Supplementary Materials

Tuning sorbent properties to reduce the cost of direct air capture

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S1. INPUT VARIABLES FOR ECONOMIC MODEL

The process parameters correspond to a polyamine-silica sorbent in a polymeric contactor (see Figure 1). The sorbent is used in a temperature swing adsorption process with the following steps: (1) adsorption, (2) vacuum to remove interstitial air from module, (3) heat to desorption temperature, (4) desorption, and (5) cool to adsorption temperature. See section "2.2 System Design" in the manuscript for more details on the sorbent and process setup.

Table S1 includes the parameters used in the analysis for each of the figures in the manuscript.

- The parameters used in **Figures 2-4** are representative for a polyamine-silica sorbent in a polymeric contactor.
- In **Figure 6**, experimentally-measured values are input for the heat of adsorption and working capacity. All other parameters are the same as Figures 2-4.
- **Figure 7** (except for the "best case scenario") also utilizes the same parameters as Figures 2-4, but the working capacity and cycle time are varied between minimum and maximum values.
- For the **"best case scenario" in Figure 7,** many of the parameters are changed to highly optimistic values. The minimum cycle time and maximum working capacity are used.

Table S1. Summary of input variables used for the calculation of LCOC in Figures 2-4, 6-7. Figures 2-4 use representative parameters for a polyamine-silica sorbent in a polymeric structured contactor. The working capacity and heat of adsorption are changed to experimentally-measured parameters in Figure 6. Figure 7 varies the working capacity and cycle time between minimum and maximum values. The "best case scenario" in Figure 7 uses highly optimistic values for many of the parameters.

Parameter	Fig. 2-4	Fig. 6	Fig. 7	Fig. 7 Optimistic	Unit
Plant					
Desired annual cumulative CO_2 captured	1,000,000 / 1.00	1,000,000 / 1.00	1,000,000 / 1.00	1,000,000 / 1.00	t-CO_2/yr / Mt-CO_2/yr
CO_2 feed concentration	0.0006069	0.0006069	0.0006069	0.0006069	wt. fraction
Process					
Cycle time	1800	1800	600 – 21,600	600	s
Adsorption step time (includes cooling step)	1200	1200	400 – 14,400	400	s
Desorption step time (includes heating step)	600	600	200 – 7,200	200	s
CO_2 product purity	0.95	0.95	0.95	0.98	
Temperature swing ($T_{des} - T_{ads}$)	70	70	70	60	C
Vacuum pressure	20	20	20	20	kPa
Initial CO_2 recovery	0.50	0.50	0.50	0.70	
Air superficial velocity	4*	4*	4*	5.4*	m/s
Adsorbent					
CO_2 working capacity of amine + support	1	0.455 – 0.84	0.25 – 3.0	3.0	mol/kg
CO_2 heat of adsorption	-70	-57 to -80	-70	-60	kJ/mol
CO_2/N_2 selectivity	45,631	45,631	45,631	200,000	
N_2 heat of adsorption	-15	-15	-15	-15	kJ/mol
Comp. 1 heat capacity	1.20	1.20	1.20	1.20	kJ/(kg·K)
Comp. 2 heat capacity	0.68	0.68	0.68	0.68	kJ/(kg·K)
Composition & Dimensions of Contactor & Module					
Amine + support loading in contactor	0.50	0.50	0.50	0.80	mass frac
Support loading in polymer +support	0.37	0.37	0.37	0.70	mass frac
Mass ratio of amine to support	0.70	0.70	0.70	0.70	
Amine loading in contactor	0.21	0.21	0.21	0.33	mass frac
Support loading in contactor	0.29	0.29	0.29	0.47	
CO_2 working capacity of contactor	0.50	0.50	0.50	0.80	mol/kg contactor
Pressure drop	500	500	500	500	Pa
Contactor density (including porosity)	730	730	730	730	kg/m^3
Polymer heat capacity	1.50	1.50	1.50	1.50	kJ/(kg·K)
Module					
Module void fraction	0.5	0.5	0.5	0.4**	
Module diameter	1.0	1.0	1.0	0.86**	m
Module length	0.37*	0.37*	0.37*	0.05**	m

*These variables are solved for and not input by the user (see section S3.C).

**See note below table (on pg. 7) for explanation.

Table S1 Contd. Summary of input variables used for the calculation of LCOC in Figures 2-4, 6-7.

Parameter	Figure 2-4	Fig. 6	Fig. 7	Fig. 7 optimistic	Unit
Sorbent & Contactor Costs					
Amine (PEI) raw material cost	3	3	3	2	$/kg
Support (silica) raw material cost	2	2	2	1	$/kg
Amine replacement cost	5	5	5	5	$/module
Contactor cost including polymer, solvents, stainless steel housing	25	25	25	10	$/kg
Contactor system installation factor	3	3	3	**3**	
Energy Costs					
Electricity cost	0.06	0.06	0.06	0.06	$/MWh
Isentropic efficiency of turbine	0.75	0.75	0.75	0.75	
Heat recovery efficiency	0.85	0.85	0.85	0.85	
Cost of steam	0.02929	0.02929	0.02929	0.02929	$/kg
Carbon footprint	171	171	171	17	kg CO_2/MWh$_e$
Blowers and Vacuum Pumps					
Number of modules per blower	20*	20*	20*	20*	
Number of modules per vacuum pump	10	10	10	10	
Isentropic pump efficiency	0.70	0.70	0.70	0.70	
Initial pressure of vacuum pump	101.3	101.3	101.3	101.3	kPa
Final pressure of vacuum pump	20	20	20	50	kPa
Heat capacity ratio (vacuum pump)	1.1	1.1	1.1	1.1	
Economic					
Plant capital lifetime (for annualization)	10	10	10	20	years
Contactor lifetime (for annualization)	5	5	5	10	years
Interest rate	10%	10%	10%	10%	
Plant capital recovery factor	0.16	0.16	0.16	0.117	
Contactor capital recovery factor	0.264	0.264	0.264	0.163	
Plant utilization	0.90	0.90	0.90	0.90	
CEPCI of project (2022)	816	816	816	816	
Indirect capital cost	0.35	0.35	0.35	0.35	Fraction of direct capital
Contingency	0.30	0.30	0.30	0.20	
Piping	0.40	0.40	0.40	0.10	
Number of operators per shift	8	8	8	8	
Number of shifts	8	8	8	8	
Base labor rate[2]	79,370	79,370	79,370	79,370	$/yr
Other					
Transport - Midwest (IL Basin)[3]	2.07	2.07	2.07	2.07	$/t-$CO_2$
Storage - Midwest (IL basin)[3]	8.32	8.32	8.32	8.32	$/t-$CO_2$

Note on change to module size in optimistic scenario in Figure 7:

In the optimistic scenario in Figure 7, the module void fraction was reduced from 0.5 to 0.4 because a higher packing fraction is more desirable but challenging to achieve.

The model is set up with the module length, module diameter, number of contactors per blower, individual blower flow rate, and air velocity as inputs, and the recovery is an output. In the optimistic scenario, the recovery increases from 50% to 70%. Because the CO_2 product is kept constant at 1 Mt-CO_2/year, increasing the recovery reduces the amount of air fed to the system ($q_{air,\,feed}$). If increasing the recovery was the only change made (with no change to sorbent amount or module size), we would expect the superficial velocity (v) to decrease (see eq. S17).

$$q_{air,\,feed} = A_{total,\,modules}\left[m^2\right] \cdot v\left[\frac{m}{s}\right] \# (S17)$$

However, in the optimistic scenario, the working capacity also increases from 1.0 mmol/g to 3.0 mmol/g, which decreases the volume of sorbent (V_{SC}) required. When the sorbent volume decreases, we can either reduce the length, total cross-sectional area, or both. If we reduce the cross-sectional area ($A_{total,\,modules}$) of the sorbent by too much, then we will need to use an unreasonably high superficial velocity (v) to achieve the same volumetric air flow rate (see eq. S17). Therefore, we choose to reduce both the cross-sectional area and length. We reduce the module diameter (from 1.0 m to 0.86 m), which reduces the total cross-sectional area and increases the necessary superficial velocity to 5.4 m/s. To compensate for the remaining necessary reduction in sorbent volume, we also reduce the module length (l_{module}) from 0.37 m to 0.05 m.

$$A_{total,SC}\left[m^2\right] = \frac{V_{SC}\left[m^3\right]}{l_{module}\left[m\right]} \# (S13)$$

These "decisions" are made by the solver system described in section S3.C.

Table S2. Parameters used for the calculation of LCOC in Fig. 6. The equilibrium capacity, heat of adsorption, and % of initial capacity at 20 cycles are experimentally-measured values. The % of initial capacity at 2 years was set to achieve an equivalent LCOC for all four sorbents.

Parameter	Commercial PEI	0.05HP-PEI	0.10HP-PEI	0.15HP-PEI
Equilibrium capacity (mmol/g)	1.2	1.1	0.83	0.65
Working capacity (mmol/g) (70% of equilibrium)	0.84	0.77	0.581	0.455
Heat of ads. (kJ/mol)	-80	-77	-69	-57
First decay constant rate (cycle^{-1})	6.34×10^{-4}	0	0	0
% of initial capacity at 20 cycles	98.6	100	100	100
Second decay constant rate (cycle^{-1})	2.15×10^{-5}	1.90×10^{-5}	9.12×10^{-6}	4.05×10^{-6}
% of initial capacity at 2 years	50	55	75	88

S2. SUPPLEMENTAL FIGURES

Blower and vacuum pump scheduling is used in the model to reduce capital cost while maintaining the system within physically reasonable bounds. **Figure S1** illustrates the unintended downtime that can occur for modules without proper placement and scheduling of the blower and vacuum pumps. The centralized blower and vacuum pump design is illustrated in **Figure S2a**. **Figure S2b** shows the corresponding schedule, and **Figure 2c** illustrates the rotation of the vacuum pumps around the modules. The result is a system of 20 modules per 1 blower and 2 vacuum pumps, and this design is used in the analysis for **Figures 2-4, 6-7**.
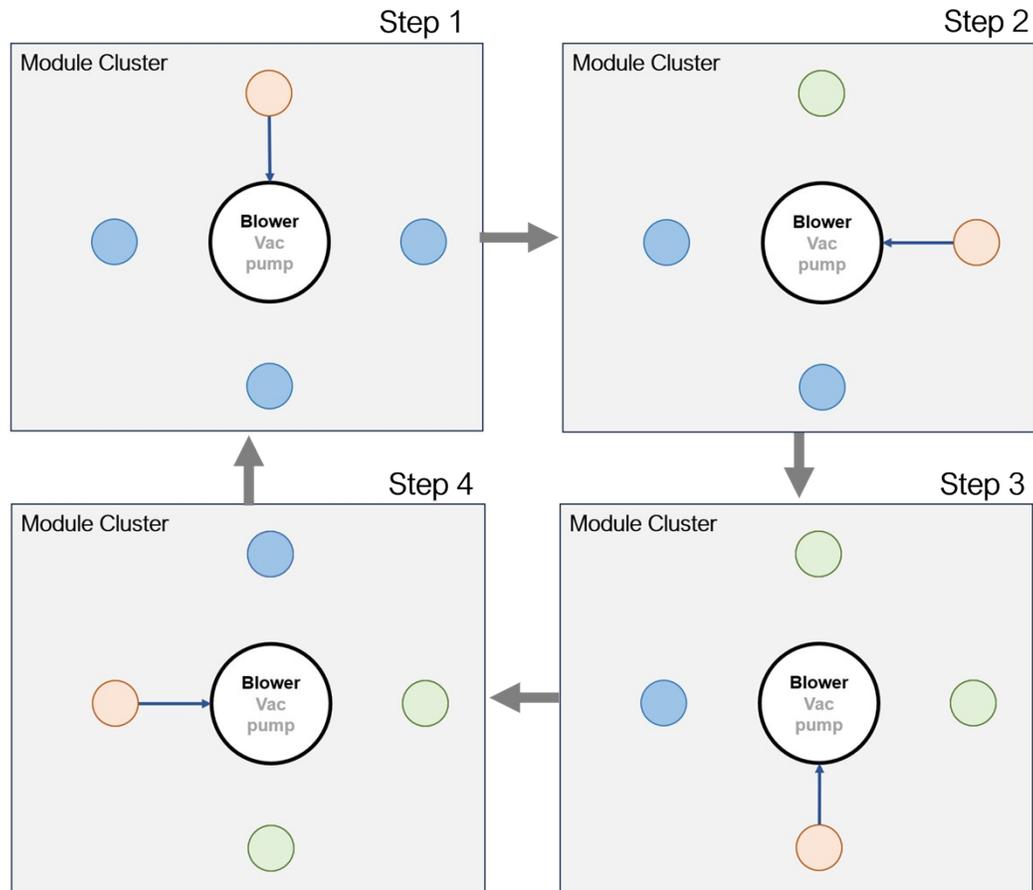
Figure S1. Schematic illustrating the consequences of inadequate blower and vacuum pump scheduling. Four modules (circles) are centered around one blower and vacuum pump. Red indicates desorption (using vacuum pump), green indicates adsorption (using blower), and blue indicates not in operation. The timing is based on a 2:1 ratio of adsorption to desorption time. When the first module finishes adsorption (step 4), the vacuum pump is still being used by the fourth module, which leads to downtime for the first module.
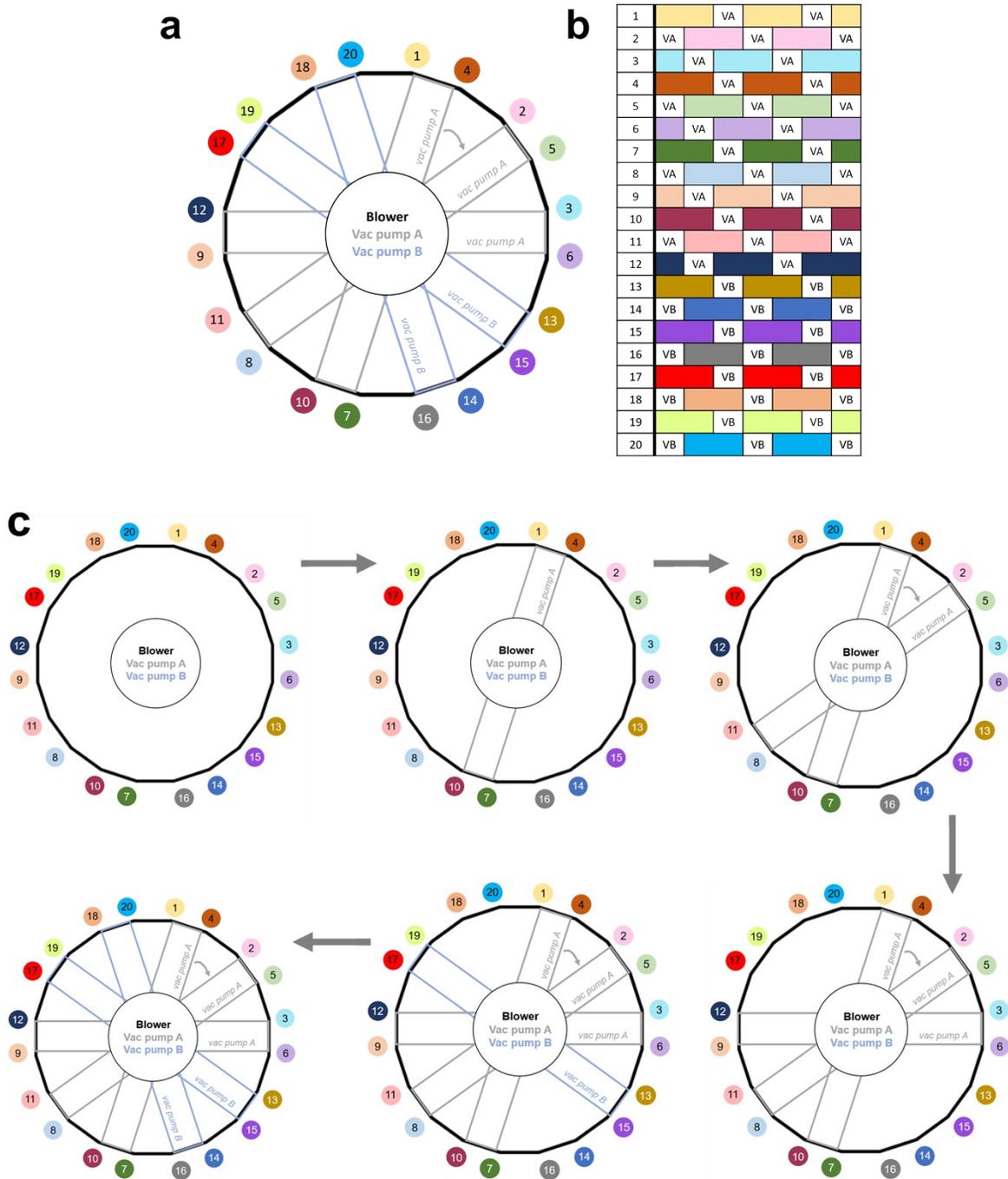
Figure S2. (a) Arrangement of modules (small, numbered circles) around a centralized blower and vacuum pumps and **(b)** the corresponding schedule based on a 2:1 ratio of adsorption to desorption time. This schedule allows for 18 – 22 modules to share one blower and two vacuum pumps. "VA" and "VB" denote vacuum pumps A and B, and the colored bars represent the adsorption step. **(c)** Illustration of vacuum pump service rotating around the circle of modules. Four modules in parallel can use a vacuum pump at the same time. Vacuum pump connections A and B rotate at the same time.

