Supplementary Materials

Defining targets for adsorbent material performance to enable viable BECCS processes

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Table S1. Supplemental information used for the calculation of LCOC in Figures 2-6.1-5

| Parameter                                      | Value         | Unit         |
|------------------------------------------------|---------------|--------------|
| **Flue Gas**                                  |               |              |
| Moisture in fuel                              | 0.101         | wt. %        |
| Ash in fuel                                   | 0.014         | wt. %        |
| Absolute humidity of air                      | 0.1           | kg H₂O/kg air|
| Excess air coefficient - λ                    | 1.2           |              |
| Stoichiometric oxygen volume                  | 0.8565        | m³N O₂/kg fuel|
| Stoichiometric volume of dry air              | 4.0786        | m³N dry air/kg fuel|
| Stoichiometric volume of wet air              | 4.7353        | m³N wet air/kg fuel|
| Air flow rate into boiler                     | 510,616       | m³N air/h    |
| Carbon dioxide volume                         | 0.8223        | m³N CO₂/kg fuel|
| Water vapor volume                            | 1.2804        | m³N H₂O/kg fuel|
| Sulfur dioxide volume                         | 0.00042       | m³N SO₂/kg fuel|
| Nitrogen volume                               | 3.2264        | m³N N₂/kg fuel|
| Stoichiometric flue gas volume                | 5.3295        | m³N flue gas/kg fuel|
| Real flue gas volume                          | 6.2765        | m³N flue gas/kg fuel|
| Oxygen volume                                 | 0.947         | m³N O₂/kg fuel|
| Flue gas density                              | 1.0           | kg/m³        |
| **Boiler/Turbine**                            |               |              |
| Power generated                               | 100*          | MW           |
| Mass of biomass                               | 5000          | tonne/day    |
| Combustion efficiency                         | 0.80          |              |
| Deaerator pressure                            | 861.8         | kPa          |
| Boiler steam temperature                      | 727.6         | K            |
| Boiler steam pressure                         | 6205          | kPa          |
| Blowdown rate                                 | 0.03          |              |
| Turbine isentropic efficiency                 | 0.70          |              |
| Generator efficiency                          | 0.95          |              |
| Mass flow of steam                            | 228           | kg/s         |
| Fuel energy required                          | 0.925         | GJ/s         |
| Biomass (willow) LHV                          | 17.6          | MJ/kg        |
| Water loss fraction                           | 0.10          |              |
| **Adsorption**                                |               |              |
| Cycle time                                    | 300           | s            |
| Adsorption step time                          | 200           | s            |
| Contactor to sorbent mass ratio               | 1.0           |              |
| Sorbent bulk density                          | 1300          | kg/m³        |
| Contactor bulk density                        | 1300          | kg/m³        |
| Contactor porosity                            | 0.25          |              |
| Sorbent cost per mass                         | 40            | $/kg sorbent |
| Sorbent cost per contactor external surface area | 5.43         | $/m²         |
| Contactor system cost per external surface area | 10           | $/m²         |
| Contactor system installation factor          | 2.50          |              |
| CO₂ product purity                            | 0.95          |              |
| N₂ heat of desorption                         | 15            | kJ/mol       |
| H₂O heat of vaporization                     | 40.8          | kJ/mol       |
| CO₂:N₂ selectivity                            | 19            |              |
| CO₂:H₂O selectivity                           | 1             |              |

*varies slightly with sorbent parameters
Table S1 continued. Supplemental information used for the calculation of LCOC in Figures 2-6.6-9

| Parameter                                                                 | Value   | Unit   |
|---------------------------------------------------------------------------|---------|--------|
| **Heat Exchanger**                                                       |         |        |
| Steam temp. post-turbine (690 kPa)                                        | 520     | K      |
| Saturation temp. of steam (690 kPa)                                      | 438     | K      |
| Hot water temp. post-heat exchanger, pre-boiler (101.3 kPa)              | 388     | K      |
| ∆H\text{vap} of steam (690 kPa)                                          | 2068    | kJ/kg  |
| C_p of steam (400 K – 500 K)                                             | 2.00    | kJ/kg*K   |
| C_p of water (32 K – 443 K)                                              | 4.20    | kJ/kg*K   |
| Steam temp. pre-desorption (198 kPa)                                     | 393     | K      |
| Water temp. post-desorption (101.3 kPa)                                  | 323     | K      |
| Saturation temp. of steam (198 kPa)                                      | 393     | K      |
| ∆H\text{vap} of steam (198 kPa)                                          | 2202    | kJ/kg  |
| Fiber temp. pre-desorption                                               | 310     | K      |
| Fiber temp. post-desorption                                              | 393     | K      |
| Sorbent heat capacity                                                    | 0.80    | kJ/kg*K   |
| Polymer heat capacity                                                    | 1.60    | kJ/kg*K   |
| **Economics**                                                            |         |        |
| Capital recovery factor (CRF)                                            | 0.12    |        |
| Plant utilization                                                        | 0.90    |        |
| Indirect capital cost                                                    | 0.35    |        |
| Contingency                                                              | 0.30    |        |
| Number of operators per shift                                            | 10      |        |
| Number of shifts                                                         | 5       |        |
| Base labor rate                                                          | 79,370  | $/yr   |
| Selling price of excess electricity                                      | 0.05    | $/kWh  |
| Biomass purchase price                                                   | 50      | $/short ton |
| Lime purchase price (2007)                                               | 180.87  | $/ton  |
| Boiler water purchase cost (388 K)                                       | 0.00245 | $/kg   |
| Low pressure steam cost                                                  | 0.027   | $/kg   |
| Ash disposal cost (non-hazardous)                                        | 28.86   | $/tonne |
| SCR O&M cost                                                             | 70      | $/MW   |
| Isentropic pump efficiency                                               | 0.70    |        |
| Initial pressure of vacuum pump                                          | 101.3   | kPa    |
| Final pressure of vacuum pump                                            | 50      | kPa    |
| Heat capacity ratio (vacuum pump)                                        | 1.30    |        |
| Lime:SO\text{2} stoichiometric ratio                                     | 1.2     |        |
Table S2. Supplemental itemized capital costs used in the calculation of LCOC in Figures 2-6.10,11

