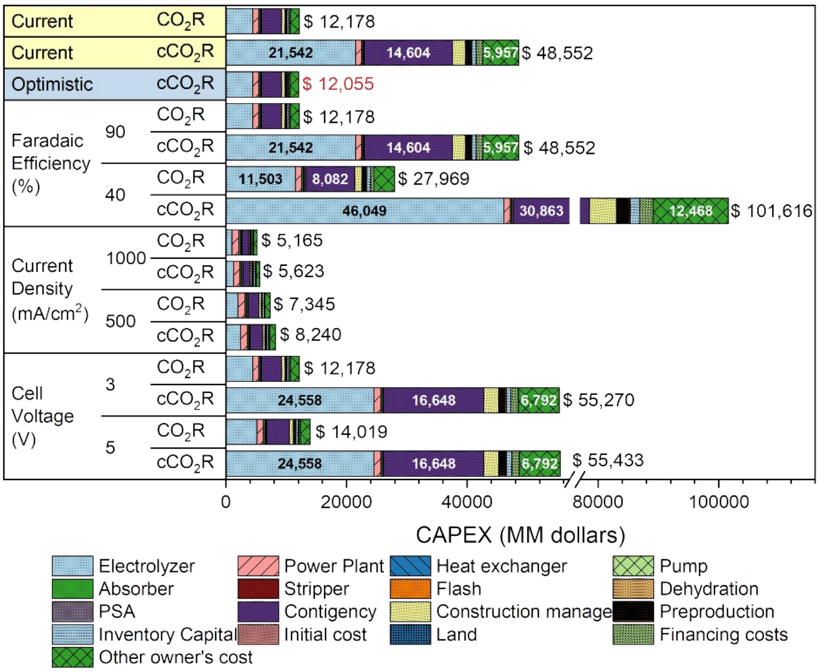


Fig. S4. Comparison of the total capital cost (CAPEX) breakdown of the cCO_2RR and CO_2RR processes under different electrochemical performances scenarios (current level, optimistic level, FE, current density, and overpotential).

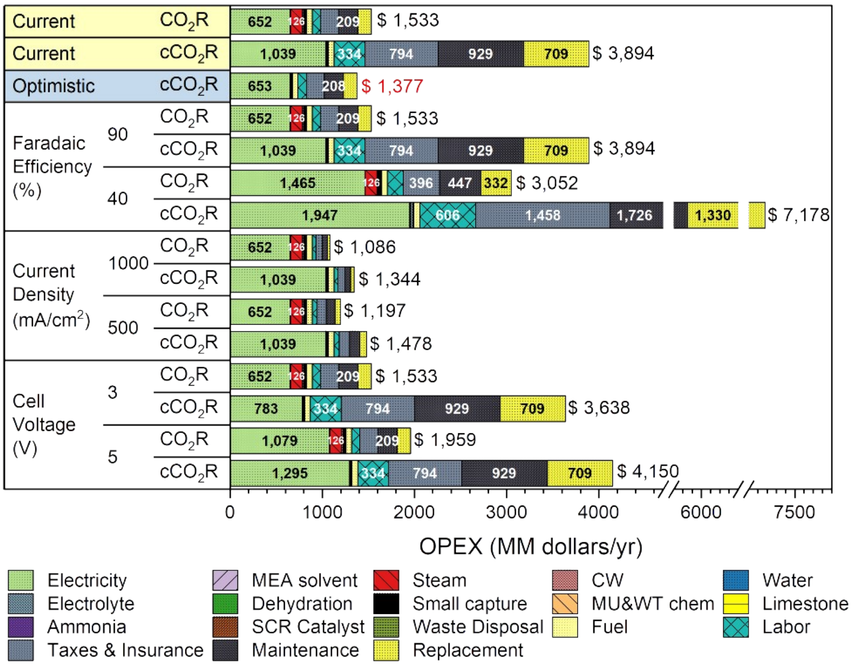


Fig. S5. Comparison of the total operating cost (OPEX) breakdown of the cCO_2RR and CO_2RR processes under different electrochemical performances scenarios (current level, optimistic level, FE, current density, and overpotential).