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An average of the energy carbon footprints of the lowest five states is used in the analysis (171 kg CO_2/MWh). The total DAC carbon footprint using the lowest five state average is shown in **Figure 2c**. Below, **Figure S3** illustrates the electricity grid distribution for each of those five states. The carbon emissions from each of these energy sources (i.e., natural gas, nuclear, wind) are listed in Table S24. Raw data from this figure along with a breakdown of the carbon footprints of each state are in Table S25.
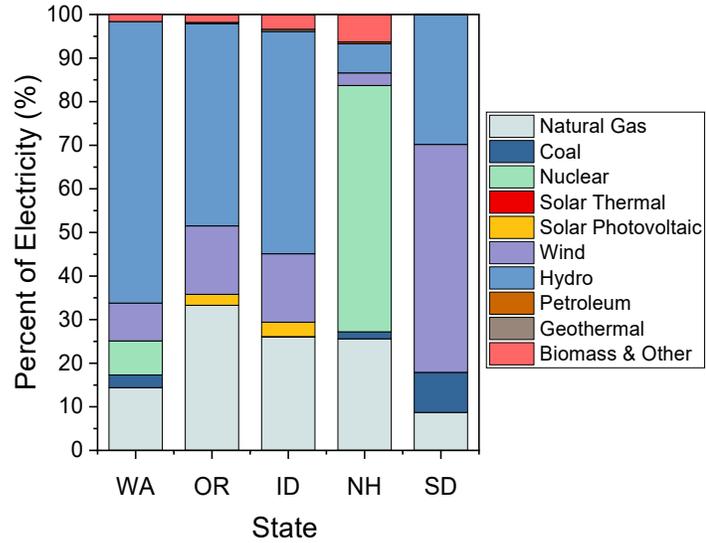
Figure S3. Electricity grid distribution from energy sources for the lowest five states referred to in Table S25.

Figures S4 – S6 contain supplemental experimental characterization (N_2 physisorption, amine efficiency, and CO_2 uptake profiles) for the modified PEI sorbents described in **Figure 5**.
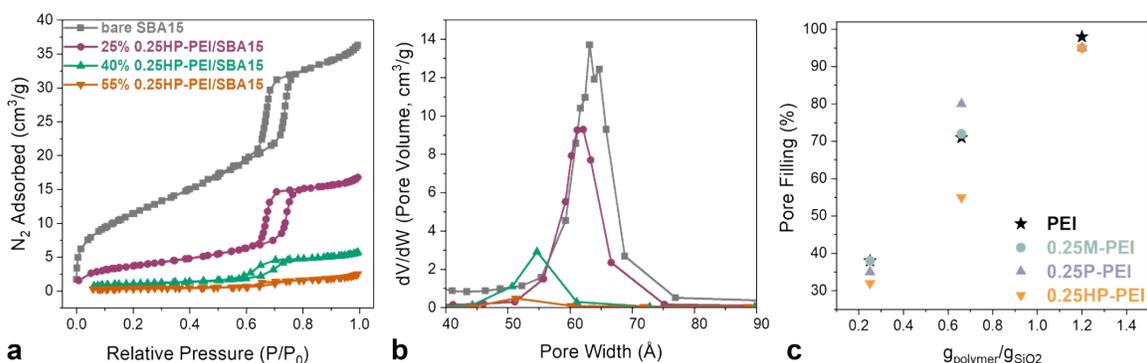
Fig S4. (a) N_2 physisorption isotherms at 77 K, (b) pore size distributions calculated by the BJH method for 0.25HP-PEI on SBA-15 composite sorbents as a function of amine loading, and (c) pore filling in SBA-15 as a function of modified polymer loading.
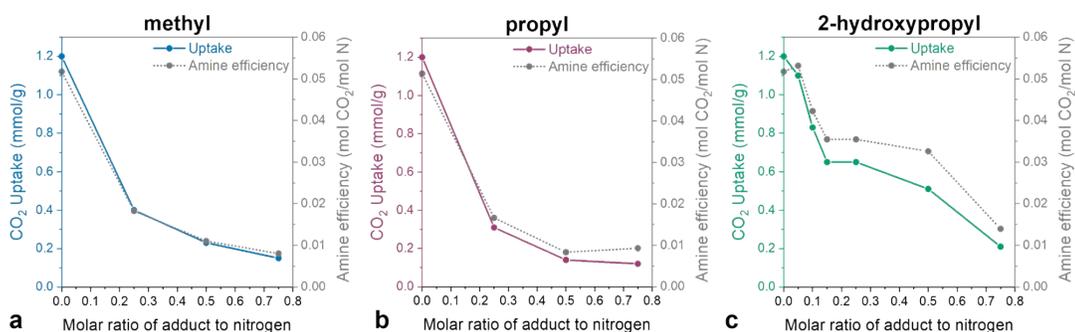
Figure S5. CO_2 uptake and amine efficiency (mol CO_2/mol N) as a function of % of adduct in PEI solution.
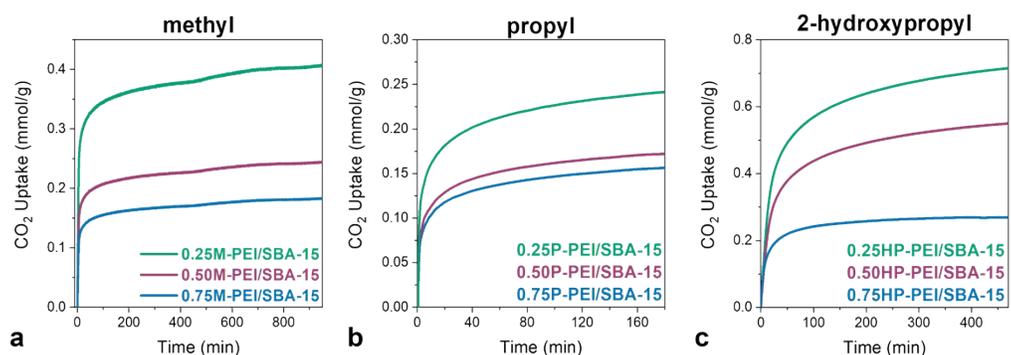
Figure S6. Gravimetric CO_2 uptake (400 ppm CO_2) of modified PEIs impregnated in SBA-15, **(a)** methyl-modified, **(b)** propyl-modified, and **(c)** 1,2 epoxypropane modified PEI. Green = 25% titrated, magenta = 50% titrated, and blue = 75% titrated.

In Figure 6c, the "accelerated" cycles of unmodified PEI from Choi et al., Nat Commun. (2016) are fit to a piecewise exponential function.[1] In Figure S7, we show the cycles fit to all three functions, linear (Fig. S7a), exponential (Fig. S7b), and piecewise exponential (Fig. S7c). We also show the cycles for a modified PEI sample (0.15EP-PEI) from Choi et al. fit to linear (Fig. S7d), exponential (Fig. S7e), and piecewise exponential (Fig. S7f).[1]
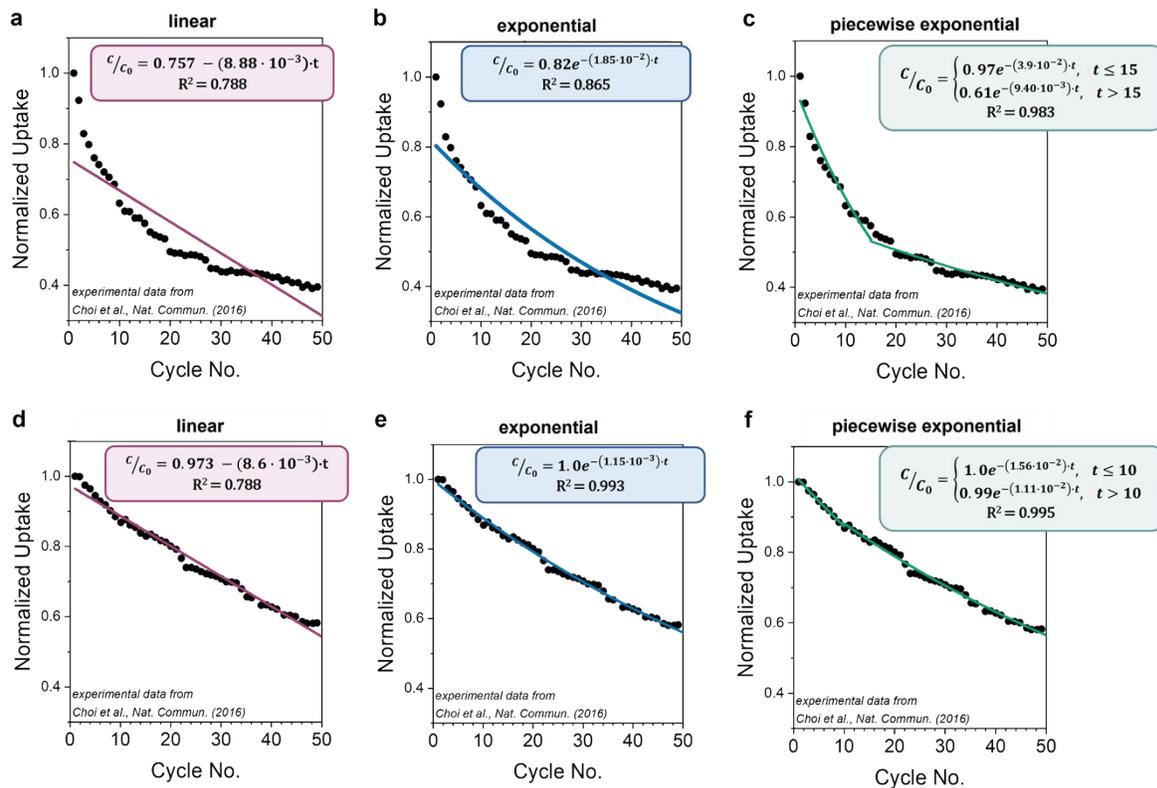
Figure S7. **Fitting accelerated cyclic capacity fade data. (a-c)** unmodified PEI deactivation profile fit to linear, exponential, and piecewise exponential functions. **(d-f)** 0.15EP-PEI (2-hydroxybutyl functionalized PEI) from Choi et al.[1] to linear, exponential, and piecewise exponential function.

The thermal energy will change each cycle as the sorbent degrades. In the model, the thermal energy is fit to a cubic function, which is integrated to determine the cumulative thermal energy. Examples of the fit are shown below in **Figure S8**. These examples correspond to the results in **Figure 4** with exponential, linear, and piecewise exponential capacity decay profiles.
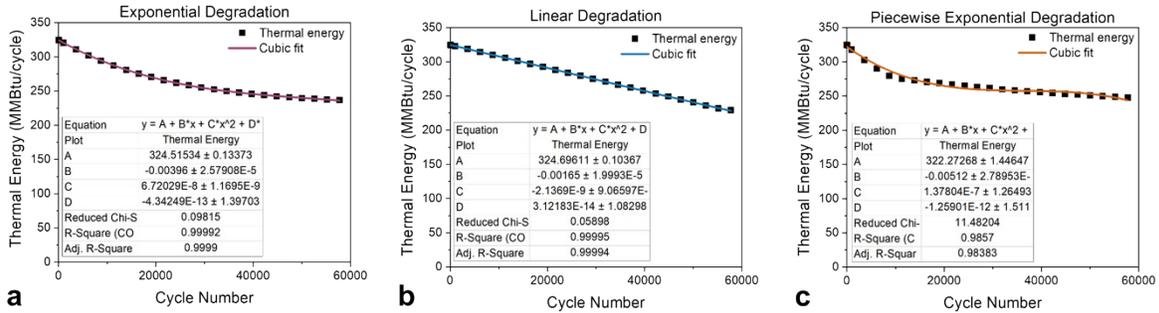
Figure S8. **Examples of fitting thermal energy to cubic function.** Examples correspond to results in Figure 3. (a) Exponential decay, $k = 4.40 \times 10^{-5}$ cycle^{-1}, (b) Linear decay, $k = 1.76 \times 10^{-5}$ cycle^{-1}, (c) Piecewise exponential decay, $k_1 = 7.33 \times 10^{-5}$ cycle^{-1}, $k_2 = 1.94 \times 10^{-5}$ cycle^{-1}, and $t_s = 0.6$ years.

