

## Supplementary Information

### **Electrochemical Direct Air Capture with Intermittent Renewable Energy: Techno-economic Insights from Solar-Driven Electrodialysis Systems**

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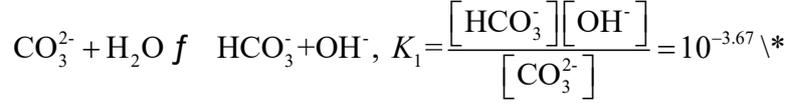
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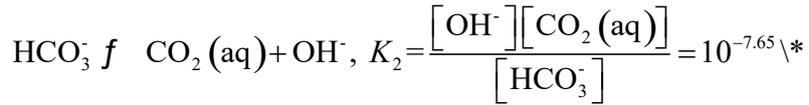
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## 1. Solution Regeneration

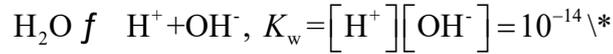
The carbonate/bicarbonate reactions and corresponding equilibrium constants in the regenerative solution are expressed as<sup>1</sup>:



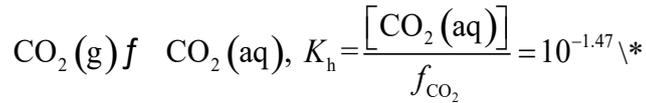
MERGEFORMAT (S1)



MERGEFORMAT (S2)



MERGEFORMAT (S3)



MERGEFORMAT (S4)

where  $f_{\text{CO}_2}$  is the fugacity of  $\text{CO}_2$ . Assuming the degassing is performed without vacuum and under the condition of atmospheric pressure and 400 ppm  $\text{CO}_2$  concentration, it's assumed as 0.0004 atm for the degassed solution equilibrium calculation. The constants are used to calculate the ion concentrations under equilibrium conditions.

Figure S1 illustrates the detailed changes in pH and DIC throughout the solution regeneration cycle. As the solution consists of carbonate/bicarbonate, the pH remains alkaline throughout the loop, even after electrochemical acidification. Acidification and basification occur within the BPMED cells, which operate as a closed system with no DIC change. DIC variations occur during the absorption and degassing processes, while the concentration of metal ions ( $\text{K}^+$ ) remains constant. The governing equation for the cycle can be expressed as:

$$\text{DIC}_2 = \text{DIC}_3 \quad \text{\textbackslash* MERGEFORMAT (S5)}$$

$$DIC_1 = DIC_4 \quad \backslash * \text{MERGEFORMAT (S6)}$$

$$[K^+]_1 = [K^+]_2 \quad \backslash * \text{MERGEFORMAT (S7)}$$

$$[K^+]_3 = [K^+]_4 \quad \backslash * \text{MERGEFORMAT (S8)}$$

The ion concentrations of each solution state are primarily determined using equilibrium constants, with particular consideration given to the limitations of CO<sub>2</sub> absorption. Based on estimates of the CO<sub>2</sub> absorption performance of Carbon Engineering's air contactor,<sup>2</sup> the outflow solution exhibits a notable DIC gap compared to the equilibrium (ideal) conditions with the inlet air. The extent of CO<sub>2</sub> absorption can be quantified using the absorption ratio, defined as:

$$R_a = \frac{DIC_2}{DIC_{2,eq}} \quad \backslash * \text{MERGEFORMAT (S9)}$$

where  $DIC_2$  represents the actual DIC after CO<sub>2</sub> absorption (black solid lines), while  $DIC_{2,eq}$  is the ideal DIC (red dashed lines) under equilibrium conditions. Under actual operating conditions, the absorption ratio is assumed to be 0.9, which is considered an attainable estimate.<sup>2</sup>