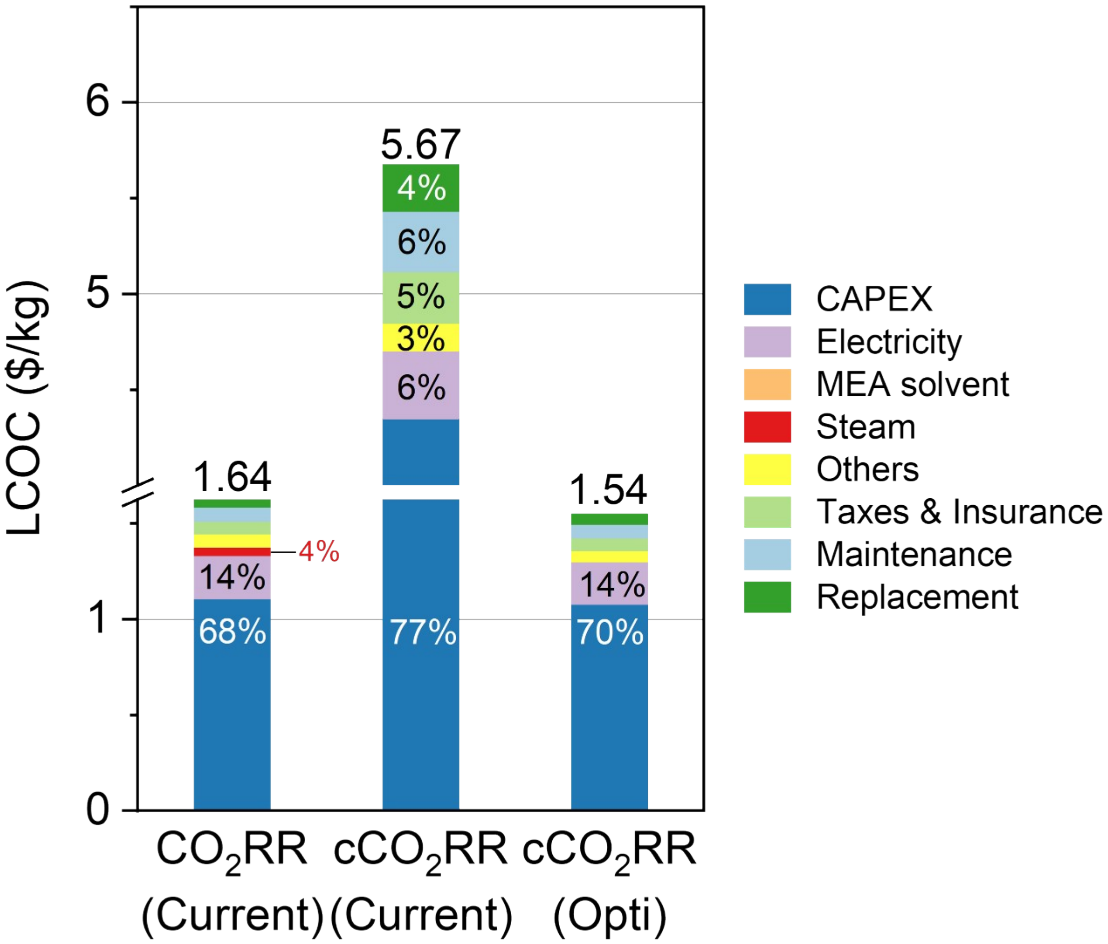


Fig. S6. Cost breakdown based on LCOC of both CCU processes under different electrochemical performances scenarios.

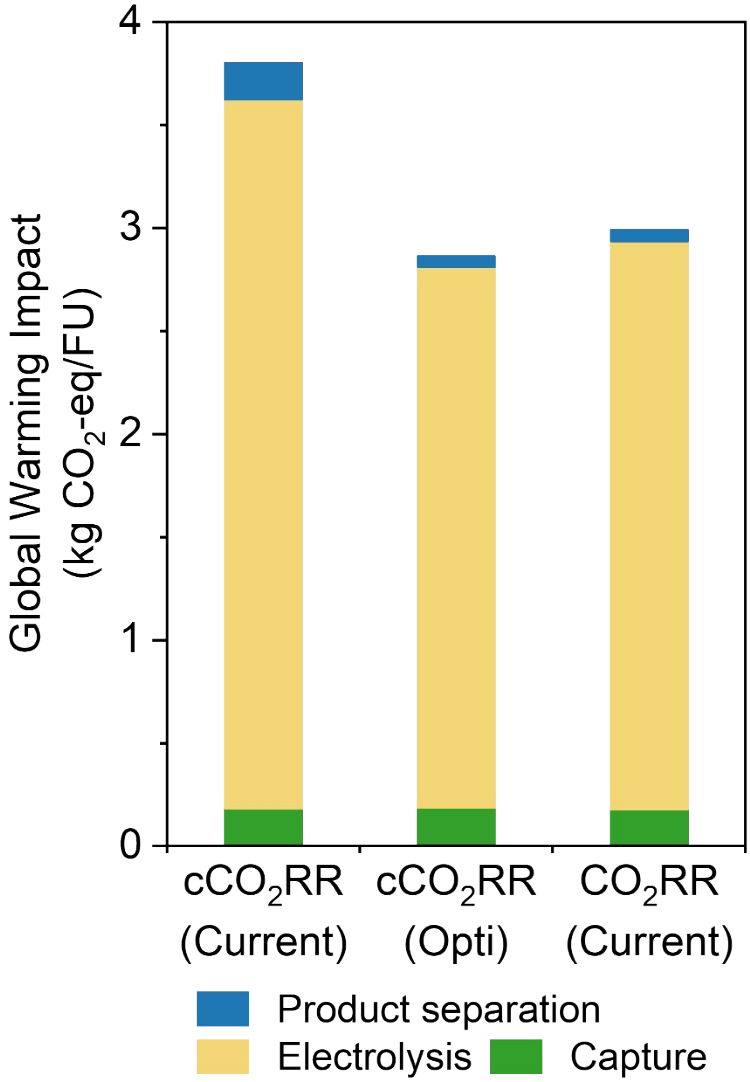


Fig. S7. Comparison of the global warming impact (GWI) according to unit processes.

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