Measurement of gases inside TGA during adsorption-desorption cycle

It was hypothesized in section 3.5 that the TGA used is not air-tight, resulting in trace oxygen in the furnace during adsorption-desorption cycles. To evaluate this, a mass spectrometer was connected to the outlet of the TGA during the second adsorption-desorption cycle of the unmodified PEI in **Fig. 6b**. The ion current is normalized to the value at the start of measurement, which is thought to be lab air in the line connecting the TGA outlet to the mass spec. After making the connection, N_2 flowing through the TGA fills the line, which is observed as an increase in N_2 concentration and decrease in CO_2, H_2O, Ar, and O_2 concentrations. When the gas is switched to 400 ppm CO_2 in N_2 at the start of adsorption, we observe the CO_2 concentration rise again. The CO_2 concentration also increases as the temperature is increased above 70 °C during desorption, corresponding to CO_2 desorbing from the sample. If we assume (1) a linear calibration curve between ion current and concentration and (2) that the values at the start of the measurement correspond to lab air, then we estimate the oxygen concentration in the TGA furnace as approximately 0.02%, or approximately 0.02 kPa partial pressure.
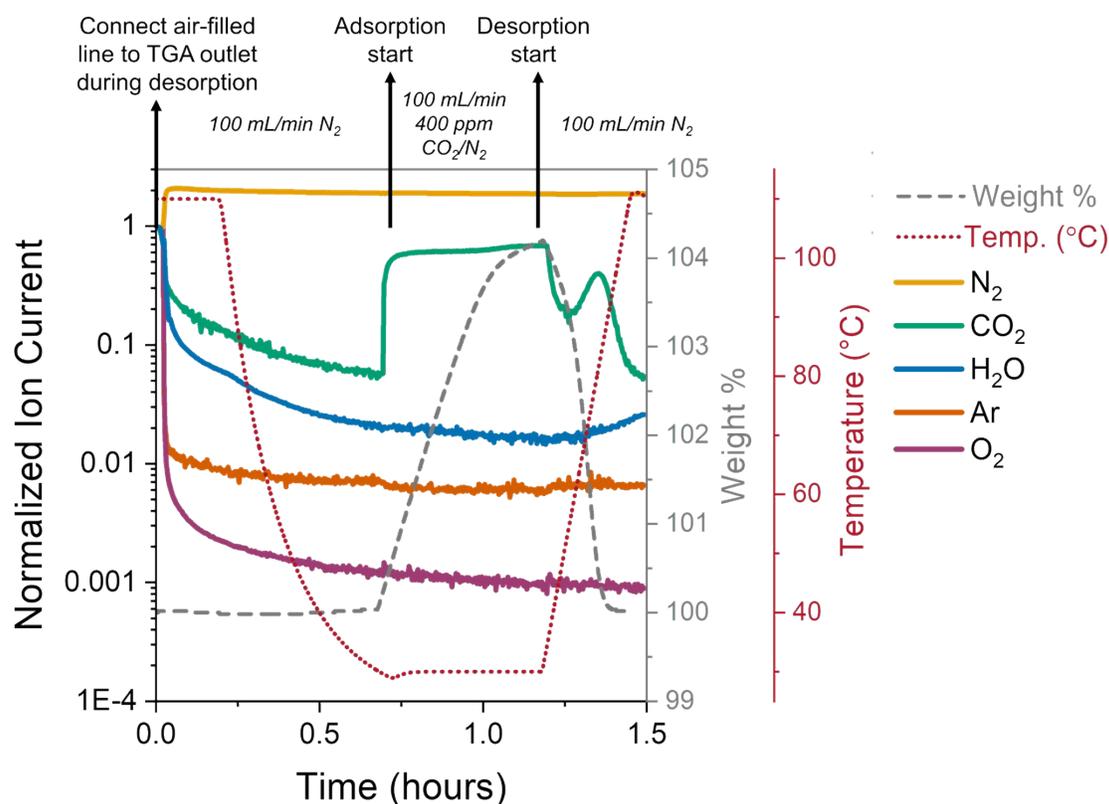
Figure S9. **Gases inside TGA during adsorption-desorption cycles.** The outlet of the TGA was connected to a Pfeiffer Omnistar mass spectrometer during the second adsorption-desorption cycle of the unmodified PEI in Fig. 6b. The measured ion currents (left y-axis) are normalized to the value at the start of the measurement (assumed to be lab air). The sample weight is also normalized to the value at the start of the measurement (weight %, right y-axis).

S3. DETAILED ECONOMIC MODEL METHODS

This section describes the economic model used for the analysis in **Figures 2-4, 6-7**. The levelized cost of CO_2 capture (LCOC) is the primary metric from the economic model. It is comprised of the capital cost ($C_{capital}$), operation & maintenance cost ($C_{O\&M}$), sorbent cost ($C_{sorbent}$), and the cumulative amount of CO_2 captured N_{CO_2} (equation S1).

$$LCOC = \frac{Q_{capital}^{N_{CO_2}} + C_{O\&M} + C_{sorbent}}{N_{CO_2}} \#(S1)$$

The following sections (S3.A – S3.K) provide more details on the DAC economic model and how the LCOC is calculated. Sections 3.A, 3.B, and 3.C focus on setting up the system, including flow rates and sizing. Section S3.D describes how to calculate the cumulative amount of CO_2 captured both with and without sorbent degradation. Section 3.E uses the sorbent amount from S3.B to calculate the total sorbent cost ($C_{sorbent}$). Section 3.F summarizes the components of the total capital cost and provides the scaling relationships used in sections S3.G, S3.H and S3.I, which focus on the four primary components of capital cost: contactor, blowers, vacuum pumps, auxiliary equipment, and downstream processing. Section S3.J provides an overview of the total operation & maintenance cost, including specifics on the non-energy cost components. The energy component of operations & maintenance cost is then covered in section S3.K, including specifics on how to account for sorbent degradation. Finally, section S3.L focuses on how to calculate the carbon footprint and include it in the LCOC.

S3.A. Feed and Initial Product Flow Rates

The first task in the model is setting up the feed and product flow rates, which will affect all other cost components. The target amount of CO_2 captured in one year (before sorbent degradation is considered) is an input in the model, and the system flow rates will depend on that value. The flow rate of air feed into the system ($m_{air,feed}$) depends on the total volumetric blower flow rate ($q_{air,feed}$) and air density ($\rho_{air,feed}$). The total volumetric blower flow rate will be calculated in S3.B in eq. S17.

$$m_{air,feed}\left[\frac{kg}{day}\right] = q_{air,feed}\left[\frac{m^3}{s}\right] \cdot \rho_{air,feed}\left[\frac{kg}{m^3}\right] \cdot \left[\frac{3600\ s}{hr}\right]\left[\frac{24\ hr}{day}\right] \#(S2)$$

The CO_2 feed flow rate ($m_{CO2,feed}$) is calculated using the CO_2 concentration in air ($x_{CO_2,\ feed}$) and the flow rate from eq. S2 ($m_{air,feed}$).

$$m_{CO2,feed}\left[\frac{kg}{day}\right] = x_{CO_2,\ feed}\left[\frac{kg\ CO_2}{kg\ air}\right] \cdot m_{air,feed}\left[\frac{kg}{day}\right] \#(S3)$$

Similarly, the N_2 feed flow rate depends on the N_2 concentration in air ($x_{N_2,\ feed}$) and the flow rate of air.

$$m_{N2,feed}\left[\frac{kg}{day}\right] = x_{N_2,\ feed}\left[\frac{kg\ N_2}{kg\ air}\right] \cdot m_{air,feed}\left[\frac{kg}{day}\right] \#(S4)$$

The initial flow rate of CO_2 product (i.e., the flow rate without sorbent degradation or before it occurs) ($m_{CO_2,product}^{k=0}$) depends on the amount of CO_2 captured in one cycle $n_{CO_2}^{k=0}(t_i)$ and the number of cycles per day (N_{cycles}).

$$m_{CO_2,product}^{k=0}\left[\frac{kg}{day}\right] = n_{CO_2}^{k=0}(t_i)\left[\frac{kg}{cycle}\right] \cdot N_{cycles}\left[\frac{cycles}{day}\right] \#(S5)$$

The number of cycles per day (N_{cycles}) depends on the utilization fraction (U) and the cycle time (T_{cycle}). The utilization fraction is discussed more in section S3.F; it accounts for the time that the plant is not in operation. A lower utilization fraction will reduce the number of cycles in one year. Thus, to capture the same amount of CO_2 in one year (which is the target of the model), more CO_2 will need to be captured each cycle.

$$N_{cycles}\left[\frac{cycles}{day}\right] = \frac{\left[\frac{86400\ s}{day}\right] \cdot U\left[\frac{operational\ time}{total\ time}\right]}{T_{cycle}\left[\frac{s}{cycle}\right]} \#(S6)$$

The total desorption product flow rate ($m_{product}$) depends on the CO_2 product flow rate $(m_{CO2,product})$ and the CO_2 concentration in the product (i.e., purity) ($x_{CO_2,\ product}$).

$$m_{product} = \frac{m_{CO2,product}}{x_{CO_2,\ product}} \#(S7)$$

The instantaneous recovery ($X_{R,i}$) is the recovery measured at one cycle; it does not consider the amount captured in previous cycles. If the sorbent is degrading, the instantaneous recovery will change every cycle. It can be calculated from the amount of CO_2 captured in the cycle that the instantaneous recovery is being evaluated at ($n_{CO_2}(t_i)$) and the amount of CO_2 fed in that cycle ($n_{CO_2,\ feed} / N_{cycles}$).

$$X_{R,i} = \frac{n_{CO_2}(t_i)\left[\frac{kg\ CO_2}{cycle}\right]}{n_{CO_2,\ feed}\left[\frac{kg\ CO_2}{day}\right] \cdot \frac{1}{N_{cycles}}\left[\frac{day}{cycles}\right]} \#(S8)$$

The amount of CO_2 captured in a specific cycle ($n_{CO_2}(t_i)$) depends on the total mass of sorbent (m_S) and working capacity at that cycle ($q_{\delta,i}$). Calculating the total mass of sorbent is described in S3.B in eq. S10.

$$n_{CO_2}(t_i)\left[\frac{kg\ CO_2}{cycle}\right] = m_S\ [kg\ sorbent] \cdot q_{\delta,i}\left[\frac{mol\ CO_2}{kg\ sorbent}\right] \cdot \left[\frac{0.044\ kg}{mol\ CO_2}\right] \#(S9)$$

Table S3. Summary of "Feed and Product Flow Rates" section of model. Input variables from S1 are used with no sorbent degradation.

Parameter	Input or Equation	Variable	Value	Unit
AIR FEED				
Air feed total flow rate	Eq. S2	$m_{air,feed}$	8.82×10^9	kg/day
CO_2 feed concentration	Input	$x_{CO_2,\ feed}$	0.00061	kg CO_2/ kg air feed
CO_2 feed flow rate	Eq. S3	$m_{CO2,feed}$	5.48×10^6	kg/day
H_2O feed concentration	Input	$x_{H_2O,\ feed}$	0.01397	kg H_2O/ kg air feed
H_2O feed flow rate		$m_{H2O,feed}$	1.22×10^8	
N_2 feed concentration	Input	$x_{N_2,\ feed}$	0.745	kg N_2/ kg air feed
N_2 feed flow rate	Eq. S4	$m_{N2,feed}$	6.57×10^9	kg/day

DESORPTION PRODUCT				
Desorption product total flow rate	Eq. S7	$m_{product}$	2.88 x 10^6	kg/day
CO_2 product concentration	Input	$x_{CO_2,\ product}$	0.95	fraction
CO_2 product flow rate if no degradation	Eq. S5	$m_{CO_2,product}^{k=0}$	2.74 x 10^6	kg/day
Instantaneous recovery (cycle 1 shown as example)	Eq. S8	$X_{R,i}$	0.50	fraction

Table S4. Summary of "Amount of CO_2 Captured" section of model. Input variables from S1 are used with no sorbent degradation.

Parameter	Input or Equation	Variable	Value	Unit
CO_2 captured if no degradation	Input	$m_{CO_2,product}$	1,000,000	t-CO_2/yr
			2.74 x 10^6	kg/day
Utilization	Input	U	0.90	fraction
Number of cycles per day	Eq. S6	N_{cycles}	43.2	cycles/day
CO_2 captured in current cycle (cycle 1 shown as example)	Eq. S9	$n_{CO_2}(t_i)$	6.34 x 10^4	kg/cycle
			63.42	tonne/cycle
			6.34 x 10^7	g/cycle
			1.44 x 10^6	mol/cycle
CO_2 captured per cycle if no degradation	Eq. S5 or Eq. S9 at t = 1	$n_{CO_2}^{k=0}(t_i)$ or $n_{CO_2}(t=1)$	63.42	tonne/cycle
			1.44 x 10^6	mol/cycle

Note: The flow rates and CO_2 captured will vary with the input variables. The above values are one example.

S3.B. Amount of Sorbent & Module Sizing

The adsorbent is assumed to be in a polymeric structured contactor. We will use the following terminology throughout this work:

- **Sorbent:** active material that is binding CO_2
- **Contactor**: sorbent and inactive material (e.g., polymer) formed into a geometry (e.g., fiber, monolith, laminate)
- **Module**: entire capture unit (sorbent, polymer, stainless steel housing)
- **Cluster**: several capture units that share vacuum pump(s) and blower(s); modules in clusters are on an adsorption/desorption schedule
- **Array:** multiple clusters in one area

The model is agnostic to the geometry of the contactor (i.e., fibers, monolith, laminate) but does consider the sorbent loading inside the contactor (i.e., sorbent to polymer mass fraction), contactor density, module void fraction, module cross-sectional area, and module length.

The mass of necessary adsorbent depends on the desired product flow rate (not considering sorbent degradation) ($m_{CO_2,product}^{k=0}$), number of cycles per day (N_{cycles}), and initial working capacity of the sorbent ($q_{\delta,0}$).

$$m_S\ [kg\ sorbent] = \dfrac{m_{CO_2,product}^{k=0}\left[\dfrac{kg\ CO_2}{day}\right]\cdot \dfrac{1}{N_{cycles}}\left[\dfrac{day}{cycles}\right]\cdot \left[\dfrac{mol\ CO_2}{0.044\ kg}\right]}{q_{\delta,0}\left[\dfrac{mol\ CO_2}{kg\ sorbent}\right]}\#(S10)$$

The mass of polymer is determined using the mass of sorbent from eq. S10 (m_S) and the sorbent loading fraction inside the contactor ($\varnothing_s$).

$$m_P\ [kg\ polymer] = \left(\dfrac{m_S}{\varnothing_s}\right)\cdot (1-\varnothing_s)\#(S11)$$

The mass of the structured contactor includes both the adsorbent and polymer. The structured contactor volume is estimated using eq. S12, where V_{SC} is volume of structured contactor, m_S and m_P are the mass of sorbent and polymer, and ρ_{SC} is the density of contactor. In cases where the contactor has porosity, such as porous polymer supports, the porosity should be included in the density used.

$$V_{SC}\ [m^3] = \dfrac{(m_s + m_p)\ [kg\ contactor]}{\rho_{SC}\left[\dfrac{kg\ contactor}{m^3}\right]}\#(S12)$$

The total cross-sectional area of the contactors (again referring to only the sorbent and polymer, not the module cross-sectional area) is the total contactor volume from eq. S12 (V_{SC}) divided by the module length (which is the same as the contactor length). Longer module lengths will lead to smaller facial areas (with a constant sorbent volume) and higher gas velocities, while shorter module lengths will lead to larger facial areas and lower gas velocities.

$$A_{total,SC}[m^2] = \dfrac{V_{SC}[m^3]}{l_{module}\ [m]}\#(S13)$$

The modules are modeled here as square cuboids. The individual module cross-sectional area (A_{module}) is therefore a function of the side length of the square faces (d_{module}). To change the geometry of the module, Eq. S14 should be modified.

$$A_{module}[\ m^2] = (d_{module}[m])^2\#(S14)$$

The total cross-sectional area of the modules ($A_{total,\ modules}$) is estimated using the cross-sectional area of the contactors from eq. S13 ($A_{total,\ SC}$) and a set void fraction of the module (φ_{module}). The void fraction is the ratio of empty volume to total volume in the module. The void fraction results from incomplete packing of contactor into the module housing.

$$A_{total,\ modules}\ [m^2] = \dfrac{A_{total,\ SC}\ [m^2]}{(1-\varphi_{module})}\#(S15)$$

The number of modules ($N_{modules}$) is calculated by dividing the total cross-sectional area from eq. S15 ($A_{total,\ modules}$) by the cross-sectional area of one module from eq. S14 (A_{module}).

$$N_{modules} = \dfrac{A_{total,\ modules}\ [m^2]}{A_{module}\ [m^2]}\#(S16)$$

The total volumetric blower flow rate ($q_{air, feed}$) is the total module cross-sectional area ($A_{total, modules}$) multiplied by the superficial velocity (v). The total volumetric blower flow rate was used in the previous section to determine the flow rate of air into the system. Superficial velocity is an input that is varied to maintain the recovery at a set value. The superficial velocity is lower than the interstitial velocity (velocity in the channels) due to the solid fraction of the module. The relationship between module size, superficial velocity, and recovery is discussed more in section S3.C.

$$q_{air, feed} = A_{total, modules} \, [m^2] \cdot v \left[\frac{m}{s} \right] \#(S17)$$

Table S5. Summary of "Amount of Sorbent & Module Sizing" section of model. Input variables from S1 are used with no sorbent degradation.