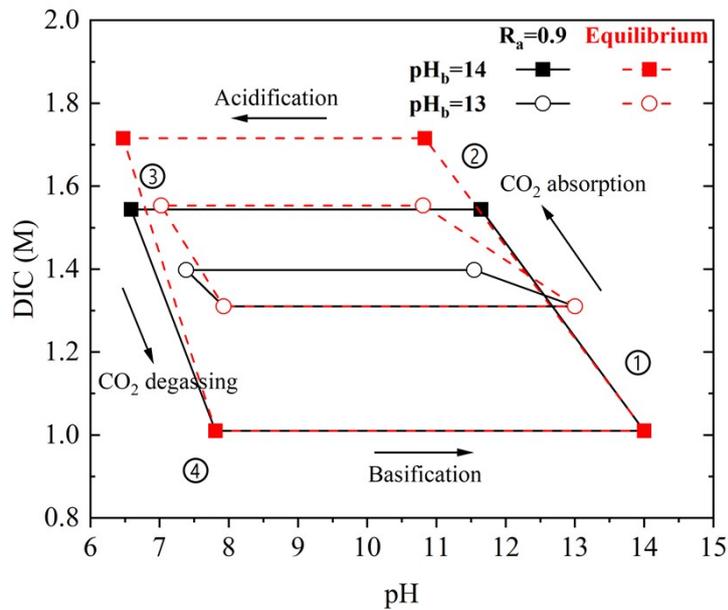


Figure S1. DIC-pH diagram of carbonate/bicarbonate solution regenerative cycle.

The cycles with basified pH values of 13 and 14 are also compared in Figure S1. The

DIC change reflects the DAC capacity of the liquid absorbent. At a basified pH of 13, the DIC change is only 0.1 M, less than half of the equilibrium value, indicating lower absorption efficiency. In contrast, at a basified pH of 14, the actual DIC change reaches approximately 0.5 M, exceeding 70% of the equilibrium value. A higher basified pH and greater pH-swing amplitude enhance both the potential and kinetics of chemical absorption, as well as improve degassing performance. Additionally, a basified pH of 14 closely aligns with the absorbent conditions used in Carbon Engineering's commercial plant, which employs a 1 M KOH solution. This alignment ensures that subsequent cost estimations for the air contactor are more reliable. Therefore, the higher basified pH of 14 has been adopted in this study.

## 2. Mathematical Models of System Components

### 2.1 BPMED Cell

The single cell voltage of the BPMED incorporating HER and OER can be expressed as:

$$E_{\text{cell}} = E_{\text{ws}}^0 + E_{\text{a}} + E_{\text{c}} + E_{\text{BPM}} + E_{\text{ohm}} \quad \backslash*$$

MERGEFORMAT (S10)

where  $E_{\text{ws}}^0$  is the standard thermodynamic potential for the water electrolysis reaction, set at 1.229 V under standard temperature conditions.  $E_{\text{a}}$  and  $E_{\text{c}}$  represent the activation overpotentials at the anode and cathode, respectively, and are determined using the Butler-Volmer equation <sup>3</sup>:

$$E_{\text{a}} = \frac{RT}{F} \ln \left( \frac{i}{2i_{0,\text{a}}} \right) \quad \backslash* \text{MERGEFORMAT (S11)}$$

$$E_{\text{c}} = \frac{RT}{F} \ln \left( \frac{i}{2i_{0,\text{c}}} \right) \quad \backslash* \text{MERGEFORMAT (S12)}$$

where  $i_{0,\text{a}}$  and  $i_{0,\text{c}}$  are the anode and cathode exchange current densities, respectively, set to 0.0048 and 0.19 mA cm<sup>-2</sup> based on reported values.<sup>3</sup>

The potential of the BPM,  $E_{\text{BPM}}$ , is primarily determined by the pH difference across the membrane and can be expressed as<sup>4</sup>:

$$E_{\text{BPM}} = n_{\text{BPM}} (0.52 + 0.059 \Delta \text{pH}) \quad \text{MERGEFORMAT} \quad (\text{S13})$$

where the term 0.52V accounts for the reaction overpotential associated with the BPM's water dissociation process.<sup>5</sup>