| Equipment            | Ref. Installed Cost | Scaling | Ref. Capacity | New Capacity | Cost exp. | Ref. Year | New Year | New Installed Cost |
|----------------------|---------------------|---------|---------------|--------------|-----------|-----------|-----------|-------------------|
| Feed handling        | $24,155,013         | Biomass | 104 t/hr      | 210 t/hr     | 0.6       | 2007      | 2019      | $42,590,278        |
| Boiler               | $48,652,589         | Steam   | 235 t/hr      | 1040 t/hr    | 0.6       |           |           | $137,171,228       |
| Turbogenerator       | $16,145,591         | Power   | 41.3 MW       | 100 MW       | 0.6       | 2007      | 2019      | $31,724,059        |
| Water softener       | $133,894            | Steam   | 235 t/hr      | 1040 t/hr    | 0.6       |           |           | $377,501           |
| Deaerator            | $872,600            | Steam   | 235 t/hr      | 1040 t/hr    | 0.6       | 2007      | 2019      | $2,460,211         |
| Amine injection      | $68,842             | Steam   | 235 t/hr      | 1040 t/hr    | 0.6       |           |           | $194,093           |
| Heat exchanger       | $83,863             | Steam   | 393 t/hr      | 1040 t/hr    | 0.6       |           |           | $173,551           |
| Cooling tower        | $2,186,673          | Steam   | 235 t/hr      | 1040 t/hr    | 0.6       | 2007      | 2019      | $647,676.84        |
| Blower               | $162,336            | Flue gas| 833 t/hr      | 1541 t/hr    | 0.6       |           |           | $272,138.03        |
| Vacuum pump          | $29,553,222         | CO₂     | 134 t/hr      | 340 t/hr     | 0.6       |           |           | $56,170,971        |
| SCR Unit             | $500/MW             | Power   | 350 MW        | 100 MW       | ----      | 1999      |           | $77,765            |
| **TOTAL**            |                     |         |               |              |           |           |           | **$271,859,472**   |

Figure S1. Example of function of deficit steam as number of cycles with quadratic approximation used in LCOC calculations. The parameters for this example are an initial working capacity of 0.5 mol/kg, decay constant of $4.86 \times 10^{-6}$ cycle$^{-1}$, and heat of adsorption of -40 kJ/mol.
Figure S2. Dataset from GALE used to estimate the correlation between the heat of adsorption and affinity.12

Example S1: Example LCOC calculation for adsorbent with constant capacity (no degradation).

The LCOC is a function of capital costs, sorbent system cost, operation & maintenance costs, revenue, and the cumulative amount of CO₂ captured (equation 1).

S1.A. CAPITAL COSTS

The total capital cost is the sum of direct capital, indirect capital, and contingency.

\[ C_{\text{total\ capital}} = \text{direct capital} + \text{indirect capital} + \text{contingency} \]  

(D1)

Direct capital

The direct capital includes the purchase and installation costs of the equipment and is scaled by the capital recovery factor (CRF) and utilization (equation S2). The CRF is a function of interest rate (i) and the number of annual payments (N) (equation S3). A CRF of 0.12 was chosen for this study. To illustrate the effect of CRF on capital cost, if a capital cost of $100,000 was to be paid back in 10 annual payments, with a CRF of 0.12, then the annual payment would be $12,000. The utilization is a ratio of operating capacity to design capacity (equation S4). A utilization of 0.90 was used. Multiplying the total capital by the CRF transforms the total capital ($/plant lifetime) into annual payments ($/year). Dividing the total capital by the utilization scales the total capital from an operating capacity to a design capacity.

\[ C_{\text{levelized\ capital}} = C_{\text{total\ capital}} \cdot \frac{\text{CRF}}{\text{Utilization}} \]  

(S2)

\[ \text{CRF} = \frac{i(1 + i)^N}{(1 + i)^N - 1} \]  

(S3)

\[ \text{Utilization} = \frac{\text{Operating\ capacity}}{\text{Design\ capacity}} \]  

(S4)

In the BECCS design, the direct capital includes the purchase and installation cost of the feed handling unit, boiler, boiler accessories, selective catalytic reduction (SCR) unit, flue gas desulfurization (FGD) unit and baghouse, turbogenerator, heat exchanger, cooling tower system, blower, and vacuum pump. The capital cost of each piece of equipment was estimated using quotes from the 2011 NREL biomass-to-ethanol process design. A scaling law (equation S5) was used to scale the quotes (non-
linearly due to the cost exponent, n) from the NREL project capacity and year to the equipment capacity and year of the BECCS project.$^{8,10}$

$$CC_{a,2} = CC_{b,1} \left( \frac{A_a}{A_b} \right)^n \left( \frac{I_2}{I_1} \right)$$  \hspace{1cm} (S5)

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 the BECCS project, and “b” and “1” refer to the NREL biomass-to-ethanol design. Table S2 shows the scaling variables and results.

**Indirect capital and contingency**

Indirect capital covers the transportation of equipment, insurance, purchase taxes, construction overhead, contractor engineering expenses, etc. Contingency includes any unforeseen expenses. Indirect capital and contingency were set as 35% and 30% of the direct capital, respectively.

$$C_{total \_ capital} = direct \_ capital + 0.30 \cdot (direct \_ capital) + 0.35 \cdot (direct \_ capital)$$  \hspace{1cm} (S6)

**Capital cost calculation example**

In this design, the estimated direct capital is $271,859,500, resulting in a total capital of $448,568,100 and levelized capital of $59,809,000/year. Regardless of the working capacity, the process is designed to capture the same amount of CO$_2$ per year (2,678,800 t-CO$_2$). Therefore, the levelized capital cost (without the sorbent system cost) is $22.30/t-CO$_2$ for all working capacities.