Parameter	Input or from Eq.	Variable	Value	Unit
Initial sorbent working capacity	Input	$q_{\delta,0}$	1.0	mol/kg sorbent
Mass of sorbent	Eq. S10	m_S	1,441	tonne
Sorbent wt. fraction in contactor	Input	$\emptyset_s$	0.50	fraction
Mass of polymer	Eq. S11	m_P	1,441	tonne
Contactor density	Input	ρ_{SC}	730	kg/m³
Contactor volume	Eq. S12	V_{SC}	3,951	m³
Module diameter	Input (see S3.C)	d_{module}	1.00	m
Module length	Input (see S3.C)	l_{module}	0.38	m
Module cross sectional area	Eq. S14	A_{SC}	1.00	m²
Total contactor cross-sectional area	Eq. S13	$A_{total,SC}$	10,374	m²
Module void fraction	Input	φ_{module}	0.50	
Total module cross-sectional area	Eq. S15	$A_{total,modules}$	20,747	m²
Number of modules	Eq. S16	$N_{modules}$	20,747	
Superficial velocity	Input (see S3.C)	v	4	m/s
Total blower flow rate	Eq. S17	$q_{air,feed}$	82,989	m³/s

Note: The module size will vary with the input variables. The above values are one example.

S3.C. Relationship Between Velocity, Recovery, and Module Size

When performing an economic analysis without inputs from a process model, it is important to make sure that variables remain constrained within physically reasonable bounds. For the results shown here, the initial CO_2 recovery is fixed at 50%. Superficial velocity, module diameter, module length, number of contactors per blower, and individual blower flow rate are all related to the recovery, so these can be varied to maintain the recovery at the set value, but they should be varied within physically reasonable constraints. Additionally, if all of these parameters are varied, there will be multiple solutions. To have only one solution, it is necessary to fix three out of the five variables. Following the blower and vacuum pump scheduling scheme in Figure S2, we fixed the number of contactors per blower as 20. We also fixed the module diameter as 1.0 m and individual blower flow rate as 80 m³/s. The superficial velocity and module length are then varied to achieve a recovery of 50%.

Table S6. Solver parameters and constraints.

Variable	Constraint
Recovery	= 0.50
Superficial Velocity	Between: 2 m/s - 6 m/s [4 m/s used in analysis, except for Fig. 7 best-case scenario which uses 5.4 m/s (see note on SI pg. 7)]
Module Diameter	Between: 0.5 – 1.5 m [1.0 m used in analysis, except for Fig. 7 best case scenario which uses 0.86 m (see note on SI pg. 7)]
Module Length	Between: 0.1 – 1.5 m
Number of contactors per blower	= 20
Individual Blower Flow Rate	Less than: 100 m³/s [80 m³/s used in analysis]

S3.D. Cumulative Amount of CO₂ Captured for Different Forms of Degradation

The cumulative amount of CO_2 captured depends on the working capacity in each cycle. The working capacity ($q_{\delta,i}$) can be written as a function of the initial working capacity ($q_{\delta,0}$), a degradation decay constant (k) that corresponds to the degradation rate, and time (t_i). Three working capacity functions are shown below in Eq. S19, S21, and S24 for three forms of degradation: linear, exponential, and piecewise exponential. The working capacity in these equations is integrated over the sorbent's lifetime to find the cumulative amount of CO_2 captured in an en-masse replacement method (i.e., when the sorbent is replaced all at once).

The current sorbent lifetime [t_i] is the cycle time [T_{cycle}] multiplied by the number of cycles [N_{cycles}]. To convert from cycles to clock time, it is divided by the utilization [U] to account for the plant not always being in operation.

$$t_i \, [year] = T_{cycle} \left[\frac{s}{cycle}\right] \cdot N_{cycles} \, [cycles] \cdot \left[\frac{hr}{3600 \, s}\frac{day}{24 \, hr}\frac{year}{365 \, days}\right] \cdot \frac{1}{U} \#(S18)$$

Linear Degradation

Working capacity:

$$q_{\delta,i} \left[\frac{mol}{kg}\right] = q_{\delta,0} \left[\frac{mol}{kg}\right] - k \, [cycle^{-1}] \cdot t_i \, [cycles] \#(S19)$$

Cumulative amount of CO₂ captured (integral of eq. S19):
There are three equations: (1) if the sorbent is not degrading, (2) if the sorbent is degrading but the capacity is not yet zero, and (3) if the sorbent capacity has already reached zero.

$$N_{CO_2, \, i}(t_i) = \begin{cases} N^*_{CO_2} \cdot t_i, & if \; k = 0 \\[2ex] m_S \left(q_{\delta,0} \cdot t_i - \dfrac{k \cdot t_i^2}{2} \right), & if \; k \neq 0 \; and \; t_i < \dfrac{q_{\delta,0}}{k} \\[3ex] m_S \left(q_{\delta,0} \cdot \dfrac{q_{\delta,0}}{k} - \dfrac{k \cdot \left(\frac{q_{\delta,0}}{k}\right)^2}{2} \right), & if \; k \neq 0 \; and \; t_i > \dfrac{q_{\delta,0}}{k} \end{cases} \#(S20)$$

Exponential Degradation

Working capacity:

$$q_{\delta,i} \left[\frac{mol}{kg}\right] = q_{\delta,0} \left[\frac{mol}{kg}\right] \cdot e^{-k \, [cycle^{-1}] \cdot t_i \, [cycles]} \#(S21)$$

Cumulative amount of CO₂ captured (integral of eq. S21):
There are two equations: (1) if the sorbent is not degrading and (2) if the sorbent is degrading.

$$N_{CO_2, \, i}(t_i) = \begin{cases} N^*_{CO_2} \cdot t_i, & if \; k = 0 \\[2ex] m_S \dfrac{q_{\delta,0}}{k} (1 - e^{-k \cdot t_i}), & if \; k \neq 0 \; and \; t_i < \dfrac{q_{\delta,0}}{k} \end{cases} \#(S22)$$

If a partial replacement method is used where a portion of the sorbent is replaced each cycle at some fixed replacement rate m_r (kg sorbent/cycle), then eq. S23 can be used.

$$N_{CO_2, i} = q_{\delta,0} m_s \left(\frac{t_i \cdot \dot{m}_r}{\dot{m}_r + m_s k} + \frac{m_s}{\dot{m}_r + m_s k} \left(1 - e^{-(\dot{m}_r + m_s k) \cdot t_i} \right) + \frac{m_s \dot{m}_r}{\dot{m}_r + m_s} \left(1 - e^{-(\dot{m}_r + m_s k) \cdot t_i} \right) \right) \#(S23)$$

Piecewise exponential (two part)

Working capacity:

$$q_{\delta,i} = \begin{cases} q_{\delta,0} e^{-k_1 \cdot t}, & t < t_s \\ q_{\delta,t_s} e^{-k_2 \cdot t}, & t \geq t_s \end{cases} \#(S24)$$

Cumulative amount of CO₂ captured:

$$N_{CO_2, i}(t_i) = \begin{cases} N_{CO_2}^* \cdot t_i, & if\ k = 0 \\ m_s \dfrac{q_{\delta,0}}{k_1} \left(1 - e^{-k_1 \cdot t_i} \right), & if\ k \neq 0\ and\ t_i < t_s \\ m_s \dfrac{q_{\delta,t_s}}{k_2} \left(1 - e^{-k_2 \cdot (t_i - t_s)} \right) + m_s \dfrac{q_{\delta,0}}{k_1} \left(1 - e^{-k_1 \cdot t_s} \right), & if\ k \neq 0\ and\ t_i > t_s \end{cases} \#(S25)$$

These can be compared to the cumulative of CO₂ captured without degradation $(N_{CO_2}^{K=0})$, which is simply the set capture rate $(N_{CO_2}^*)$ multiplied by the current sorbent lifetime (t_i).

The cumulative recovery at a specific time is the ratio of the cumulative amount of CO₂ captured from eq. S20, S22, S23, or S25 $(N_{CO_2, i})$ to the cumulative amount of CO₂ fed $(m_{CO2, feed} \cdot t_i)$.

$$X_{R,cumulative} = \frac{N_{CO_2, i} \left[\frac{tonne\ CO_2}{lifetime} \right]}{m_{CO2, feed} \left[\frac{kg}{day} \right] \cdot t_i \left[\frac{year}{lifetime} \right] \cdot \left[\frac{365\ day}{year} \frac{tonne}{1000\ kg} \right]} \#(S26)$$

Table S7. Summary of "Cumulative Amount of CO₂ Captured" section of model. Input variables from S1 are used with no sorbent degradation.

Parameter	Input or from Eq.	Variable	Value	Unit
Current sorbent capacity	Eq. S19, S21, S24	$q_{\delta,i}$	1.0	mol/kg sorbent
Cycle time	Input	T_{cycle}	1800	s
Current sorbent lifetime (chosen as example)	Eq. S18	t_i	1.04	year
			16,474	cycles
Cumulative CO₂ captured at current sorbent lifetime	Eq. S20, S21, S3, S25	$N_{CO_2, i}$	1,044,774	t-CO₂/lifetime
Desired annual CO₂ captured	Input	$N_{CO_2}^*$	1,000,000	t-CO₂/year
Cumulative CO₂ captured without degradation		$N_{CO_2}^{K=0}$	1,044,774	t-CO₂/lifetime
Cumulative recovery	Eq. S26	$X_{R,cumulative}$	0.50	

Note: The CO_2 captured will vary with the input variables. The above values are one example.

S3.E. Total Sorbent Cost

Because this analysis focuses on amine-supported materials in structured polymeric contactors, the cost of the sorbent is only the active material (amine) and the labor cost for replacement. The sorbent replacement method is washing the amines out and then re-infusing them[4], so the support (silica) and contactor (polymer) are annualized as capital as they will not be replaced at the same time.

The sorbent material cost ($C_{sorbent, material}$) is the bare module cost ($BM_{S,1}$) multiplied by the amount of sorbent required ($m_{s,1}$) (calculated in Eq. S10). The cumulative amount of CO_2 captured (N_{CO_2}) already has utilization factored in.

$$C_{sorbent, material}\left[\frac{\$}{tonne\ CO_2}\right] = (BM_{S,1} \cdot m_{s,1})\left[\frac{\$}{lifetime}\right] \cdot \frac{1}{N_{CO_2}}\left[\frac{lifetime}{tonne\ CO_2}\right] \#(S27)$$

The sorbent replacement labor cost ($C_{replacement}$) is accounted for using a replacement cost per module ($C_{rep., module}$), number of modules ($N_{modules}$), and number of replacements per year ($\frac{1}{t_{life}}$).

$$C_{replacement}\left[\frac{\$}{tonne\ CO_2}\right] = \left(C_{rep., module} \cdot N_{modules} \cdot \frac{1}{t_{life}}\right)\left[\frac{\$}{year}\right] \cdot \frac{1}{N_{CO_2}}\left[\frac{year}{tonne\ CO_2}\right] \#(S28)$$

The total sorbent cost is the sum of the sorbent material and replacement costs.

$$C_{sorbent} = C_{sorbent, material} + C_{replacement} \#(S29)$$

Table S8. Summary of "Sorbent Cost" section of model. Input variables from S1 are used with no sorbent degradation. Parameters are evaluated at a sorbent lifetime of 1.04 years.

Parameter	Input or from Eq.	Variable	Value	Unit
Polyamine bare module cost	Input	$BM_{S,1}$	3.00	$/kg
	Eq. S27	$BM_{S,1} \cdot m_{s,1}$	1,780,092	$/lifetime
	Eq. S27	$\dfrac{BM_{S,1} \cdot m_{s,1}}{N_{CO_2}}$	1.70	$/t-CO$_2$
Number of replacements per year		$\dfrac{1}{t_{life}}$	0.96	
Number of module replacements per year	Portion of Eq. S28	$N_{modules} \cdot \dfrac{1}{t_{life}}$	19,858	
Replacement cost per module	Portion of Eq. S28	$C_{rep., module}$	5.00	$/module
Replacement cost per year	Portion of Eq. S28	$C_{rep., module} \cdot N_{mo}$	99,291	$/year
	Eq. S28	$C_{replacement}$	0.10	$/t-CO$_2$
Total sorbent cost	Eq. S29	$C_{sorbent}$	1.80	$/t-CO$_2$

Note: The amount of sorbent will vary with the input variables. The above values are one example.

S3.F. Total Capital Cost

The total capital cost is the sum of direct capital, indirect capital, piping, and contingency. The engineering economic analysis procedure from Turton et al. was followed, and the module costing technique was used for the contactor, blower, and vacuum pump costs.[5] A summary of the primary capital cost inputs is provided in Table S9.

$$C_{total\ capital} = direct\ capital + indirect\ capital + piping + contingency \#(S30)$$

Table S9. Capital cost inputs.

Parameter	Value
Plant capital recovery factor (CRF)	0.16
Plant utilization	0.90
Indirect capital cost	35% of DC
Contingency	30% of DC
Piping	40% of DC

Direct capital

The direct capital includes the purchase and installation costs of the equipment (discussed further in S3.G, S3.H, and S3.I) and is scaled by the capital recovery factor (CRF) and utilization (equation S29) to obtain the levelized capital ($/year).

$$C_{levelized\ capital}\left[\frac{\$}{year}\right] = C_{total\ capital}[\$] \times \frac{CRF}{Utilization}\#(S31)$$

The CRF is a function of the interest rate (*i*) and the number of annual payments (*N*) (eq. S32). The utilization is a ratio of operating capacity to design capacity (eq. S33). Multiplying the total capital by the CRF converts the total capital ($/plant lifetime) into annual payments ($/year). Dividing the total capital by utilization scales the total capital from an operating capacity to a design capacity.

$$CRF = \frac{i(1+i)^N}{(1+i)^N - 1}\#(S32)$$

$$Utilization = \frac{Operating\ capacity}{Design\ capacity}\#(S33)$$

Since the amount of CO_2 captured varies between years when sorbent degradation is included, the capital cost and amount of captured CO_2 are calculated per sorbent lifetime rather than per year. The levelized capital is reported in units of $/lifetime by scaling the capital by the number of years in the sorbent's lifetime (t_{life}).

$$C_{levelized\ capital}\left[\frac{\$}{t-CO_2}\right] = \left(C_{total\ capital} \cdot \frac{CRF}{Utilization}\right)\left[\frac{\$}{yr}\right] \cdot t_{life}\left[\frac{yr}{lifetime}\right] \cdot \frac{1}{N_{CO_2}}\left[\frac{lifetime}{t-CO_2}\right]\#(S34)$$

In the DAC plant, the direct capital includes contactors, blowers, vacuum pumps, auxiliary equipment, and downstream compression and purification. These are discussed in more detail in S3.G, S3.H, and S3.I.

$$C_{direct\ capital} = C_{contactors} + C_{blowers} + C_{vacuump\ pumps} + C_{auxiliary} + C_{downstream}\#(S35)$$

A scaling law was used to scale reference values or quotes to the equipment capacity and year of this project (2022),[8,10]

$$CC_{a,2} = CC_{b,1}\left(\frac{A_a}{A_b}\right)^n\left(\frac{I_2}{I_1}\right)\#(S36)$$

where "CC" is the installation cost (installation factors already included), "A" is an equipment attribute or capacity, and "I" is the chemical engineering plant cost index (CEPCI). Subscripts *"a"* and *"2"* refer to this DAC design, and *"b"* and *"1"* refer to the original source.