The ohmic loss  $E_{\text{ohm}}$  is determined by the resistance of the solution electrolyte and membranes. It can be expressed as:

$$E_{\text{ohm}} = i \cdot [R_{\text{e}} + n_{\text{BPM}} R_{\text{BPM}} + (n_{\text{BPM}} + 1) R_{\text{CEM}}] \quad \text{MERGEFORMAT} \quad (\text{S14})$$

where  $R_{\text{BPM}}$  and  $R_{\text{CEM}}$  are the electric resistances of a single BPM and CEM, respectively. In this study, they are set as 10  $\Omega \text{ cm}^2$  and 4  $\Omega \text{ cm}^2$ , respectively.<sup>4</sup>  $n_{\text{BPM}}$  is the number of repetitive BPM units in a cell. The resistance of the solution electrolytes  $R_{\text{e}}$ , considering the effect of gas volumetric fraction  $\varphi$ , can be expressed as<sup>6</sup>:

$$\sigma_{\text{mp}} = \sigma \frac{8(2-\varphi)(1-\varphi)}{(4+\varphi)(1-\varphi)} \quad \text{MERGEFORMAT} \quad (\text{S15})$$

$$R_{\text{e}} = \frac{L}{\sigma_{\text{mp}}} \quad \text{MERGEFORMAT} \quad (\text{S16})$$

where  $\sigma$  and  $\sigma_{\text{mp}}$  are the conductivity of the pure liquid and multi-phase electrolytes.  $L$  is the thickness of the electrolyte compartment.

For the BPMED cell with  $\text{K}_3/\text{K}_4\text{Fe}(\text{CN})_6$  redox couple, the single cell voltage is expressed as:

$$E_{\text{cell}} = E_{\text{a}} + E_{\text{c}} + E_{\text{BPM}} + E_{\text{ohm}} \quad \text{MERGEFORMAT} \quad (\text{S17})$$

The calculation of each potential follows the methods described above. The electrode exchange current density for the  $\text{K}_3/\text{K}_4\text{Fe}(\text{CN})_6$  redox couple set to 229  $\text{mA cm}^{-2}$ .<sup>7</sup>

The power consumption of the entire BPMED stack is calculated as the sum of the power consumption of single cells:

$$P_{\text{BPMED}} = n_{\text{cell}} i A_{\text{cell}} E_{\text{cell}} \quad \backslash * \text{MERGEFORMAT (S18)}$$

where  $n_{\text{cell}}$  is the total number of single cells in the stack, and  $A_{\text{cell}}$  is the cell effective (single membrane) area.

## 2.2 Typical-Day Solar Irradiance and PV Power Output

The solar irradiance model is developed to predict the varying PV power conditions over a typical day. This model captures the temporal variation of solar radiation throughout the day, enabling precise simulation of PV power output. Based on Baig et al.'s modified Gaussian distribution function model,<sup>8</sup> the solar radiation intensity is described as follows:

$$r_t = \frac{1}{2\sigma\sqrt{2\pi}} \left[ \exp\left(\frac{-(t-12)^2}{2\sigma^2}\right) + \cos\left(\frac{180^\circ(t-12)}{S_0-1}\right) \right] \quad \backslash *$$

MERGEFORMAT (S19)

$$\sigma = \frac{1}{r_{t=12}\sqrt{2\pi}} \quad \backslash * \text{MERGEFORMAT (S20)}$$

$$S_0 = \frac{2}{15} \arccos(-\tan \varphi \cdot \tan \delta) \quad \backslash * \text{MERGEFORMAT (S21)}$$

$$\delta = \arcsin\left(\sin 23.45^\circ \sin\left(\frac{360^\circ(284+n)}{365}\right)\right) \quad \backslash * \text{MERGEFORMAT}$$

(S22)

where  $r_t$  is the ratio of half-hourly radiation to the day radiation,  $\sigma$  is a constant determined by the noon time value of  $r_{t=12}$ , and  $S_0$  is the duration of daylight hours.