**S1.B. SORBENT SYSTEM COST**

**Amount of sorbent**

The adsorption/desorption unit was designed to contain enough sorbent to theoretically capture all of the CO$_2$ in the flue gas stream at the sorbent's initial capacity. Even though the actual recovery is defined as 90%, the amount of sorbent is still designed to recover 100% of the CO$_2$. Therefore, the amount of sorbent is based on the amount of CO$_2$ exiting the boiler and the initial sorbent capacity (equation S7). The working capacity is either set as a parameter or determined from the heat of adsorption for results in Figure 2. The cycle time is set at 300 seconds (288 cycles/day). Using the combustion stoichiometry described in the manuscript, the elemental composition of willow in Table 1 was converted to volumetric quantities of each component per kg of willow. For example, based on the boiler properties provided in Table S1, 0.8223 m$^3$ CO$_2$/kg of willow are emitted. The NO$_x$ emissions were determined using experimental results based on the boiler temperature and excess air ratio. NO$_x$ emissions decrease with decreasing excess air ratio and increasing boiler temperature.$^{13}$ Given that 5,000 tonnes/day of willow are used, it was calculated that the boiler emits 8,154,600 kg/day of CO$_2$ in the flue gas, or 25,500 kg/cycle. For an initial working capacity of 2.0 mol/kg, 289,500 kg of sorbent is required.

$$m_S = \frac{N_{CO_2, \ _ideal} \cdot \frac{mol}{day}}{C_{initial} \frac{mol}{kg}} \cdot \frac{day}{\# \ cycles} \hspace{1cm} (S7)$$

**Determining working capacity from the heat of adsorption (results in Figure 2)**

The dataset extracted from GALE for affinity and heat of adsorption values is shown in Figure S2. By plotting the natural logarithm of CO$_2$ affinity ($b_{CO_2}$) versus heat of adsorption ($\Delta H_{ads}$), equation S8 was obtained. The correlation has a coefficient of determination ($R^2$) of 0.74, indicating that the linear equation does not predict all of the variance and should be used as an approximation.

$$\Delta H_{ads} = 3.0899 \cdot \ln(b_{CO_2}) + 30.491 \hspace{1cm} (S8)$$
The affinity constant was calculated for various heats of adsorption using equation S8. The dataset was simulated at a temperature of 298 K. The affinity constant was scaled to adsorption and desorption temperatures using equation S9. The effective uptakes for adsorption and desorption were found using the Langmuir model in equation S10, where \( P_{\text{ads}} \) is 0.13 bar, and \( P_{\text{des}} \) is 1 bar. The saturation uptake is a constant, chosen value. The effective working capacity is the effective desorption uptake subtracted from the effective adsorption uptake (equation S11).

\[
\ln \left( \frac{b_{\text{ads or des}}}{b_{298}} \right) = -\frac{\Delta H_{\text{ads, CO}_2}}{R} \left( \frac{1}{T_{\text{ads or des}}} - \frac{1}{T_{298}} \right)
\]

\[
q_{\text{ads or des}} = q_{\text{sat}} \cdot b_{\text{ads or des}} \cdot P_{\text{ads or des}} \left( 1 + b_{\text{ads or des}} \cdot P_{\text{ads or des}} \right)
\]

\[
\Delta q = q_\theta = q_{\text{ads}} - q_{\text{des}}
\]

**Sorbent system cost**

The cost of the adsorption unit includes the costs of sorbent and contactors. A relationship from Sinha and Realf was used:

\[
C_{\text{ads, unit}} = \frac{(BM_S + BM_C \cdot f_{C}) \cdot A_{\text{external}} \cdot t_{\text{cycle}} \cdot \frac{1}{Utilization}}{N_{\text{cycle}} t_{\text{life}}}
\]

where \( BM_S \) and \( BM_C \) are the sorbent and contactor system bare module cost, respectively, per m² of external contactor surface area, \( rc:s \) is the mass ratio of contactor to sorbent, \( A_{\text{external}} \) is the external contactor surface area (m²), \( f_{C} \) is the contactor installation factor, \( t_{\text{cycle}} \) is the cycle time (s/cycle), \( N_{\text{cycle}} \) is the tonnes of CO₂ captured in one cycle (t-CO₂/cycle), and \( t_{\text{life}} \) is the sorbent lifetime (s). The utilization factor (0.90) is also included in this equation, as the design capacity will differ from the operation capacity.

A sorbent cost of $40/kg ($5.43/m²) and contactor cost of $10/m² was used. To estimate the necessary contactor surface area, a basis of hollow fiber sorbents was used, which results in equations S13 and S14.

\[
V_{c&s} = \frac{(m_S + m_C)}{(\rho_s + \rho_c)} \left( 1 - \phi_{\text{contactor}} \right)
\]

\[
A_{\text{external}} = \frac{4 \cdot V_{c&s} \cdot d_{\text{fiber}}^2}{(d_{\text{fiber}}^2 - d_{\text{bore}}^2)}
\]

\( V_{c&s} \) refers to the volume of the contactor and sorbent, which is the volume of fibers in the scenario of hollow fiber sorbents. \( m_S \) is the mass of necessary sorbent calculated in equation S7, and \( m_C \) is equivalent to \( m_S \) in this analysis because the contactor to sorbent mass ratio is kept constant at 1.0. \( \rho_s \) and \( \rho_c \) are the sorbent and contactor bulk density, respectively, and \( \phi_{\text{contactor}} \) is the porosity of the contactor (assumed to be 0.25). The chosen properties are mid-range values for adsorbents and polymers. The volume was converted to external surface area using the inner and outer diameters of the fibers in equation S14. \( d_{\text{fiber}} \) is the outer diameter of the fiber (assumed to be 1.2 mm), and \( d_{\text{bore}} \) is the diameter of the bore (assumed to be 0.32 mm). The total sorbent system cost is scaled by an installation factor of 2.5.