Indirect capital, contingency, and piping

Indirect capital covers the transportation of equipment, insurance, purchase taxes, construction overhead, contractor engineering expenses, etc. Contingency includes any unforeseen expenses. Indirect capital, contingency, and piping were set as 35%, 30%, and 40% of the direct capital, respectively. These are added to the direct capital cost to obtain the total capital (eq. S30)

$$C_{indirect\ capital} = 0.35 \cdot direct\ capital\ \#(S37)$$

$$C_{contingency} = 0.30 \cdot direct\ capital\ \#(S38)$$

$$C_{piping} = 0.40 \cdot direct\ capital\ \#(S39)$$

Table S10. Summary of "CAPEX Totals" section of model. Input variables from S1 are used with no sorbent degradation. Parameters are evaluated at a sorbent lifetime of 1.04 years.

Parameter	Input or from Eq.	Variable	Value	Unit
Direct Capital	Eq. S35	$C_{direct\ capital}$	477,377,192	$
Indirect Capital	Eq. S37	$C_{indirect\ capital}$	167,082,017	$
Contingency	Eq. S38	$C_{contingency}$	143,213,158	$
Piping	Eq. S39	C_{piping}	190,950,877	$
Total Capital (non-annualized)	Eq. S30	$C_{total\ capital}$	978,623,243	$
Levelized Capital	Eq. S31	$C_{levelized\ capital}$	481,159,710	$/yr
Levelized Capital	Eq. S34	$C_{levelized\ capital}$	179.70	$/t-CO$_2$

Note: The plant capacity and capital will vary with the input variables. The above values are one example.

S3.G. Contactor Cost

The cost of the contactor includes the materials, manufacturing, and installation costs and is annualized.

$$C_{contactor} = (BM_{S,2} \cdot m_{s,2} + BM_C \cdot m_C) \cdot F_{BM,C} \#(S40)$$

where $BM_{S,2}$ and BM_C are the support and contactor system bare module cost, respectively, and $F_{BM,C}$ is the contactor's installation or bare module cost factor. The bare module cost factor accounts for factors associated with installation of the equipment, including (1) materials required for installation, (2) labor to install the equipment and material, (3) freight, insurance, and taxes, (4) construction overhead, (5) contractor engineering expenses, (6) contingency, (7) contractor fee, (8) site development, (9) auxiliary buildings, and (10) off-site and utilities.

Table S11. Summary of "Contactor Cost" section of model. Input variables from S1 are used with no sorbent degradation. Parameters are evaluated at a sorbent lifetime of 1.04 years.

Parameter	Input or from Eq.	Variable	Value	Unit
Amine support (silica) bare module cost	Input	$BM_{S,2}$	2.00	$/kg
	Portion of Eq. S40	$BM_{S,2} \cdot m_{s,2}$	1,695,326	$/lifetime
Contactor bare module cost	Input[6]	BM_C	25.00	$/kg
	Portion of Eq. S40	$BM_C \cdot m_C$	36,082,841	$/lifetime
Contactor bare module cost factor	Input	f_C	3.00	
Installed contactor cost	Portion of Eq. S40	$BM_C \cdot m_C \cdot f_C$	113,334,801	$/lifetime
Total contactor cost	Eq. S40	$C_{contactor}$	65.23	$/t-CO_2

Note: The contactor amount will vary with input variables. The above values are one example.

S3.H. Blower & Vacuum Pump Capital Cost

The total blower flow rate ($q_{air,feed}$) was calculated in eq. S17. The number of necessary blowers ($N_{blowers}$) is the total blower flow rate divided by a set individual blower flow rate (q_{blower}) (see section S3.C).

$$N_{blowers} = \frac{q_{air,feed}}{q_{blower}} \#(S41)$$

The number of modules per blower ($N_{modules}/_{blower}$) is determined by dividing the number of modules by the number of blowers. The number of blowers and vacuum pumps to the number of modules is determined by the configuration of modules and blowers. To reduce the number of modules, the modules are arranged in a circle around a central blower and vacuum pumps. Shown in Fig. S2, one blower and two vacuum pumps can be utilized for 20 modules. The superficial velocity and module length were varied to maintain $N_{modules}/_{blower}$ at 20 and q_{blower} at a set value (see section S3.C).

$$N_{modules}/_{blower} = \frac{N_{modules}}{N_{blowers}} \#(S42)$$

The purchase cost (C_P°) of each blower and vacuum pump follows the form in eq. S43

$$log_{10} C_P^\circ = K_1 + K_2 log_{10}(A) + K_3 (log_{10}(A))^2 \#(S43)$$

where A is the capacity parameter, which is the gas flowrate [m^3/s] for blowers and the shaft power (kW) for vacuum pumps. The parameters (K_1, K_2, K_3) are from Turton et al.[5] and are listed in Table S11.

The purchase cost (C_P°) is converted to the bare module cost (C_{BM}) using a bare module factor (F_{BM}); the bare module factors are also in Table S11. The bare module cost is then scaled from the reference year (2001 for the blower and vacuum pump reference text) to the current project year (2022) using eq. S36. The bare module factor contains indirect expenses and contingency, so they are not applied to the blowers and vacuum pumps again when computing the total capital cost.

$$C_{BM} = F_{BM} \cdot C_P^\circ \#(S44)$$

The total cost of the blowers or vacuum pumps ($C_{blowers}, C_{vacuum\,pumps}$) is the bare module cost (C_{BM}) of each multiplied by the number of blowers or vacuum pumps ($N_{blowers}, N_{vacuum\,pumps}$).

$$C_{blowers} (or\ C_{vacuum\,pumps}) = C_{BM} \cdot N_{blowers} (or\ N_{vacuum\,pumps}) \#(S45)$$

The number of vacuum pumps per blower ($N_{vac\,pump}/_{blower}$) is an input value based on the scheduling discussed previously (Fig. S2). The number of vacuum pumps ($N_{vacuum\,pumps}$) is calculated using equation S46.

$$N_{vacuum\,pumps} = N_{vac\,pump}/_{blower} \cdot N_{blowers} \#(S46)$$

Determining the total shaft power of the vacuum pumps ($E_{VP,total}$) is discussed in section S3.K. The shaft power of individual vacuum pumps ($E_{VP,individual}$) is calculated using the total shaft power and the number of vacuum pumps from Eq. S46. The individual shaft power is used as the capacity in the cost equation (eq. S43).

$$E_{VP,individual} = \frac{E_{VP,total}}{N_{vacuum\ pumps}} \#(S47)$$

Table S12. Blower and vacuum pump cost equipment parameters used with Eq. S43.[5]

Parameter	Axial vane fan	Positive displacement pump
K1	3.1761	3.4471
K2	-0.1373	0.1350
K3	0.3413	0.1438
Min Size (m^3/s)	1	1
Max Size (m^3/s)	100	100
Bare Module Factor	4	3

Table S13. Summary of "Blower & Vacuum Pump CAPEX" section of model. Input variables from S1 are used with no sorbent degradation. Parameters are evaluated at a sorbent lifetime of 1.04 years.

Parameter	Input or from Eq.	Variable	Value	Unit
Blowers				
Total blower flow rate	Eq. S17	$q_{air,\,feed}$	82,989	m^3-air/s
Individual blower flow rate	Input (see S3.C)	q_{blower}	80	m^3/s
Number of blowers	Eq. S41	$N_{blowers}$	1,037	
Number of modules per blower	Eq. S42	$N_{modules/blower}$	20	
purchased cost, Cp	Eq. S43	C_P°	14,167	$/blower
bare module cost	Eq. S44	C_{BM}	56,667	$/blower
Total installed cost, 2001	Eq. S45	$C_{blowers}$	58,784,579	$
Total installed cost, 2022	Eq. S36	$C_{blowers}$	121,654,113	$
Levelized capital cost of blowers		$C_{levelized,\,blowers}$	30.28	$/t-CO$_2$
Vacuum Pumps				
Number of vacuum pumps per blower	Input	$N_{vac\,pump/blower}$	2	
Number of vacuum pumps	Eq. S43	$N_{vacuum\,pumps}$	2,075	
total kw		$E_{VP,total}$	266.91	kW
Individual vacuum pump shaft power	Eq. S47	$E_{VP,individual}$	0.13	kW
purchased cost, Cp	Eq. S43	C_P°	2,789	$/pump
bare module cost	Eq. S44	C_{BM}	8,366	$/pump
Total installed cost, 2001	Eq. S45	$C_{vacuum\,pumps}$	17,357,947	$
Total installed cost, 2022	Eq. S36	$C_{vacuum\,pumps}$	35,922,102	$
Levelized capital cost of vacuum pumps		$C_{levelized,\,vac\,pumps}$	8.94	$/t-CO$_2$

Note: The blower and vacuum pump capacity will vary with the input variables. The above values are one example.

S3.I. Auxiliary & Downstream Compression & Purification Capital Cost

Values from references were scaled to the current capacity and project year using eq. S36.

The downstream compression and purification system cost is from a quote provided by Trimeric, Inc.. The system is based on a liquefication/distillation process for removing inert species (O_2) from the CO_2. Trimeric used the QGESS design values of CO_2 (> 95 mol%), H_2O (< 500 ppmv), N_2 and Ar (< 1 to 4 mol%), and O_2 (< 10 ppmv).

The auxiliary equipment includes (1) sorbent handling, (2) sorbent preparation, storage, and feed, (3) cooling water system, (4) spend sorbent handling system, (5) instrumental control, (6) site preparation, improvements, facilities, (7) buildings & structures (admin., machine shop, warehouse, etc.), (8) steam distribution system, and (9) system controls equipment. The costs are the bare erected costs from the "Direct Air Capture Case Studies: Sorbent System" report from NETL (Exhibit 6-8).[3]

Table S14. Summary of "Downstream Compression & Purification CAPEX" section of model.

Equipment	Ref. Installed Cost	Scaling	Ref. Capacity	New Capacity	Cost exp.	Ref. Year	New Year	New Installed Cost
Downstream compression & purification	$87,596,398	CO_2 product	140 t/hr	114 t/hr	0.9			$99,773,569
Sorbent handling	$553,000	CO_2 product Sorbent capacity			0.6			$3,013,168
Sorbent prep., storage, and feed	$290,000	CO_2 product Sorbent capacity			0.6			$1,580,142
Cooling water system	$3,220,000	CO_2 product Sorbent capacity			0.6			$17,545,028
Spent sorbent handling system	$1,082,000	CO_2 product Sorbent capacity			0.6			$5,895,565
Instrumentation & control	$10,091,000	CO_2 product Sorbent capacity	100,000 t/yr	1,000,000 t/yr	0.6	2020	2022	$54,983,501
Site prep., improvements, facilities	$5,383	CO_2 product Sorbent capacity			0.60			$31,810
Buildings & structures (admin., machine shop, warehouse, etc.)	$1,943,000	CO_2 product Sorbent capacity			0.60			$10,586,953
DAC steam distribution system	$1,734,000	CO_2 product Sorbent capacity			0.70			$11,894,530
DAC system controls equipment	$601,000	CO_2 product Sorbent capacity			0.15			$1,161,910
						TOTAL		$206,466,177

S3.J. Total Operation & Maintenance Cost

The operation & maintenance cost $(C_{O\&M})$ includes labor (C_L), utilities (C_{UT}), transportation and storage ($C_{T\&S}$), and indirect/general expenses (eq. S48). Utility costs include thermal ($C_{thermal}$) and electrical ($C_{electrical}$) energy (eq. S49). Indirect/general expenses include direct manufacturing costs (direct supervisory and clerical labor, maintenance and repairs, operating supplies, laboratory charges, and patents and royalties), fixed manufacturing costs (local taxes and insurance, plant overhead costs), and general manufacturing expenses (administration costs, distribution and selling costs, and research and development). These are calculated as fractions of the total capital (non-annualized).

$$C_{O\&M} = C_L + C_{UT} + C_{T\&S} + C_{indirect/general} \#(S48)$$

$$C_{UT} = C_{thermal} + C_{electrical} \#(S49)$$

The electrical energy cost $(C_{electrical})$ is the total electrical energy $(E_{electrical})$, which includes blowers (E_{blower}), vacuum pumps ($E_{vacuum\ pump}$), and downstream processing ($E_{downstream}$) (eq. S51), multiplied by the cost of electricity ($p_{electricity}$) (eq. S50).

$$C_{electrical}\left[\frac{\$}{tonne\ CO_2}\right] = E_{electrical}\left[\frac{KWh}{tonne\ CO_2}\right] \cdot p_{electricity}\left[\frac{\$}{KWh}\right] \#(S50)$$

$$E_{electrical} = E_{blower} + E_{vacuum\ pump} + E_{downstream} \#(S51)$$

Similarly, the thermal energy cost $(C_{thermal})$ is the total thermal energy $(E_{thermal})$, which is the regeneration energy ($E_{regeneration}$) (eq. S53), multiplied by the cost of thermal energy ($p_{thermal}$) (eq. S52). Determining the amount of thermal and electrical energy required is described further in section S3.K.

$$C_{thermal}\left[\frac{\$}{tonne\ CO_2}\right] = E_{thermal}\left[\frac{MMBtu}{tonne\ CO_2}\right] \cdot p_{thermal}\left[\frac{\$}{MMBtu}\right] \#(S52)$$

$$E_{thermal} = E_{regeneration} \#(S53)$$

The labor (C_L) and transportation and storage costs $(C_{T\&S})$ are also included.

$$C_L\left[\frac{\$}{year}\right] = N_{operators} \cdot N_{shifts} \cdot S_{base\ labor}\left[\frac{\$}{year}\right] \#(S54)$$

$$C_{T\&S} = C_{transport} + C_{storage} \#(S55)$$

Table S15. Summary of "Energy OPEX Totals" section of model. Input variables from S1 are used with no sorbent degradation. Parameters are evaluated at a sorbent lifetime of 1.04 years.

Parameter	Input or from Eq.	Variable	Value	Unit
Electricity Cost	Input	$p_{electricity}$	0.06	$/KWh
Electrical Energy	Eq. S51	$E_{electrical}$	50.27	GJ/cycle
			0.79	GJ/t-CO_2
			220.20	KWh/t-CO_2
Blower Electrical Energy	Table S17	E_{blower}	0.800	GJ/t-CO_2
Vacuum Pump Electrical Energy	Table S18	$E_{VP,total}$	0.060	GJ/t-CO_2
Downstream Electrical Energy	Table S19		0.504	GJ/t-CO_2
Cumulative Electrical Energy			899,111	GJ/lifetime
Electrical Cost	Eq. S50	$C_{electrical}$	13.21	$/t-$CO_2$
Steam Cost	Input	$p_{thermal}$	0.03	$/kg
Thermal Energy	Eq. S53	$E_{thermal}$	3.43×10^8	kJ/lifetime
			5.41	GJ/t-CO_2
Thermal Cost	Eq. S52	$C_{thermal}$	80.30	$/t-$CO_2$
Total Utilities	Eq. S49	C_{UT}	93.51	$/t-$CO_2$

Table S16. Summary of "Non-energy OPEX Totals" section of model. Input variables from S1 are used with no sorbent degradation. Parameters are evaluated at a sorbent lifetime of 1.04 years.