The real-time solar radiation intensity can then be calculated by specifying the noon-time radiation intensity  $I_{t=12}$  :

$$I_g = \frac{I_{t=12}}{r_{t=12}} r_t \quad \backslash * \text{MERGEFORMAT (S23)}$$

The output power of the PV panels is calculated as follows<sup>9</sup>:

$$P_{pv} = A_{pv} P_{STC} \frac{I_g}{I_{STC}} \left[ 1 + \alpha_{temp} (T - T_{STC}) \right] \quad \text{MERGEFORMAT} \quad (S24)$$

where  $P_{STC}$  is the PV output power at the standard test condition (1000 W m<sup>-2</sup>, 298 K), which is 188 W in this study.<sup>10</sup>  $\alpha_{temp}$  is the temperature coefficient, assumed as -0.39% K<sup>-1</sup>.<sup>10</sup> The real-time variation of solar radiation and PV power can be determined for any given location and day.

### 2.3 PEMFC, Electrolyzer, and Hydrogen Storage

The electrolyzer is used to convert surplus PV electricity into hydrogen energy for storage. The stored hydrogen is then utilized by the PEMFC system to sustain BPMED operation during periods of insufficient direct PV power. The power of the electrolyzer and PEMFC is calculated based on the power gap between the PV plant and the BPMED stack:

$$P_{PEMFC} = \max(0, P_{BPMED} - P_{PV}) \quad \text{MERGEFORMAT} \quad (S25)$$

$$P_{EL} = \max(0, P_{PV} - P_{BPMED}) \quad \text{MERGEFORMAT} \quad (S26)$$

Assuming constant energy efficiencies for both the electrolyzer and PEMFC, the hydrogen generation and consumption rates are expressed as:

$$q_{H_2,g} = \frac{\eta_{EL} P_{EL}}{HHV_{H_2}} \quad \text{MERGEFORMAT} \quad (S27)$$

$$q_{H_2,c} = \frac{P_{PEMFC}}{\eta_{PEMFC} HHV_{H_2}} \quad \text{MERGEFORMAT} \quad (S28)$$

where  $HHV_{H_2}$  is the high heating value of hydrogen. The electrolyzer and PEMFC efficiencies ( $\eta_{EL}$  and  $\eta_{PEMFC}$ ) are assumed to be 80%<sup>11</sup> and 50%<sup>12</sup>, respectively.

In the BPMED-PEMFC system, the hydrogen generation rate is derived from the BPMED current density as:

$$q_{H_2,g} = \frac{n_{\text{cell}} i A_{\text{cell}}}{2F} \quad \backslash * \text{MERGEFORMAT (S29)}$$

Discretizing the simulated system operation period and assuming stable state of each time point, the real-time hydrogen storage amount is expressed as:

$$m_{H_2,t+1} = m_{H_2,t} + (q_{H_2,g} - q_{H_2,c}) \Delta t \quad \backslash * \text{MERGEFORMAT (S30)}$$

The system's required hydrogen storage capacity is determined by the difference between the maximum and minimum hydrogen storage levels:

$$C_{H_2} = \max(m_{H_2,t}) - \min(m_{H_2,t}) \quad \backslash * \text{MERGEFORMAT (S31)}$$

## 2.4 Battery Storage

A constant efficiency assumption is also applied to the battery energy storage system. The real-time battery energy storage amount is expressed as:

$$E_{BS,t+1} = E_{BS,t} + \left( \eta_c P_{c,t} - \frac{P_{d,t}}{\eta_d} \right) \Delta t \quad \backslash * \text{MERGEFORMAT (S32)}$$