It should be noted that with some sorbent/contactor combinations, such as amine-infused polymer/silica hollow fibers, the sorbent could be regenerated without simultaneously replacing the contactor (via re-infusing amines into the fibers). In that case, the sorbent and contactor lifetimes would be different. To maintain simplicity in this analysis, it is assumed the sorbent and contactor have the same lifetime, with the limiting lifetime being that of the sorbent.

**Sorbent system cost calculation example**

For a sorbent with a constant working capacity of 2.0 mol/kg, the sorbent and contactor cost is $64,859,000. In one year, 2,678,800 t-CO₂ is captured, so a sorbent with a one-year lifetime costs $26.90/t-
CO\textsubscript{2}. The sorbent cost will increase as the capacity decreases. For example, a sorbent with a one-year lifetime and 1.0 mol/kg working capacity will cost $53.80/t-CO\textsubscript{2}.

\section*{S1.C. OPERATION AND MAINTENANCE}

The operation & maintenance cost is the sum of energy (\(C_E\)) and non-energy (\(C_{NE}\)) costs (equation S15), where the non-energy costs include the labor (\(C_L\)), utilities (\(C_{UT}\)), waste treatment (\(C_{WT}\)), and raw material (\(C_{RM}\)), equation S16. Each cost is calculated per year.

\begin{align*}
C_{O&M} &= C_{NE} + C_E \\
C_{NE} &= C_L + C_{UT} + C_{WT} + C_{RM}
\end{align*} \\
(S15) \hspace{1cm} (S16)

\textbf{Non-energy: Labor}

A labor amount of 10 operators per shift and 5 shifts was used. The estimated 2019 U.S. annual mean wage for power plant operators was $79,370/year, as reported by the U.S. Bureau of Labor Statistics.\textsuperscript{9} Therefore, the total labor cost is $3,968,500/year.

\textbf{Non-energy: Utilities}

Utilities include boiler water and deficit steam for desorption. The purchase cost of 115°C boiler water is $0.00245/kg, and the low-pressure steam cost is $0.027/kg.\textsuperscript{8} To determine how much boiler water the process requires, boiler and turbogenerator calculations were performed and based on 100 MW power production. The Steam System Modeler Tool (SSMT) from the U.S. DOE was used for the calculations.\textsuperscript{2,3} The boiler is based on a boiler in an NREL design that produces 6205.3 kPa / 454°C steam.\textsuperscript{10} The isentropic efficiency was set as 70\%, generator efficiency as 95\%, and outlet pressure of steam as 689.5 kPa.

The DOE calculator first finds the specific enthalpy of inlet steam (6205.3 kPa, 454°C, set by boiler type). The ideal specific enthalpy of outlet steam (689.5 kPa) is found by equating the ideal inlet and outlet entropies and then using the ideal outlet entropy to find the ideal outlet enthalpy. The actual outlet enthalpy is found via equation S17, where \(\eta_T\) is the isentropic efficiency (70\%) and \(\dot{h}\) is the enthalpy.

\begin{equation}
\dot{h}_{\text{outlet}} = \dot{h}_{\text{inlet}} - \eta_T(\dot{h}_{\text{inlet}} - \dot{h}_{\text{outlet,ideal}})
\end{equation} \\
(S17)

Using the outlet enthalpy, the temperature of the outlet steam is determined to be 246.7°C. Given a 95\% generator efficiency, 100 MW power, and the inlet and outlet enthalpy, the steam mass flow is 288 kg/s.

The amount of water was increased by 10\% in O&M costs to account for water loss in the boiler system. If primary steam is condensed due to less steam being required for desorption, the condensed steam is cycled back as boiler feedwater, lowering the amount of required water. Thus, the cost of water in utilities ranges from $2,800/hr for a 0.25 mol/kg working capacity sorbent to $1,600/hr for a 2.25 mol/kg working capacity sorbent, both with a heat of adsorption of -40 kJ/mol.

The inlet steam requirement for the turbogenerator was used to calculate the fuel energy required with the DOE boiler calculator. First, the steam enthalpy and energy were determined (6205.3 kPa, 454°C, 228 kg/s). Given the steam mass flow and blowdown rate (3\%), the feedwater mass flow, blowdown mass flow, blowdown energy, and boiler energy are determined using equations S18 – S21. The specific enthalpy of the blowdown is determined using a deaerator pressure of 861.8 kPa.

\begin{align*}
\dot{m}_{\text{feedwater}} &= \frac{\dot{m}_{\text{steam}}}{1 - x_{\text{blowdown}}} \\
\dot{m}_{\text{blowdown}} &= \dot{m}_{\text{feedwater}} \cdot x_{\text{blowdown}} \\
\dot{E}_{\text{blowdown}} &= \dot{h}_{\text{blowdown}} \cdot \dot{m}_{\text{blowdown}} \\
\dot{E}_{\text{boiler}} &= \dot{E}_{\text{steam}} + \dot{E}_{\text{blowdown}} - \dot{E}_{\text{feedwater}}
\end{align*} \\
(S18) \hspace{1cm} (S19) \hspace{1cm} (S20) \hspace{1cm} (S21)

Given an 80\% combustion efficiency, the required fuel energy is 0.925 GJ/s. An average willow LHV of 17.6 MJ/kg (dry basis) was used to determine the mass flow rate of biomass necessary, 58.4 kg/s.\textsuperscript{19} The LHV of the material will significantly affect how much power a given amount of material can produce via combustion. A biomass cost of $0.06/kg ($50/short ton) was used.\textsuperscript{7}
Low-pressure (689.5 kPa) steam is removed from the turbogenerator. This will decrease the power of the BECCS plant compared to completely condensing the steam in the turbine. If all of the steam were condensed, this process would produce 223 MW. The low-pressure steam is used to make secondary steam for the desorption step. An energy balance was used to determine the amount of steam available for desorption. Steam exits the turbogenerator at 520 K / 689.5 kPa. In the heat exchanger, it provides sensible energy as it cools from 520 K to its saturation temperature (438 K), latent energy at 438 K, and then sensible energy as it cools from 438 K to 388 K (the temperature of the water re-entering the boiler). The hot water exiting the desorption step is 323 K / 101.3 kPa. It requires sensible energy to heat to its saturation temperature (393 K) and latent energy at 393 K (the saturated steam temperature needed for desorption). The heat capacity and heat of vaporization values are shown in Table S1. In conclusion, the uncontaminated steam exiting the turbogenerator produces a maximum of 281 kg/s of secondary steam (198 kPa).