Parameter	Input or from Eq.	Variable	Value	Unit
Number of Operators	Input	$N_{operators}$	8.00	$/yr
Number of Shifts	Input	N_{shifts}	8.00	$/yr
Base labor rate	Input	$S_{base\ labor}$	79,370	$/t-$CO_2$
Labor cost	Eq. S54	C_L	5,079,680	$/t-$CO_2$
Transport - Midwest (IL Basin)	Input	$C_{transport}$	2.07	$/t-$CO_2$
Storage - Midwest (IL basin)	Input	$C_{storage}$	8.32	$/t-$CO_2$
T&S	Eq. S55	$C_{T\&S}$	10.39	$/t-$CO_2$
Labor & T&S cost	Portion of Eq. S48	$C_L + C_{T\&S}$	15.47	$/t-$CO_2$

Table S17. Summary of "Indirect/General OPEX Totals" section of model. Input variables from S1 are used with no sorbent degradation. Parameters are evaluated at a sorbent lifetime of 1.04 years.

Parameter	Calculated as	Value	Unit
Maintenance	1.5% of total capital	14,679,349	$/yr
Insurance	0.5% of total capital	4,893,116	$/yr
Local taxes & fees	0.5% of total capital	4,893,116	$/yr
Indirect labor	3.5% of labor and maintenance	6,915,660	$/yr
Other OPEX cost		31.38	$/t-$CO_2$

S3.K. Energy Operations & Maintenance Cost

Electrical Energy

The electrical energy required by the process includes the blower and vacuum pump energies (eq. S51), which are determined via eqn. S56 and S567 from Sinha et al.[7],

$$E_{blower} = \Delta P Q_{FG} t_{ads} \quad \#(S56)$$

where ΔP is the pressure drop across the sorbent (Pa), Q_{FG} is the volumetric flow rate of flue gas through the sorbent during the adsorption step (m^3/s), and t_{ads} is the time of the adsorption step per cycle (s).

$$E_{VP,total} = -\frac{\eta P_1 V_1^{\gamma}\left(V_2^{1-\gamma} - V_1^{1-\gamma}\right)}{1-\gamma} \quad \#(S57)$$

η is the isentropic efficiency of the pumps, P_1 is the initial pressure (Pa), V_1 and V_2 are the initial and final volumes (m^3), respectively, and γ is the ratio of the desorption stream heat capacity at constant pressure to that at constant volume. The final volume for the vacuum pump is calculated using the initial pressure (101.325 kPa), initial volume (amount of air in module), γ value, and final pressure (vacuum pressure), following the relationship:

$$V_1 = \frac{n_{CO_2}(t_i)\,[kg]}{\rho_{CO_2}\left[\frac{kg}{m^3}\right]} \quad \#(S58)$$

The following relationship is used to find V_2:

$$PV^{\gamma} = constant \quad \#(S59)$$

Table S18. Summary of "Blower OPEX – Electrical" section of model. Input variables from S1 are used with no sorbent degradation. Parameters are evaluated at a sorbent lifetime of 1.04 years.

Parameter	Input or from Eq.	Variable	Value	Unit
Adsorption step time	Input	t_{ads}	1200	s
Pressure drop	Input	ΔP	500	Pa
Blower energy	Eq. S56	E_{blower}	785,144	kJ/t-CO$_2$
			49.79	GJ/cycle

Table S19. Summary of "Vacuum Pump OPEX – Electrical" section of model. Input variables from S1 are used with no sorbent degradation. Parameters are evaluated at a sorbent lifetime of 1.04 years.

Parameter	Input or from Eq.	Variable	Value	Unit
Isentropic Pump Efficiency	Input	η	0.70	
Contactor Porosity	Input		0.25	
P1	Input	P_1	101,325.00	Pa
V1	Eq. S58	V_1	4,938	m^3
V1^(1-gam)	Portion of Eq. S57	$V_1^{1-\gamma}$	0.43	
Vacuum pressure (P2)	Input	P_2	20,000	Pa
V2	Eq. S57	V_2	21,590	m^3
V2^(1-gamma)	Portion of Eq. S57	$V_2^{1-\gamma}$	0.37	
P1V1^gam	Portion of Eq. S59	PV^{γ}	1.17 x 10^9	
P2V2^gam	Portion of Eq. S59	PV^{γ}	1.17 x 10^9	
Gamma	Input	γ	1.10	
Vacuum Pump Energy	Eq. S57	$E_{VP,total}$	480,430	kJ/cycle

			0.48	GJ/cycle
			7,575	kJ/t-CO$_2$
			267	kJ/s or kW

Downstream compression & purification operation & maintenance costs are scaled from the Trimeric quote by simply scaling by the capacities.

Table S20. Summary of "Downstream OPEX – Electrical" section of model. Input variables from S1 are used with no sorbent degradation. Parameters are evaluated at a sorbent lifetime of 1.04 years.

Parameter	Input or from Eq.	Value	Unit
REFERENCE - annual variable O&M	Input	869,983	$/yr
REFERENCE - power requirement for CO$_2$ compression	Input	0.14	MWh/t-CO2
REFERENCE - power requirement	Input	19.32	MWh
CURRENT - power requirement		15.75	MWh
CURRENT -annual variable O&M		709,379	$/yr
REFERENCE Annual Non-PT&I Fixed O&M	Input	958,616	$/year
REFERENCE PT&I Fixed O&M	Input	1,441,247	$/year
REFERENCE and CURRENT Total O&M		3,109,242	$/year
Downstream O&M costs		3.11	$/t-CO2

The total electrical energy is the sum of the blower, vacuum pump, and downstream electrical requirements. As the sorbent degrades, the electrical energy can change. Thus, the electrical energy is fitted to a third-order polynomial (cubic) expression.

$$E_{electrical} = A \cdot t_{cycle}^3 + B \cdot t_{cycle}^2 + C \cdot t_{cycle} + D \#(S60)$$

The parameters of the quadratic (A, B, C, D) fit were continuously updated in the model using a three-parameter LINEST function in Excel. The cumulative amount of energy was determined by integrating equation S64 over the sorbent's lifetime.

$$E_{electrical} = \frac{A}{4} \cdot t_{lifetime}^4 + \frac{B}{3} \cdot t_{lifetime}^3 + \frac{C}{2} \cdot t_{lifetime}^2 + D \cdot t_{lifetime} \#(S61)$$

Thermal Energy

The energy required for desorption of CO$_2$ includes latent heat ($\Delta H_{ads} \dot{n}_{CO_2}$) and sensible heat for each of the three components (amine, silica support, polymer). Specifically, this will depend on the CO$_2$ heat of adsorption of the sorbent ΔH_{ads}, the amount of CO$_2$ adsorbed/desorbed ($\dot{n}_{CO_2}$), the amount of each component (m_1, m_2, m_3), the heat capacity of each component ($C_{P,1}$, $C_{P,2}$, $C_{P,3}$), and the temperature swing (ΔT).

$$E_{desorption,\ required} = \Delta H_{ads} \dot{n}_{CO_2} + m_1 C_{P,1} \Delta T + m_2 C_{P,2} \Delta T + m_3 C_{P,3} \Delta T \#(S62)$$

The model allows for N_2 and H_2O desorption energy contributions to be turned on or off depending on whether this should be considered. If both are turned on, the full equation becomes:

$$E_{desorption,\ required} = \Delta H_{ads}\dot{n}_{CO_2} + \Delta H_{ads,N_2}\dot{n}_{N_2} + \Delta H_{ads,H_2O}\dot{n}_{H_2O} + m_1 C_{P,1}\Delta T + m_2 C_{P,2}\Delta T + m_3 C_{P,3}\Delta T \#(S63)$$

The selectivity of CO_2 to component "i" is defined in Eq. S64, where x refers to the concentration in the adsorbed phase and y refers to the concentration in the bulk phase. The selectivities for both CO_2 to N_2 and CO_2 to H_2O are inputs, and they are used to calculate the amount of H_2O and N_2 that adsorb.

$$S = \frac{\dfrac{x_{CO_2\ (ads.)}}{x_{i\ (ads.)}}}{\dfrac{y_{CO_2\ (bulk)}}{y_{i\ (bulk)}}}\#(S64)$$

The CO_2/N_2 and CO_2/H_2O selectivities and concentrations in the bulk phase (i.e., $\dfrac{y_{CO_2\ (bulk)}}{y_{i\ (bulk)}}$) are used to calculate the ratios of concentrations in adsorbed phase (i.e., $\dfrac{x_{CO_2\ (ads.)}}{x_{i\ (ads.)}}$). The ratio of concentrations in the adsorbed phase are used to determine how much N_2 and H_2O adsorb, and therefore desorb, each cycle based on the cumulative amount of CO_2 that adsorbs. Then, this is used in combination with their respective heats of adsorption in Eq. S63 to find the energy required to desorb each of the species.

Similar to electrical energy, the amount of thermal energy changes as the sorbent degrades and amount of CO_2 captured changes. The amount of required thermal energy is fitted to a third-order polynomial of the number of cycles. Examples of the fit are provided in Figure S8.

$$E_{thermal} = A \cdot t_{cycle}^3 + B \cdot t_{cycle}^2 + C \cdot t_{cycle} + D\#(S65)$$

The cumulative amount of thermal energy is determined by integrating equation S65 over the sorbent's lifetime.

$$E_{thermal} = \frac{A}{4} \cdot t_{lifetime}^4 + \frac{B}{3} \cdot t_{lifetime}^3 + \frac{C}{2} \cdot t_{lifetime}^2 + D \cdot t_{lifetime}\#(S66)$$

Table S21. Summary of "Regeneration OPEX for sensible and CO_2 - thermal" section of model. Input variables from S1 are used with no sorbent degradation. Parameters are evaluated at a sorbent lifetime of 1.04 years.

Parameter	Input or from Eq.	Variable	Value	Unit
Temperature Swing	Input	ΔT	70.00	K
CO_2 Heat of Adsorption	Input	ΔH_{ads}	-70.00	kJ/mol
Sensible energy to heat comp. 1	Portion of Eq. S58	$m_1 C_{P,1}\Delta T$	4.98×10^7	kJ/cycle
			0.0346	MJ/mol CO_2
			12.6	MJ/m³ contactor
Sensible energy to heat comp. 2	Portion of Eq. S58	$m_2 C_{P,2}\Delta T$	4.03×10^7	kJ/cycle
			0.0280	MJ/mol CO_2
			10.2200	MJ/m³ contactor
Sensible energy to heat polymer	Portion of Eq. S58	$m_3 C_{P,3}\Delta T$	1.52×10^8	kJ/cycle
			0.1050	MJ/mol CO_2
			38.3	MJ/m³ contactor
Total sensible energy	Portion of Eq.	sum of 3	0.167	MJ/ mol CO_2

Parameter	Input or from Eq.	Variable	Value	Unit
	S58			
Latent energy to desorb CO2	Portion of Eq. S58	$\Delta H_{ads}\dot{n}_{CO_2}$	1.01×10^8	kJ/cycle
			0.0700	MJ/mol CO_2
			25.5	MJ/m^3 contactor
Sensible fraction of heat			0.71	fraction
Energy to desorb CO_2 (cycle specific)	Eq. S58	$E_{desorption}$	0.238	MJ/mol CO_2
			3.42×10^8	kJ/cycle
Cumulative energy to desorb CO_2	Eq. S65	$E_{desorption}$	5.40	GJ/t-CO_2
			5.35	GJ/lifetime

Table S22. Summary of "Regeneration OPEX for N_2 - thermal" section of model. Input variables from S1 are used with no sorbent degradation. Parameters are evaluated at a sorbent lifetime of 1.04 years.

Parameter	Input or from Eq.	Variable	Value	Unit
CO_2 adsorption	Eq. S21	$\dot{n}_{CO_2}$	1.44×10^6	mol CO_2/lifetime
Nitrogen				
CO_2/N_2 selectivity	Input	S	62,944	
y CO_2/N_2 (bulk)	From inputs	$\dfrac{y_{CO_2 \, (bulk)}}{y_{i \, (bulk)}}$	5.31×10^{-4}	
x CO_2/N_2 (ads)	Eq. S64	$\dfrac{x_{CO_2 \, (ads.)}}{x_{i \, (ads.)}}$	33.42	
N_2 ads.	Eq. S64	$\dot{n}_{N_2}$	4.31×10^4	mol-N_2/lifetime
N_2 heat of sorption	Input	$\Delta H_{ads,N_2}$	15	kJ/mol
Cumulative energy to desorb N_2	Portion of Eq. S63		10,658	GJ/lifetime
			0.01	GJ/t-CO_2
Purity				
CO_2	Eq. S21		1.44×10^6	mol CO_2/lifetime
N_2	Eq. S64		4.31×10^4	mol-N_2/lifetime
Other species from non-zero vacuum			32,730	mol/cycle
			5.39×10^8	mol/lifetime
Purity			0.95	

S3.L. Including Carbon Footprint

The electrical energy and thermal energy calculations are described in the previous section, S3.K. The calculation for electrical energy carbon footprint uses an energy source carbon footprint (Table S24-S26) and the total electrical energy from S3.K. The calculation for thermal energy carbon footprint is described at the end of this section.

Total Carbon Footprint

Table S23. Summary of "Total carbon footprint" section of model. Input variables from S1 are used with no sorbent degradation. Parameters are evaluated at a sorbent lifetime of 1.04 years.

Parameter	Value	Unit
CO_2 emissions from electricity	171.00	kg/MWh
	0.171	$t\text{-}CO_2$/MWh
Blower and vacuum pump electrical energy	8.61×10^5	$kJ/t\text{-}CO_2$
	0.24	$MWh/t\text{-}CO_2$
Compression Energy	0.14	$MWh/t\text{-}CO_2$
Total electrical energy	0.38	$MWh/t\text{-}CO_2$
Carbon footprint from electrical energy	0.06	$t\text{-}CO_2$ emitted $/t\text{-}CO_2$ captured
Carbon footprint from thermal energy (see table S27)	0.06	$t\text{-}CO_2$ emitted $/t\text{-}CO_2$ captured
Carbon footprint from total energy	0.12	$t\text{-}CO_2$ emitted $/t\text{-}CO_2$ captured
Carbon footprint from equipment materials & construction	0.01	$t\text{-}CO_2$ emitted $/t\text{-}CO_2$ captured
CO_2 emissions from sorbent	3.14	kg CO_2/kg sorbent
Carbon footprint from sorbent	0.01	$t\text{-}CO_2$ emitted $/t\text{-}CO_2$ captured
Total carbon footprint	0.13	$t\text{-}CO_2$ emitted $/t\text{-}CO_2$ captured

Electrical Carbon Footprint

Table S24. Carbon emissions per MWh of electricity for various energy sources.

Energy Source	Emissions	
	kg CO_2/MWh	*kg CO_2/GJ*
Natural Gas	506	140.6
Coal	1074	298.3
Nuclear	7	1.9
Solar Thermal	38	10.6
Solar Photo-voltaic	48	13.3
Wind	17	4.7
Hydro	17	4.7
Petro	1124	312.2
Geo-thermal	118	32.8
Biomass	1340	372.2

Table S25. Energy carbon footprints of the five states with the lowest values compared to 2020 U.S. Mix.