The charging efficiency  $\eta_c$  (ratio of the amount of energy stored in the battery to the amount of energy input) and discharging efficiency  $\eta_d$  (ratio of the useful energy output to the stored energy in the battery) are both assumed to be 95%.<sup>13</sup> The charging and discharging powers bridge the gap between PV power and BPMED power as:

$$P_{\text{charging}} = \max(0, P_{\text{PV}} - P_{\text{BPMED}}) \quad \backslash * \text{MERGEFORMAT (S33)}$$

$$P_{\text{discharging}} = \max(0, P_{\text{BPMED}} - P_{\text{PV}}) \quad \backslash * \text{MERGEFORMAT (S34)}$$

The required battery storage capacity is determined by the difference between the maximum and minimum battery energy levels:

$$C_{BS} = \max(E_{BS,t}) - \min(E_{BS,t}) \quad \backslash * \text{MERGEFORMAT (S35)}$$

### 3. Model Validation

#### 3.1 Validation of BPMED cell voltage

The experimental data from Castaño et al.<sup>14</sup> was collected to validate the BPMED cell with HER and OER. The cell design in this experiment differs slightly from the simulation design in this study. Specifically, the experimental cell consists of five BPMs and four CEMs, with each compartment having a thickness of 480  $\mu\text{m}$ . An additional BPM was used to sustain the ionic current within the system instead of a CEM.

The solution in the end compartments consisted of 0.5 M  $\text{K}_2\text{SO}_4$ , while the solution supplied to the acidic and basic compartments was  $\text{K}_2\text{CO}_3$  and  $\text{KHCO}_3$ , providing an overall concentration of 1 M  $\text{K}^+$  and 0.6 M DIC. The cell was tested under varying current densities, with the solution flow rate adjusted to maintain a load ratio of 0.9 (defined as the current divided by the  $\text{K}^+$  flow rate supplied to the compartments).

The mathematical model established in this study was applied to simulate the cell configuration used in Castaño et al.'s experiment.<sup>14</sup> Figure S2 presents the comparison of the simulation results with the experimental data.

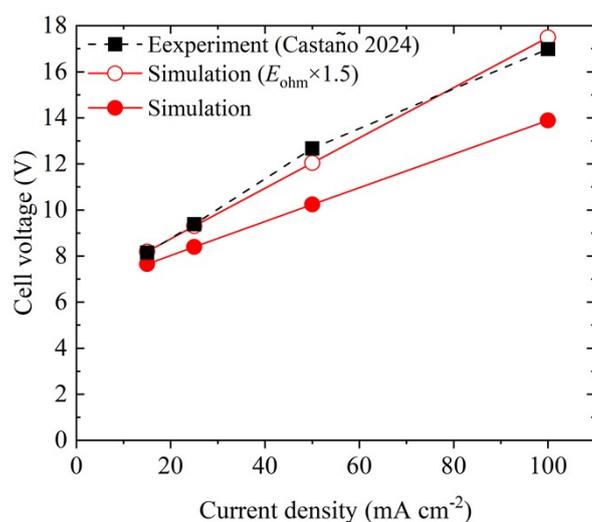


Figure S2. Comparison between experimental and simulated cell voltages of the BPMED with HER and OER.

The measured cell voltage keeps higher than the simulation results, and the gap increase

with the current density. However, both results show a linear increasing trend, implying the increase in cell voltage is dominated by the ohmic loss within the current density range. From Figure S2, the corrected cell voltage with 50% ohmic loss increasing shows the measured cell voltage in the study by Castaño et al.<sup>14</sup> consistently exceeds the simulation results, with the gap increasing as current density rises. However, both datasets exhibit a linear trend, suggesting that the rise in cell voltage is primarily governed by ohmic losses within the current density range tested.