To validate the boiler and turbine methodology, the results presented here are compared to similar analyses. In an NREL biomass-to-ethanol design report, 234,784 kg/hr of steam (454°C, 6100 kPa) enters the turbine. Of that steam, 12% is extracted at 1200 kPa, 35% is extracted at 860 kPa, and 53% is condensed at -90 kPa. This method predicts an electricity generation of 37.5 MW, similar to the 41 MW they predict.

Jin et al. designed a steam-Rankine system. The boiler produces steam at 16,000 kPa and 550°C, and all of the steam is condensed in the boiler. In contrast, the boiler in this analysis produces steam at 6,200 kPa and 454°C, and the steam is removed at low pressure from the turbine to be used for desorption in the CCS system. If steam of that pressure and temperature were used in this system, and it were all condensed in the boiler, the electricity generation would increase from 100 MW to 262 MW. A steam reheat step is included in the Jin et al. process, and there is no steam reheat step in this analysis. The steam reheat step would likely increase the power from the 262 MW predicted by this model to the 296 MW they predict. The similarity in results further validates this portion of the model.

To desorb CO₂, the steam must provide enough energy to (1) heat the sorbent, (2) heat the contactor, (3) provide the heat of CO₂ desorption of the sorbent, (4) provide the heat of N₂ desorption, and (5) provide the heat of vaporization of H₂O. It should be noted that if steam stripping is used, the water will not be desorbed every cycle. A water energy penalty is added here to cover desorption processes that desorb water every cycle. The influence of CO₂/N₂ and CO₂/H₂ selectivities is included in the required regeneration energy of the adsorbent. The values used in the energy calculation are in Table S1. The required energy calculation is in equation S17, where the subscripts “A” and “C” refer to adsorbent and contactor, respectively, and “x” refers to the adsorbed amount. Using the required energy found via equation S22, the necessary mass flow rate of steam was determined, assuming that the steam condenses and cools 70°C, using equation S23. Comparing the necessary mass flow rate of steam for desorption to the amount of secondary steam produced either reveals excess steam or a steam deficit. If less secondary steam is required than the maximum amount produced, less primary steam is removed from the turbogenerator. The portion of the primary steam not removed is condensed to generate additional electricity. If there is a steam deficit, the cost is added to the utilities cost in non-energy operation and maintenance.

\[
E_{\text{desorption, required}} = \Delta H_{\text{ads}} \hat{n}_{CO_2} + m_A C_{PA} \Delta T_A + m_C C_{PC} \Delta T_C + \Delta H_{\text{vap, H}_2O} \frac{\hat{n}_{CO_2}}{x_{CO_2}/x_{H_2O}} + \Delta H_{\text{ads, N}_2} \frac{\hat{n}_{CO_2}}{x_{CO_2}/x_{N_2}} \quad (S22)
\]

\[
E_{\text{desorption, provided}} = \hat{m}_{\text{steam}} \Delta H_{\text{vap}} + \hat{m}_{\text{steam}} C_{P, \text{steam}} \Delta T_{\text{steam}} \quad (S23)
\]

The ratio of adsorbed CO₂ to adsorbed N₂ (X_{CO₂/X_{N₂}}) or H₂O (X_{CO₂/X_{H₂O}}) is determined via Eq. S24, where j refers to either N₂ or H₂O, “S” refers to selectivity, and “y” refers to the amount in the bulk phase.

\[
x_{CO_2}/x_j = S_{CO_2/j} \cdot y_{CO_2}/y_j \quad (S24)
\]

In the LCOC results provided in the manuscript, the CO₂ working capacity is a chosen, fixed value, and therefore the selectivities have no influence on that value. The results would be the same for...
non-competitive adsorption, in which the selectivities do not affect the CO$_2$ capacity. However, to analyze the effect of selectivity on LCOC in competitive adsorption scenarios, the influence of the selectivities on the CO$_2$ working capacity was added in section S4, and the results of that analysis are provided in Table S4.

**Non-energy: Waste treatment**

Waste treatment includes treating the ash. The biomass fuel is 1.5% ash, with a flow rate of 5,050,700 kg/day. Therefore, there is 3.16 t-ash/hr of ash. The non-hazardous waste disposal is estimated at $36/t-waste,$ so the total waste treatment cost is $995,500/yr.

**Non-energy: Raw materials**

The raw material component of operation & maintenance is biomass, lime for the FGD unit, and catalyst for the SCR unit. The biomass feed is 58.4 kg/s, and a cost of $0.06/kg ($50/short ton) was assumed, so the biomass cost is $11,600/hr. The necessary lime amount is based on a 1:1 stoichiometric ratio of lime to SO$_2$, with 20% excess. The flue gas contains 250 kg SO$_2$/hr (3,940 mol SO$_2$/hr), so 4,730 mol lime/hr (0.40 t-lime/hr) is required. A lime cost of $209.13/t-lime was used, scaled from a 2007 cost given in the NREL report using the CEPCI values.$^{10}$ SCR operating costs were scaled from a 1999 EPA cost estimate, and a cost of $70/MW was used.$^{11}$ The total raw material cost is $102,321,700/yr.