	Washington		Oregon		Idaho		New Hampshire		South Dakota		2020 US Mix	
	%	kg CO_2/ MWh	%	kg CO_2/ MWh	%	kg CO_2/ MWh	%	kg CO_2/ MWh	%	kg CO_2/ MWh	%	kg CO_2/ MWh
Natural Gas	14.4	72.9	33.3	168.5	26	131.6	25.6	129.5	8.7	44.0	35	177.1
Coal	2.9	31.1	0	0	0.1	1.1	1.6	17.2	9.2	98.8	26	279.2
Nuclear	7.8	0.5	0	0	0	0	56.5	4.0	0	0	20	1.4
Solar Thermal	0	0	0	1.0	0	0	0	0	0	0	0	0
Solar Photo-voltaic	0	0	2.5	7.5	3.3	1.6	0	0	0	0	2	0.96
Wind	8.7	1.5	15.7	2.7	15.7	2.7	2.9	0.5	52.3	8.9	8	1.36
Hydro	64.6	11.0	46.4	7.9	51	8.7	6.7	1.1	29.7	5.0	7	1.19
Petro	0	0	0	0	0	0	0.4	4.5	0.1	1.1	0	0
Geo-thermal	0	0	0.3	0.3	0.5	0.6	0	0	0	0	0	0
Biomass	1.6	21.4	1.7	22.8	3.3	44.2	6.2	83.0	0.1	1.3		
Total		139		211		190		157		158		461

Table S26. Comparison of energy carbon footprints.

	kg CO_2/MWh	kg CO_2/GJ
Average of the five states with the lowest carbon footprints	171	47.5
2020 U.S. Mix	461	128.1
U.S. carbon intensity of grid electricity from Young et al. (2023)[8]		99

Note on Thermal Energy Carbon Footprint & Cost

The carbon footprint for thermal energy is not as direct as electrical energy. Steam is used for regeneration of the sorbent. This steam typically comes from a power plant. The power plant has to produce more electricity to make up for the electricity it loses by sending the steam for direct air capture rather than sending it to the turbine. The steam carbon footprint is therefore the carbon footprint of the extra electricity produced.

To calculate this, we first determine the conditions of the steam used for regeneration. The enthalpy and specific entropy are based on the temperature and pressure.

We next use these conditions to determine the amount of steam we need for regeneration. The amount of available energy from the steam is the difference between the enthalpy at the inlet temperature and the enthalpy at the condensation temperature. We use a heat recovery efficiency factor (85%) to determine the energy we receive from the steam (kJ/kg steam). We know how much thermal energy is needed for regeneration (GJ/t-CO_2 captured), so we can then determine the amount of steam required per t-CO_2 captured. Multiplying this by the steam price ($0.002929)[5] provides us with the total thermal energy cost.

We then consider what would happen if we instead put that steam into a turbine, specifically: (1) what would be the outlet conditions and (2) what is the energy we would obtain (energy is removed from the steam during isentropic expansion in the turbine). The outlet conditions are based on an expansion at the same pressure, and the work obtained (kJ/kg steam) is the difference in enthalpy between the inlet and outlet steam multiplied by the efficiency of the turbine (75%).

We then calculate how much work was forgone by using the steam for thermal energy rather than electricity. This is simply the energy we could obtain (kJ/kg steam) multiplied by the amount of steam needed for regeneration (kg steam/t-CO_2 captured). This forgone work is equivalent to how much extra work the power plant must generate to make up for the steam extracted for the regeneration.

We can then use the same energy source carbon footprint from the electrical energy calculations (171 kg/MWh to determine the carbon emissions associated with the forgone work, which provides the carbon footprint of the thermal energy.

Table S27. Summary of "Total carbon footprint" section of model. Input variables from S1 are used with no sorbent degradation. Parameters are evaluated at a sorbent lifetime of 1.04 years.

Parameter	Value	Unit
Conditions of steam for regeneration		
Superheat by	10	C
P_in	1.1	bar
Saturation temp. (Tsat)	102.3	C
T_in	112.3	
H (P, T)	2699.8	kJ/kg
S (P, T)	7.4	kJ/(kg·K)
Amount of steam required for thermal energy		
T_op (temp. steam condenses at)	90	C
H (T, x = 0)	377.0	kJ/kg
delta H	2322.9	kJ/kg
Efficiency of Heat Recovery	0.85	
Energy from steam	1974.4	kJ/kg
Thermal energy	7.14	GJ/t-CO_2 captured
kg steam required	3,615	kg steam/t-CO_2 captured
Cost of steam	0.03	$/kg-steam
Total thermal energy cost	105.9	$/t-$CO_2$ captured
	14.8	$/GJ
If you instead put that steam in a turbine → Outlet steam conditions and energy of turbine assuming isentropic expansion.		
T	35	C
Psat (P_out)	0.056	bar
H (S, P)	2,266	kJ/(kg·K)
S (P, T)	7.4	kJ/(kg·K)
T_out	35	C
Efficiency of Turbine	0.75	
Energy of Expansion = work	325.8	kJ/kg steam
Forgone work by using steam for thermal energy instead of electricity		
Forgone Work	1,177,837	kJ/t-CO_2 captured
	327.2	KWh/t-CO_2 captured
Electricity carbon footprint from thermal energy		
Electricity carbon footprint	0.17	t-CO_2/MWh
	0.056	t-CO_2 emitted/t-CO_2 captured

Sorbent Carbon Footprint

Table S28. Sorbent carbon footprints compiled from literature sources. PEI on silica was used in this analysis.

Sorbent	Carbon footprint Cradle-to-gate (Cradle-to-grave) $kg\ CO_{2,eq}$ / MWh	Ref.
MIL-101	5.42	*Leonzio, Shah, et al. Sustain. Prod. Consum. (2022)*
MOF-177	49.1	
MOF-5	9.14	
APDES-NFC-FD	26.2	
SI-AEATPMS	4.12	
PEI on alumina	1.45 (1.72)	*Deutz & Bardow, Nat. Energy (2021)*
PEI on silica	2.67 (3.14)	
PEI on cellulose	4.77 (6.15)	
Carbonate on silica	1.17 (1.29)	
Carbonate on activated carbon	1.42 (1.55)	
Anionic resin	4.48 (4.86)	
Avg. for 66 polymers	6.83	
Ni MOF-74	11 - 1134	*Grande, Payet, et al. SM&T (2017)*

S4. EXAMPLE RESULTS OF DAC LCOC

Table S29. Example metrics from the economic model with and without exponential degradation. Sorbent capacity fade is 50% in 1 year (initial capacity = 1.0 mol/kg, decay constant = 4.40×10^{-5} cycle^{-1}). These values correspond to the results presented in Figure 3.

Parameter	Unit	No degradation	Exponential degradation
Outputs			
Sorbent Lifetime		1.04	1.04
kg CO_2/kg sorbent/hr		0.0440	0.0313
mol CO_2 / kg contactor/hr		0.999	0.711
Instantaneous recovery at 1 year		0.50	0.24
LCOC w/o carbon footprint	$/t-CO_2$	325	450
Capital cost	$/t-CO_2$	180	253
Operation & maintenance cost	$/t-CO_2$	144	195
Sorbent cost	$/t-CO_2$	1.80	2.53
Total carbon footprint	t-CO_2,e / t-CO_2 capt.	-0.88	-0.86
LCOC w/ carbon footprint	$/t-CO_2$	368	525
Non-annualized capital ($/t-$CO_2$/yr)		979	979
kg CO_2/yr/kg sorbent		694	494
t-CO_2/yr/m^3 contactor		253	180
tonne of PEI-silica		1,441	1,441
# contactors		20,747	20,747
# blowers		1,037	1,037
# vacuum pumps		2,075	2,075
Sensible fraction of thermal energy		0.71	0.83
Sensible + CO_2 latent heat	GJ/t-CO_2	5.40	6.81
N_2 latent heat		0.01	0.01
Thermal energy		5.42	6.83
Electrical energy		0.80	1.64
CAPEX	% of total cost	55%	56%
OPEX		44%	43%
Sorbent Cost		1%	1%
Blower Capital		9.31%	9.46%
Vacuum Pump Capital		2.75%	2.79%
Auxiliary Capital		11.96%	12.15%
Downstream Capital		11.19%	11.36%
Contactor Capital		20.07%	20.39%
Blower O&M		0.05%	0.08%
Vac Pump O&M		4.03%	6.00%
Thermal (Sensible) O&M		17.44%	18.73%
Thermal (Latent) O&M		7.28%	3.79%
Downstream O&M		0.96%	0.97%
Labor & TS O&M		4.76%	3.90%
Indirect & General O&M		9.65%	9.81%
Sorbent Raw Material		0.52%	0.53%
Replacement Cost		0.03%	0.03%

Table S30. Example metrics from the economic model with and without linear degradation. Sorbent capacity fade is 50% in 1 year (initial capacity = 1.0 mol/kg, decay constant = 3.17×10^{-5} cycle^{-1}).

Parameter	Unit	No degradation	Linear degradation
Outputs			
Sorbent Lifetime		1.04	1.04
kg CO_2/kg sorbent/hr		0.0440	0.0440
mol CO_2 / kg contactor/hr		0.999	
Instantaneous recovery at 1 year		0.50	0.24
LCOC w/o carbon footprint	$/t-CO_2$	325	426
Capital cost	$/t-CO_2$	180	243
Operation & maintenance cost	$/t-CO_2$	144	180
Sorbent cost	$/t-CO_2$	1.80	2.44
Total carbon footprint	$t-CO_2,e$ / $t-CO_2$ capt.	-0.88	-0.85
LCOC w/ carbon footprint	$/t-CO_2$	368	498
Non-annualized capital ($/t-CO_2/yr)		979	979
kg CO_2/yr/kg sorbent		694	513
$t-CO_2$/yr/m^3 contactor		253	187
tonne of PEI-silica		1,441	1,441
# contactors		20,747	20,747
# blowers		1,037	1,037
# vacuum pumps		2,075	2,075
Sensible fraction of thermal energy		0.71	0.79
Sensible + CO_2 latent heat	GJ/t-CO$_2$	5.40	5.89
N_2 latent heat		0.01	0.36
Thermal energy		5.42	6.25
Electrical energy		0.80	1.57
CAPEX	% of total cost	55%	57%
OPEX		44%	42%
Sorbent Cost		1%	1%
Blower Capital		9.31%	9.63%
Vacuum Pump Capital		2.75%	2.84%
Auxiliary Capital		11.96%	12.36%
Downstream Capital		11.19%	11.56%
Contactor Capital		20.07%	20.07%
Blower O&M		0.05%	0.05%
Vac Pump O&M		4.03%	6.43%
Thermal (Sensible) O&M		17.44%	19.48%
Thermal (Latent) O&M		7.28%	3.88%
Downstream O&M		0.96%	0.99%
Labor & TS O&M		4.76%	4.06%
Indirect & General O&M		9.65%	7.37%
Sorbent Raw Material		0.52%	0.54%
Replacement Cost		0.03%	0.03%

Table S31. Example metrics from the economic model with and without piecewise exponential degradation. Sorbent capacity fade is 60% in 1 year and 70% in 3 years (initial capacity = 1.0 mol/kg, decay constant until 1 year = 5.81 x 10^{-5} cycle^{-1}, decay constant after 1 year = 6.08 x 10^{-6} cycle^{-1}).

Parameter	Unit	No degradation	Piecewise exponential degradation
Outputs			
Sorbent Lifetime		1.04	1.04
kg CO_2/kg sorbent/hr		0.0440	0.0283
mol CO_2 / kg contactor/hr		0.999	0.644
Instantaneous recovery at 1 year		0.50	0.199
LCOC w/o carbon footprint	$/t-$CO_2$	325	477
Capital cost	$/t-$CO_2$	180	279
Operation & maintenance cost	$/t-$CO_2$	144	195
Sorbent cost	$/t-$CO_2$	1.80	2.80
Total carbon footprint	t-CO_2,e / t-CO_2 capt.	-0.88	-0.85
LCOC w/ carbon footprint	$/t-$CO_2$	368	563
Non-annualized capital ($/t-$CO_2$/yr)		979	979
kg CO_2/yr/kg sorbent		694	447
t-CO_2/yr/m^3 contactor		253	163
tonne of PEI-silica		1,441	1,441
# contactors		20,747	20,747
# blowers		1,037	1,037
# vacuum pumps		2,075	2,075
Sensible fraction of thermal energy		0.71	0.86
Sensible + CO_2 latent heat	GJ/t-CO_2	5.40	7.20
N_2 latent heat	GJ/t-CO_2	0.01	0.01
Thermal energy	GJ/t-CO_2	5.42	7.21
Electrical energy	GJ/t-CO_2	0.80	2.00
CAPEX	% of total cost	55%	59%
OPEX	% of total cost	44%	41%
Sorbent Cost	% of total cost	1%	1%
Blower Capital	% of total cost	9.31%	9.86%
Vacuum Pump Capital	% of total cost	2.75%	2.91%
Auxiliary Capital	% of total cost	11.96%	12.67%
Downstream Capital	% of total cost	11.19%	11.85%
Contactor Capital	% of total cost	20.07%	20.07%
Blower O&M	% of total cost	0.05%	0.09%
Vac Pump O&M	% of total cost	4.03%	6.89%
Thermal (Sensible) O&M	% of total cost	17.44%	19.25%
Thermal (Latent) O&M	% of total cost	7.28%	3.20%
Downstream O&M	% of total cost	0.96%	1.01%
Labor & TS O&M	% of total cost	4.76%	3.83%
Indirect & General O&M	% of total cost	9.65%	6.58%
Sorbent Raw Material	% of total cost	0.52%	0.56%
Replacement Cost	% of total cost	0.03%	0.03%

S5. COMPARISON TO OTHER ECONOMIC MODELS

Table S32. Comparison to other economic models in literature.

	Value in this analysis	Values from Young et al. (2023)	NETL 0B
INPUTS			
Sorbent Property Variables			
CO_2 capacity	0.25 – 3.0	N/A	1.2 mol/kg
CO_2 heat of adsorption	-70 kJ/mol	-70 kJ/mol	N/A
Heat capacity ratio	1.1	1.4	N/A
Bed porosity	0.5	0.4	N/A
Heat capacity	3000 J kg^{-1} K^{-1}	1580 J kg^{-1} K^{-1}	N/A
Sorbent contactor density	730 kg m^{-3}	880 kg m^{-3}	384 kg m^{-3}
Bed diameter	1 m	N/A	18 m
Process Variables			
Purity	95 mol. %	99 vol.%	N/A
Feed temperature	303.15 K	288.15 K	
Desorption temperature	373.15 K	373.15 K	
Vacuum pressure	20,000 Pa	14,788 Pa	
Bed pressure drop	500 Pa		625 Pa
Adsorption time	1800 s	9671 s	10,800 s
Desorption time	1200 s	6673 s	1,080 s
Superficial velocity	4 m/s		2.5 m/s
Plant Scale	1,000,000	1,000,000	1,000,000
Capital Cost Variables			
Capital recovery factor (CRF)	0.163	0.10	0.0707
Utilization	0.90	0.90	0.85
Operation & Maintenance Cost Variables			
Base Labor Rate (Operator Salary)	$79,370 / year	$64,900 / year	$38.50 / hour
No. of operators	8	N/A	8
Thermal energy requirement	5.4 GJ/t-CO_2	9.8 GJ/t-CO_2	4.3 GJ/t-CO_2
Electrical energy requirement	0.79 GJ/t-CO_2	0.99 GJ/t-CO_2	35 MW$_e$
Overall Costs			
Levelized capital cost	$ 128 – 444/t-CO_2	$ 94 – 786/t-CO_2	$189/t-$CO_2$
Levelized fixed O&M cost	$ 163 – 260/t-CO_2	$ 64 – 142/t-CO_2	$64/t-$CO_2$
Levelized variable O&M cost		$ 169 - 386/t-CO_2	$40/t-$CO_2$

*For this analysis, low end of range = 4.0 mmol/g sorbent capacity, 10 min. cycle time, high end of range = 0.5 mmol/g sorbent capacity, 120 min. cycle time.

S6. DETAILED EXPERIMENTAL METHODS

This section describes the experimental methods used to prepare, characterize, and test the unmodified and modified PEI-silica sorbents described in Figure 5. All chemicals for the syntheses described below were purchased from Sigma–Aldrich and used directly without further purification. UHP He, ultra-zero grade air, research grade CO_2, 400 ppm CO_2/He were obtained from Airgas.