Figure S2 demonstrates that the corrected cell voltage, incorporating a 50% increase in ohmic losses, shows a steeper slope and aligns more closely with the experimental data. The estimated ohmic loss depends on the resistances of the membranes and solutions, which vary considerably based on material selection and cell fabrication processes. This suggests that the simulated cell voltage could be achieved with further optimization of the cell. At lower current densities (10 - 40 mA cm<sup>-2</sup>), which are the focus of this study, ohmic losses constitute a smaller fraction of the total cell voltage. Consequently, the original simulation results are closer to the experimental data in this range.

For the BPMED cell driven by the K<sub>3</sub>/K<sub>4</sub>Fe(CN)<sub>6</sub> redox reaction, validation was conducted using experimental data from Digdaya et al.<sup>15</sup> Their cell design includes two end compartments for redox reactions and two BPM compartments for the pH swing, separated by two CEMs installed between the acidic/basic compartments and the end compartments. The end compartments and acidic/basic compartments have thicknesses of 5 mm and 2 mm, respectively.

The electrolyte in the end compartments consisted of 0.4 M K<sub>3</sub>/K<sub>4</sub>Fe(CN)<sub>6</sub> solution, while seawater (approximated as 0.5 M NaCl solution with 3.11 M DIC) was supplied to the acidic and basic compartments. The solution flow rate was maintained at 40 mL min<sup>-1</sup> under various current densities. Figure S3 compares the experimental and simulated cell voltages, showcasing the model's validation results.

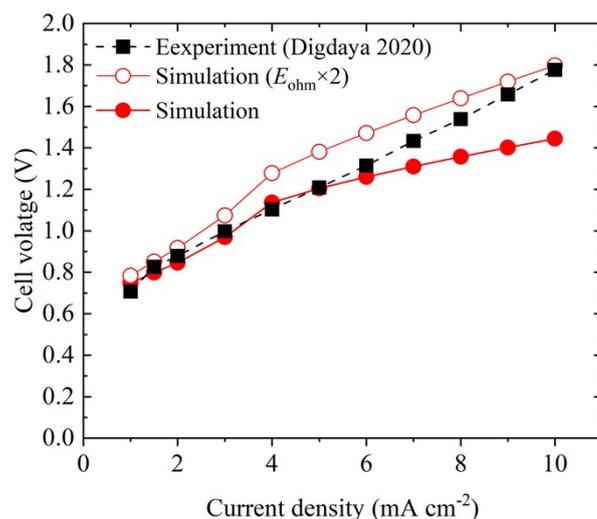


Figure S3. Comparison between experimental and simulated cell voltages of the BPMED with the  $K_3/K_4Fe(CN)_6$  redox couple.

The comparison of the redox-couple BPMED cell reveals a similar trend to that observed in the HER/OER cell. The simulated cell voltage closely matches the experimental data, though deviations in the voltage-current slope become apparent at higher current densities. Adjusting the estimation by doubling the ohmic loss provides better alignment with the experimental results. Overall, the error in the simulated cell voltage remains within an acceptable range, indicating that the model captures the key characteristics of the BPMED cell performance effectively.

### 3.2 Validation of Typical-Day Solar Radiation Intensity

The predicted hourly solar radiation intensity, based on Baig et al.'s model,<sup>8</sup> is compared with open data from the Photovoltaic Geographical Information System (PVGIS). Figure S4 illustrates the comparison results for selected locations and dates.