**Energy**

The energy component of the operation and maintenance cost would consist of the blower for adsorption and a vacuum pump for desorption. However, enough electricity is generated via the turbogenerator to cover the energy used by the process. Hence, the required energy is not factored into operation and maintenance costs but instead subtracted from the electricity to be sold to the grid in the revenue calculation. The energy required by the process is a sum of the blower and vacuum pump energies (equation S25), and the energies for the blowers and vacuum pumps were found via equations S26-27, respectively,$^{14}$

$$E_{process} = E_B + E_V$$

$$E_B = \Delta P Q_{FG} t_{ads}$$

$$E_V = -\frac{\eta P_1 V_1^\gamma (V_2^{1-\gamma} - V_1^{1-\gamma})}{1-\gamma}$$

where $\Delta P$ 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). In equation S27, $\eta$ 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 $\gamma$ is the ratio of the desorption stream heat capacity at constant pressure to that at constant volume. A $\gamma$ value of 1.3 was used. The final volume for the vacuum pump is calculated using the initial pressure (101.325 kPa), initial volume (amount of CO$_2$, N$_2$, and H$_2$O desorbed), $\gamma$ value, and final pressure (vacuum pressure, 50 kPa), following the relationship:

$$PV^\gamma = constant$$

**O&M cost example calculation**

Since the energy requirement is covered by the electricity produced by the plant, the O&M cost is only non-energy costs. The only component of O&M cost per year that is not constant with sorbent working capacity is utilities. For a sorbent with a working capacity of 2.0 mol/kg, heat of adsorption of -40 kJ/mol, and one-year lifetime ($\$2,678,800$ t-CO$_2$ captured), the O&M cost is $45.70/t-CO$_2$.

**S1.D. REVENUE**

The revenue comes from excess electricity sold (equation S29). The excess electricity, $E_{excess}$, is the difference between the total electricity produced and the electricity used in the process. The amount of
primary steam removed from the turbogenerator varies with the amount of secondary steam required for desorption. The purchase price of electricity is $0.05/KWh.\(^7\)

\[
Revenue = E_{\text{excess}} \cdot C_{\text{electricity}}
\]  
(S29)

**Revenue example calculation**

For a sorbent with a working capacity of 2.0 mol/kg and heat of adsorption of -40 kJ/mol, an additional 395,503 kg/hr of primary steam is condensed, providing an additional 5.8 MW of power. For the same sorbent, 6.9 MW of power is required for the process. Therefore, 98.9 MW can be sold to the grid. Revenue is constant with sorbent lifetime. The revenue for the described sorbent is $4,945/hr or $16.20/t-CO\(_2\).

**LCOC example calculation**

In the previous sections, the capital cost, sorbent cost, O&M cost, and revenue were determined to be $22.30/t-CO\(_2\), $26.90/t-CO\(_2\), $45.70/t-CO\(_2\), and $16.20/t-CO\(_2\), respectively. Therefore, a sorbent with a working capacity of 2.0 mol/kg, heat of adsorption of -40 kJ/mol, sorbent cost of $40/kg, lifetime of one year, and cycle time of 300 seconds will have a levelized cost of CO\(_2\) capture of $78.80.

**Example S2: Example calculation for LCOC with effects of sorbent degradation.**

The levelized costs of CO\(_2\) removal for scenarios with sorbent degradation were also calculated using equation 1. However, each component of the equation was adjusted from an annual basis to a lifetime basis to account for the amount of CO\(_2\) captured changing per cycle. It should be noted that the following adjusted method could also be used to calculate the LCOC for a sorbent with no degradation, corresponding to a decay constant of 0 in equations 3 and 4.

**S2.A. CUMULATIVE AMOUNT OF CO\(_2\) CAPTURED**

The working capacity in equation 3 was integrated over the sorbent’s lifetime to find the cumulative amount of CO\(_2\) captured in the en-masse replacement method.

\[
N_{CO_2,ufe} = \int_0^t (q_{\delta,0}e^{-k\cdot t\text{cycle}})m_S d(t_{\text{cycle}}) = m_S \frac{q_{\delta,0}}{k} (1 - e^{-k\cdot t_{ufe}})
\]  
(S30)

\(t_{\text{life}}\) is the number of cycles in the sorbent lifetime. The resulting cumulative amount of CO\(_2\) captured is used for determining the LCOC rather than an annualized amount.

Similarly, the working capacity in equation 4 was integrated over the sorbent’s lifetime to find the cumulative amount of CO\(_2\) captured in the partial replacement method.

\[
N_{CO_2,ufe} = q_{\delta,0}m_S \left( \frac{t_{ufe}m_r}{m_r + m_s} + \frac{m_s}{m_r + m_s} \left( 1 - e^{-(m_r + m_s)\cdot t_{ufe}} \right) \right) + \frac{m_s m_r}{m_r + m_s} \left( 1 - e^{-(m_r + m_s)\cdot t_{ufe}} \right)
\]  
(S31)

**S2.B. CAPITAL COST**

For degrading sorbents, levelized capital was calculated using an adjusted equation S2, shown in equation S32. The total capital, $448,568,100, is the same, and it is scaled by a capital recovery factor (0.12) and utilization (0.90) ratio. Since the amount of CO\(_2\) captured changes across years, the capital cost and amount of captured CO\(_2\) are 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.

\[
C_{\text{levelized capital}} = \left( C_{\text{total capital}} \cdot \frac{\text{CRF}}{\text{Utilization}} \right) \left[ \frac{\$}{\text{yr}} \right] \cdot t_{ufe} \left[ \frac{\text{yr}}{\text{lifetime}} \right] \cdot \frac{1}{N_{CO_2} \left[ \text{tonne CO}_2 \right]}
\]  
(S32)

**Capital cost calculation example**

The total direct capital is $271,859,500. For a sorbent with a decay constant of 6.6 \(\times\) 10\(^{-6}\) mol/kg, en-masse replacement method, and initial working capacity of 2.0 mol/kg, the cumulative amount of CO\(_2\)
captured in one year is 1,933,800 t-CO₂ (equation S30). Therefore, the levelized capital cost is $30.90/t-CO₂ for the sorbent with a lifetime of one year. The levelized capital cost will change with sorbent lifetime and decay constant since the cumulative amount of CO₂ captured changes. The levelized capital cost will not change with sorbent working capacity because the cumulative amount of captured CO₂ does not vary with working capacity. As long as the decay constant is the same, the recovery will be the same and decrease at the same rate.