S6.1 Synthesis Schemes and Fabrication Methods

Scheme 1: Synthesis of methyl modified PEI with different degrees of alkylation

This reaction was carried out with a modified version of a reported literature procedure.[9] 5 gram (6.25 mmol) of PEI was dissolved in 15 mL of dry MeOH and treated with 4.2 g (29.6 mmol), 8.23 g (58.1 mmol), and 12.34 g (85.7 mmol) 1-iodomethane at room temperature for 0.25M-PEI, 0.50M-PEI, and 0.75M-PEI, respectively. The resulting mixture was heated at 65°C for two h, 900 mg of NaOH was added after cooling. Heat was applied for an additional 12 h period, followed by removing the solvent and unreacted methyl iodide by evaporation under reduced pressure. The residue obtained appeared as yellow oily material.
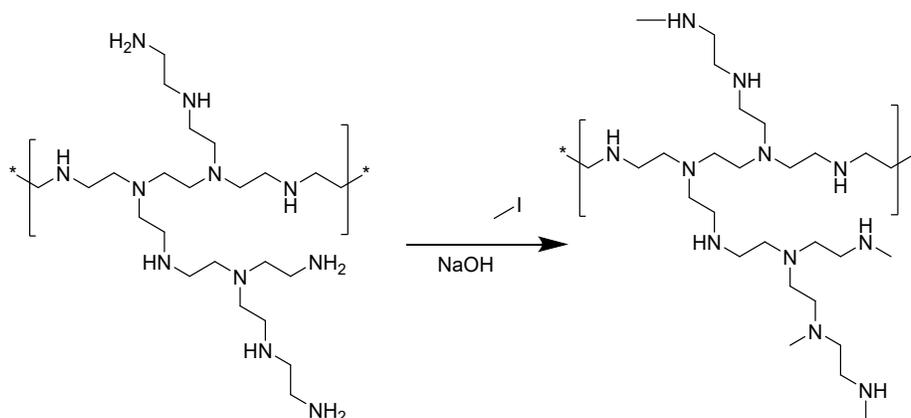
Scheme S2: Synthesis of propyl modified PEI with different degrees of alkylation

5 g (6.25 mmol) of PEI was dissolved in 15 mL of dry MeOH and treated with 4.94 g (29.6 mmol), 9.87 g (58.1 mmol) and 14.81 g (85.7 mmol) 1-iodopropane at room temperature for 0.25M-PEI, 0.50M-PEI, and 0.75M-PEI, respectively. The resulting mixture was heated at 90°C for 2 h, 900 mg of NaOH was added after cooling. Heat was applied for an additional 12 h period, followed by removing the solvent and unreacted propyl iodide by evaporation under reduced pressure. The residue obtained appeared as yellow oily material.
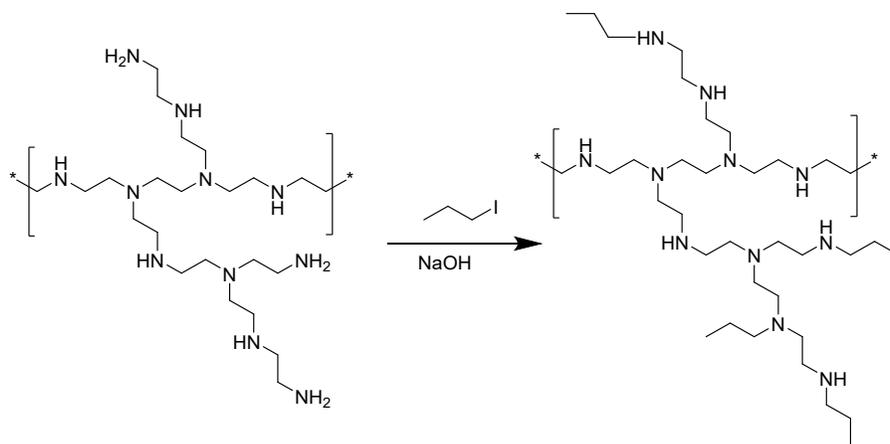
Scheme S3: Synthesis of 2-hydroxypropyl functionalized PEI with different degrees of alkylation

5 g (6.25 mmol) of PEI was dissolved in 15 mL of dry MeOH and treated with 1.68 g (29.6 mmol), 3.37 g (58.1 mmol), and 5.04 g (85.7 mmol), 1,2 epoxypropane at room temperature for 0.25HP-PEI, 0.50HP-PEI, and 0.75HP-PEI, respectively. The resulting mixture was stirred for 12 h in room temperature, followed by removing the solvent under reduced pressure. The residue obtained appeared as a colorless viscous oil.
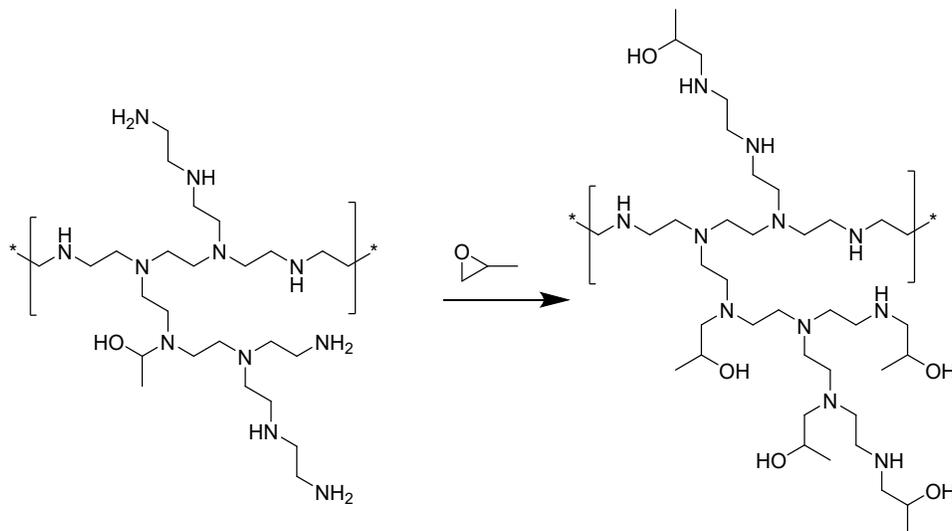
Scheme S4: Synthesis of SBA-15

SBA-15 was synthesized according to a previously published procedure.[10] 24.0 g of Pluronic P-123 block copolymer was dissolved in 600 g of deionized water and 120 mL of 12.1 (M) HCl. The components were stirred vigorously for 3 h until everything was dissolved. 46.6 g of tetraethyl orthosilicate (TEOS) was added dropwise to the mixture and stirred at 40 °C for 20 h, during which time a white precipitate formed. The solution was then heated to 100 °C and held at this temperature for 24 h without stirring. The reaction was quenched with 400 mL DI water and the precipitate was filtered and copiously washed with DI water. The filtered precipitate was dried for 12 h in an oven at 75 °C and then calcined according to the following program: heat to 200 °C at 1.2 °C min^{-1}, hold at 200 °C 1 h, heat to 550 °C at 1.2 °C min^{-1}, hold at 550 °C for 12 h, cool to room temperature. The resulting white powder was stored in ambient lab conditions.

Unmodified/modified PEI-SBA15 Composite Preparation

SBA-15 was impregnated with modified PEIs by wet impregnation. First, SBA-15 was dried overnight under vacuum (<20 mTorr) at 110 °C. The desired amount of PEI was dissolved in 15 mL methanol and added to the desired amount of SBA-15. The mixture was allowed to stir at room temperature for at least 6 h. Methanol was removed by rotary evaporation at room temperature. The resulting powder was dried overnight under vacuum (<20 mTorr) at room temperature. The resulting dried powder composites were stored in ambient lab conditions.

S6.2 Characterization

Quantitative ^{13}C NMR spectroscopy
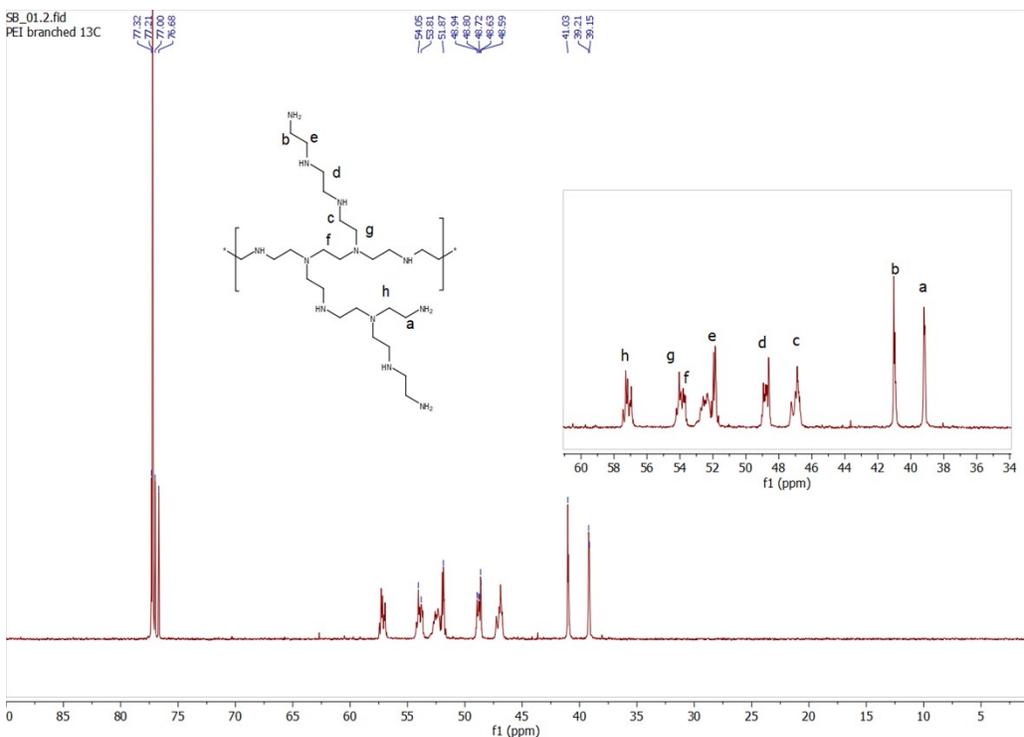
Figure S10. ^{13}C NMR spectrum of PEI in CDCl$_3$.

Inversely gated ^{13}C solution NMR experiments were conducted on a Bruker 700 and samples were prepared with CDCl$_3$ or CD$_3$OD (depending on solubility) and a single drop of chromium acetylacetonate, Cr(acac)$_2$, in deuterated DMSO to aid relaxation and minimize interference from possible base salts. Approximately 8000-10000 scans with an inversely gated pulse sequence were gathered with a relaxation time of 4 seconds. This relaxation time is more than the 3.3 seconds required for quantitative analysis determined in a complete T1 study by Geckle for branched PEI.[11]

For PEI, the distribution was calculated as primary (1°)/secondary (2°)/tertiary (3°) = $(A_a + A_b):(A_c + A_d + A_e)/2:(A_f + A_g + A_h)/3$. For R-E-PEIs, the distribution was calculated as primary (1°):secondary (2°):tertiary (3°) = $(A_a + A_b):(A_c + A_d + A_e + A_1)/2:(A_f + A_g + A_h + A_{1'})/3$. In the equations, A_i is the integrated peak area for i carbon species.[12] The ratio is calculated to be 36:38:26

N$_2$ Physisorption:

N$_2$ physisorption experiments at 77 K were conducted with a Micromeritics Tristar system. About 100-150 mg of the powder sample was activated at 110 °C under vacuum for 12 h before the measurement. the BET surface area was estimated using the N$_2$ physisorption data in the P/P_0 range of 0.05–0.2.

Table S33. N_2 physisorption data for SBA-15, unmodified PEI/SBA-15, and modified PEI/SBA-15.

Sample	Organic Loading (wt.%)	BET Surface Area (m²/g SiO₂)	Pore Volume (cm³/g SiO₂)	Pore Filling (%)
SBA-15		700	0.96	
Unmodified PEI/SBA-15	20	380	0.6	38
Unmodified PEI/SBA-15	40	93	0.28	71
Unmodified PEI/SBA-15	55	5	.016	98
0.25M-PEI/SBA-15	20	377	0.60	38
0.25M-PEI/SBA-15	40	150	0.27	72
0.25M-PEI/SBA-15	55	10	0.02	95
0.25P-PEI/SBA-15	20	384	0.60	35
0.25P-PEI/SBA-15	40	100	0.14	80
0.25P-PEI/SBA-15	55	7	0.01	95
0.25HP-PEI/SBA-15	20	389	0.7	32
0.25HP-PEI/SBA-15	40	170	0.41	55
0.25HP-PEI/SBA-15	55	14	.09	90

*modified PEI sorbents are denoted as n-x-PEI in methanol into SBA-15, where "x" denotes the functional group such as methyl (M), propyl (P) and 2-hydroxypropyl (HP) and "n" denotes the molar ratio of the functional reagent (e.g., 1,2-epoxypropane) to total moles of nitrogen in PEI.

Description of N_2 Physisorption, Pore Volume and Pore Filling Characterization

The pore network of SBA-15 consists of a majority of mesopores of a pore width equal to 7 nm and 0.96 cm³/g, as shown in BJH pore size distribution. As expected, there is a clear reduction in the pore volume and pore size in the micro and mesoporous regime of the pore size distribution of SBA-15 with increasing modified PEI loading (Fig 2 and Supplementary Table 8). In the case of 0.25HP-PEI loaded samples, at 20% loading, 32% of the pores are filled, and as the loading increases to 40%, pore filling increases to 70%; with 55% loading, the pore filling increased to 95%. The pore filling behavior is consistent with all the modified versions and unmodified PEI (800 MW), which is observed by plotting the pore filling behavior of modified PEIs and unmodified PEIs with respect to weight percent loading in SBA15. As several studies point out, the adsorption capacity of PEI polymers reaches a maximum at around 40% organic loading in SBA-15 framework[13] and decreases upon high organic loading since it can hinder the diffusion of guest molecules in the pore network; all further materials in this work have been fixed at 40% organic loading in SBA-15.

Gravimetric CO_2 adsorption and heat of adsorption

Pseudo-equilibrium CO_2 adsorption capacities were measured gravimetrically on a TA Instruments Q500 TGA. 10-15 mg of samples were pretreated by heating to 110 °C at a ramp rate of 10 °C min⁻¹ under a flow of He and held for 2 h. The samples were cooled to 30 °C and equilibrated at this analysis temperature for 1 h. Subsequently, the gas flow was switched to a premixed gas containing 400 ppm CO_2/He with 90 ml/min. The mass gain was recorded and normalized by the dry mass of the sample. Cyclic adsorption/desorption experiments were performed in a similar manner, with sorbent regeneration in between each adsorption cycle. The heat of CO_2 adsorptions were recorded using NETZSCH TGA-DSC at 30 °C under 400 ppm CO_2/He (90 ml min⁻¹) Before the measurements, the adsorbent samples were degassed at 110 °C for 1 h under He flow (90 ml min⁻¹). The heat of adsorption was calculated by normalizing the total heat flow by the amount of gas adsorbed, recorded in triplicate, and averaged.

Table S34: Amine distributions in modified PEIs.

Sample	Titration	1º Amine mol.%	2º Amine mol.%	3º Amine mol.%
PEI	0	36	38	27
0.25M-PEI	25	22	48	30
0.50M-PEI	50	13	49	38
0.75M-PEI	75	8	20	72
0.25P-PEI	25	20	45	35
0.50P-PEI	50	9	53	38
0.75P-PEI	75	2	25	73
0.05HP-PEI	5	32	38	30
0.10HP-PEI	10	30	45	40
0.15HP-PEI	15	24	42	34
0.25HP-PEI	25	22	44	34
0.50HP-PEI	50	10	50	40
0.75HP-PEI	75	5	55	40

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