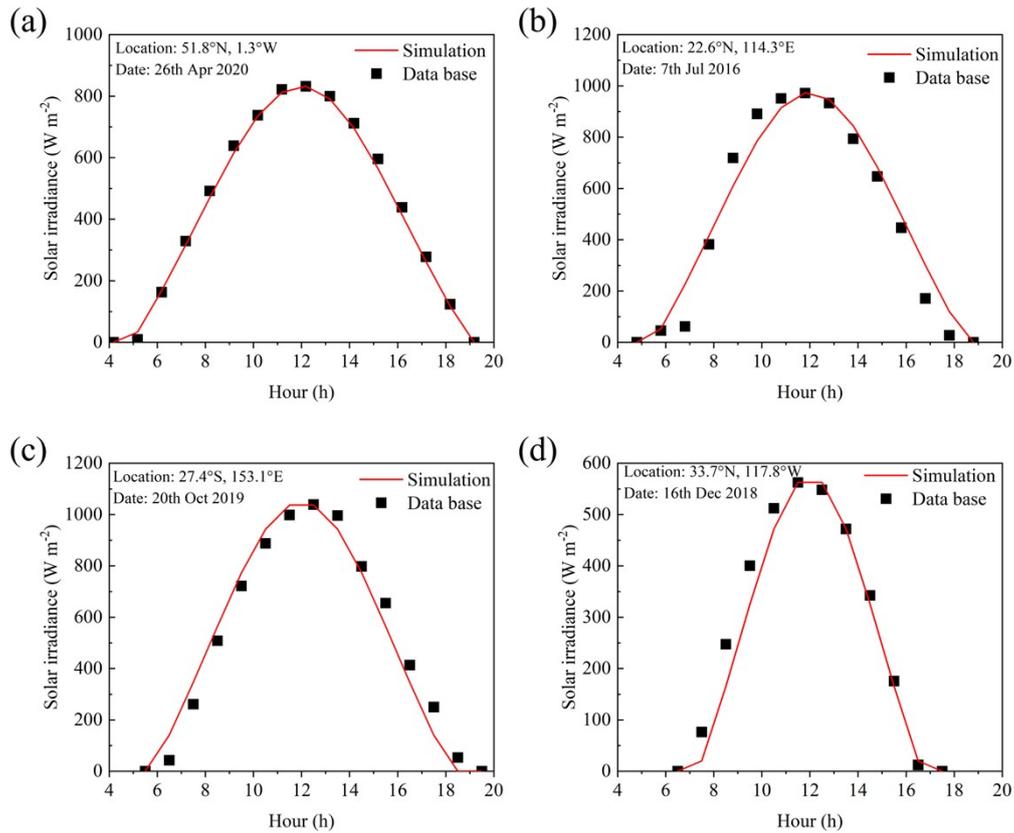


Figure S4. Comparison of daytime solar irradiance between recorded data and model predictions.

The results demonstrate reasonable agreement between the modeled and observed data, validating the reliability of Baig et al.'s model for capturing the diurnal variation in solar radiation intensity across different geographical and temporal conditions. Discrepancies observed in certain instances may be attributed to variations in local atmospheric conditions and the simplified assumptions inherent in the model.

#### 4. Simulation Operating Logic of Systems

The simulation procedure for the systems is depicted in Figure S5. Environmental conditions, including solar irradiance and temperature, along with system parameters, serve as initial inputs. Iterations are performed to determine the appropriate BP MED stack size to ensure the system's overall energy input and consumption are balanced. The systems are entirely driven by renewable solar electricity. This balance is achieved when the energy storage levels at the end of the simulation return to their initial levels, indicating that all solar energy input is fully utilized for DAC without requiring external

energy sources.