S2.C. SORBENT COST

The sorbent cost was calculated using an equation similar to S12 (equation S33). The equation is modified to account for the amount of CO₂ captured changing between cycles. Whereas equation S12 uses a factor of \((t_{cycle}/N_{cycle} \times \text{time})\) to account for the cumulative amount of captured CO₂, the cumulative amount of CO₂ captured found in equations S30 and S31 was used. The sorbent cost is a one-time purchase, so the cost per lifetime will be the same regardless of the lifetime length.

\[
C_{\text{sorbent}} = (B M S + B M C \times r_{C-S}) m_s \left[ \frac{\$}{\text{lifetime}} \right] \frac{1}{N_{CO_2}} \left[ \frac{\text{lifetime}}{\text{tonne CO}_2} \right] \cdot \frac{1}{\text{Utilization}}
\]  
(S33)

Sorbent cost calculation example
Continuing the example of a sorbent with an initial working capacity of 2.0 mol/kg, decay constant of 6.6 x 10^6 cycle^-1, and one-year lifetime (1,933,800 t-CO₂ captured), the sorbent cost is $37.30/t-CO₂.

S2.D. OPERATION & MAINTENANCE COSTS

The operation & maintenance cost will again only consist of the non-energy costs, as the energy required by the process is supplied by the energy produced via the turbogenerator.

Non-energy: raw materials, waste, labor, and utilities without steam

The utilities (not including the cost of steam), raw material, waste, and labor costs were calculated on an annual basis, following the previous method. Utilities (not including steam), raw material, labor, and waste are $9,258,900/yr, $102,322,400/yr, $3,968,500/yr, and $943,300/yr, respectively. The costs are then scaled to the sorbent’s lifetime since they are constant across the sorbent’s lifetime.

Non-energy: steam

The amount of deficit steam (depending on working capacity and heat of adsorption) changes as the amount of CO₂ captured changes. For time frames between 1-5 years, the amount of deficit steam was approximated as a quadratic function of the number of cycles:

\[
N_{\text{steam,cycle}} = A \cdot t_{cycle}^2 + B \cdot t_{cycle} + C
\]  
(S34)

The parameters of the quadratic (A, B, and C) fit were continuously updated in the model using a three-parameter LINEST function in Excel. The \(R^2\) value of the quadratic fit was over 0.998 for all variations of sorbent and process parameter combinations. Figure S1 shows an example of a case with deficit steam. The cumulative amount of deficit steam was determined by integrating equation S34 over the sorbent’s lifetime.

\[
N_{\text{steam,lifetime}} = \frac{A}{3} \cdot t_{\text{lifetime}}^3 + \frac{B}{2} \cdot t_{\text{lifetime}}^2 + C \cdot t_{\text{lifetime}}
\]  
(S35)

The cumulative cost \($/\text{lifetime}\) was added to O&M (equation S36). If there is excess steam, more primary steam is condensed, which increases the electricity in revenue.

\[
C_{\text{O&M}} = \left( C_{\text{O&M,minus steam}} \right) \left[ \frac{\$}{\text{yr}} \right] \cdot \frac{t_{\text{lifetime}}}{\text{yr}} + \left( C_{\text{deficit steam}} \right) \left[ \frac{\$}{\text{lifetime}} \right] \cdot \frac{1}{N_{CO_2}} \left[ \frac{\text{lifetime}}{\text{tonne CO}_2} \right]
\]  
(S36)

For times over five years, a steady-state recovery is reached. If the process continues without sorbent replacement in the en-masse replacement method, the steady-state recovery will be 0. In the partial
replacement method, the steady-state recovery will depend on the decay constant and replacement rate. In both scenarios, a linear function was used to model the cycles before a steady state is reached, and then the cumulative steam after that point was found by multiplying the amount per cycle by the number of cycles.

**O&M calculation example**

For a sorbent with an initial working capacity of 2.0 mol/kg, heat of adsorption of -40 kJ/mol, decay constant of $6.6 \times 10^6$ cycle$^{-1}$, en-masse replacement method, and one-year lifetime (1,993,800 t-CO$_2$ captured), the O&M cost is $59.90/t-CO_2$. If the decay constant is decreased to $2.7 \times 10^6$, the O&M cost decreases to $50.00/t-CO_2$ because more CO$_2$ is captured due to less degradation.

**S2.E. REVENUE**

Revenue from the process is a function of excess electricity that can be sold, which changes as the sorbent degrades and less CO$_2$ is captured. When there is less CO$_2$ captured, the vacuum pumps have to pull less CO$_2$ from the sorbent bed after desorption, thus decreasing their operating energy and increasing the amount of excess electricity from the turbogenerator that is sold. Less CO$_2$ captured also means that less steam is needed to desorb the CO$_2$, thus increasing the amount of primary steam that can be condensed to generate additional electricity.

Similar to the method used above for deficit steam, the excess electricity is approximated using a quadratic function (equation S37). For all variations of sorbent/process parameters, the fit of the electricity quadratic function was above 0.998. The cumulative electricity is found by integrating equation S31 over the sorbent’s lifetime, where $t_{\text{cycle}}$ is the cycle number, and the result of the integral is used in equation S38.

$$N_{\text{electricity}} = A' \cdot t_{\text{cycle}}^2 + B' \cdot t_{\text{cycle}} + C' \quad (S37)$$

The cumulative amount of excess electricity was multiplied by the purchase price to find revenue in units of $/lifetime (equation S38).

$$C_{\text{revenue}} = \left( \int_0^{N_{\text{cycle}}} \frac{N_{\text{electricity}}}{t_{\text{cycle}}} \, dt \right) \cdot C_{\text{electricity}} \left( \frac{\$}{\text{lifetime}} \right) \cdot \frac{1}{N_{\text{CO}_2}} \left( \frac{\text{lifetime}}{\text{tonne CO}_2} \right) \quad (S38)$$

**Revenue calculation example**

For a sorbent with an initial working capacity of 2.0 mol/kg, heat of adsorption of -40 kJ/mol, decay constant of $6.6 \times 10^6$ cycle$^{-1}$, and one-year lifetime, the additional electricity is 9.9 MW, with 12.8 MW required for the process. Therefore, for an electricity purchase price of $0.05/kWh and cumulative amount of 1,933,800 t-CO$_2$ captured, the revenue is $22.00/t-CO_2$.

**LCOC calculation example**

In the previous sections, the capital cost, sorbent cost, O&M cost, and revenue were determined to be $30.90/t-CO$_2$, $37.30/t-CO$_2$, $59.90/t-CO$_2$, and $22.00/t-CO$_2$, respectively. Therefore, a sorbent with a working capacity of 2.0 mol/kg, heat of adsorption of -40 kJ/mol, sorbent cost of $40/kg, one-year lifetime, decay constant of $6.6 \times 10^6$ cycle$^{-1}$, en-masse replacement method, and cycle time of 300 seconds will have a levelized cost of CO$_2$ capture of $106.00/t-CO_2$. 

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S3. Effect of biomass and electricity prices on LCOC.

The chosen biomass purchase price and electricity selling price will affect the LCOC results in this analysis. The LCOC for a non-degrading sorbent (2.0 mmol/g sorbent capacity, 0.5-year sorbent lifetime, -40 kJ/mol heat of adsorption) is shown below in Table S3 as a function of biomass purchase and electricity selling price.

Table S3. BECCS levelized cost of CO2 removal as a function of biomass purchase and electricity selling prices.

| Biomass Price ($/short ton) | Electricity Selling Price ($/kWh) |
|-----------------------------|----------------------------------|
|                             | 0.01    | 0.03    | 0.05    | 0.07    | 0.10    |
| Waste resources             |         |         |         |         |         |
| 10                          | $61     | $55     | $48     | $42     | $32     |
| 30                          | $77     | $70     | $64     | $57     | $47     |
| Used in this analysis       |         |         |         |         |         |
| 50                          | $92     | $85     | $79     | $72     | $63     |
| Sustainable resources       |         |         |         |         |         |
| 80                          | $115    | $108    | $102    | $95     | $85     |
| 100                         | $130    | $123    | $117    | $110    | $101    |

S4. Varying CO2/N2 and CO2/H2O selectivities.

In competitive adsorption, varying the selectivity of different species will affect the CO2 working capacity. For simplicity of analysis, the adsorption of all species is assumed to follow the Langmuir model, Eq. S39, where i is either CO2, H2O, or N2.

\[
\theta_i = \frac{b_i \cdot p_i}{1 + b_i \cdot p_i} \quad (S39)
\]

The sum of the fractional occupancy of the species must be 1, Eq. S40.

\[
\theta_{CO_2} + \theta_{H_2O} + \theta_{N_2} = 1 \quad (S40)
\]

The CO2/N2 and CO2/H2O selectivity of the adsorbent from Eq. S24 will determine the ratios of fractional occupancies, Eq. S41.

\[
\theta_{CO_2} \cdot \frac{x_{H_2O}}{x_{CO_2}} + \theta_{N_2} \cdot \frac{x_{H_2O}}{x_{CO_2}} = 1 \quad (S41)
\]

Using Eq. S41, the effective fractional occupancy of CO2 can be determined (Eq. S42), and then subsequently the effective CO2 working capacity (Eq. S43).

\[
\theta_{CO_2, \text{effective}} = \frac{1}{1 + \frac{x_{H_2O}}{x_{CO_2}} + \frac{x_{N_2}}{x_{CO_2}}} \quad (S42)
\]

\[
q_{CO_2, \text{ads effective}} = \theta_{CO_2, \text{effective}} \cdot q_{CO_2, \text{ads sat}} \quad (S43)
\]

The saturation capacity will still be a chosen, fixed value, but the effective CO2 fractional occupancy, and therefore the effective CO2 adsorption capacity, will vary with selectivity. If all of the species are assumed to be removed during desorption, the effective CO2 adsorption capacity is equivalent to the effective CO2 working capacity.
Table S4. Effects of CO$_2$/N$_2$ and CO$_2$/H$_2$O selectivity in competitive and non-competitive adsorption scenarios. The LCOC results are for an adsorbent with a 2.0 mmol/g CO$_2$ saturation capacity, -40 kJ/mol heat of adsorbent, and one-year lifetime.

### Competitive Adsorption
*(selectivity affects regeneration energy and CO$_2$ capacity)*

| CO$_2$/N$_2$ Selectivity | 500 (Ex: Amines) | 50 (Ex: Zeolites) | 5 (Ex: Silica) |
|--------------------------|------------------|------------------|---------------|
| 10                       | $81              | $83              | $105          |
| 1.0                      | $125             | $127             | $168          |
| 0.1                      | $962             | $966             | $1,006        |

### Non-competitive Adsorption
*(selectivity only affects regeneration energy)*

| CO$_2$/H$_2$O Selectivity | 500 (Ex: Amines) | 50 (Ex: Zeolites) | 5 (Ex: Silica) |
|---------------------------|------------------|------------------|---------------|
| 10                        | $76              | $76              | $76           |
| 1.0                       | $77              | $77              | $78           |
| 0.1                       | $137             | $137             | $140          |

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