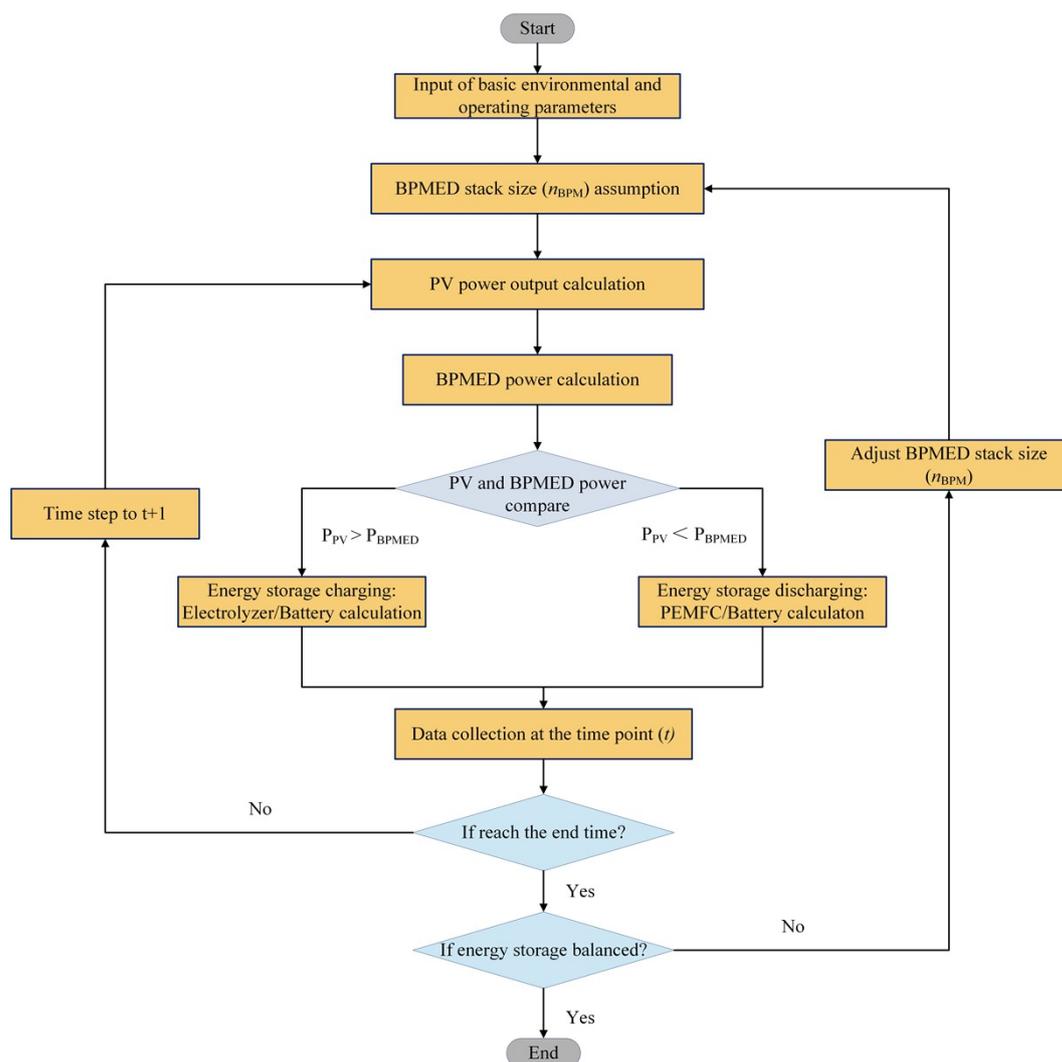


Figure S5. Generalized system simulation flow chart

The entire simulated system operation period is discretized into a series of time intervals (e.g., half an hour or one hour). The system is assumed to operate stably within each interval. The end energy storage amount is compared to the beginning value, and the BPMED stack size is iteratively adjusted to match the overall PV power input. Energy storage systems, including electrolyzers, PEMFCs, and batteries, bridge the power gap between the BPMED and PV outputs.

For the BPMED-PEMFC system, energy storage charging (hydrogen generation) is directly coupled with BPMED operation. In this case, the operating current density adjusts dynamically based on PV direct power input. During periods of low PV input (e.g., nighttime), the PEMFC consumes stored hydrogen to maintain a minimum BPMED operating load and current density. An additional internal iteration is

performed to determine this minimum BPMED current density specific to the BPMED-PEMFC system.

## 5. Effect of Repetitive BPM Unit Number in Single Cells

The BPMED-PEMFC system was simulated under various configurations with different numbers of single-cell BPMs. Figure S6 compares the BPMED current densities and daily CO<sub>2</sub> capture amounts, with all configurations evaluated under identical PV power input and an average current density of 10 mA cm<sup>-2</sup> throughout the day.

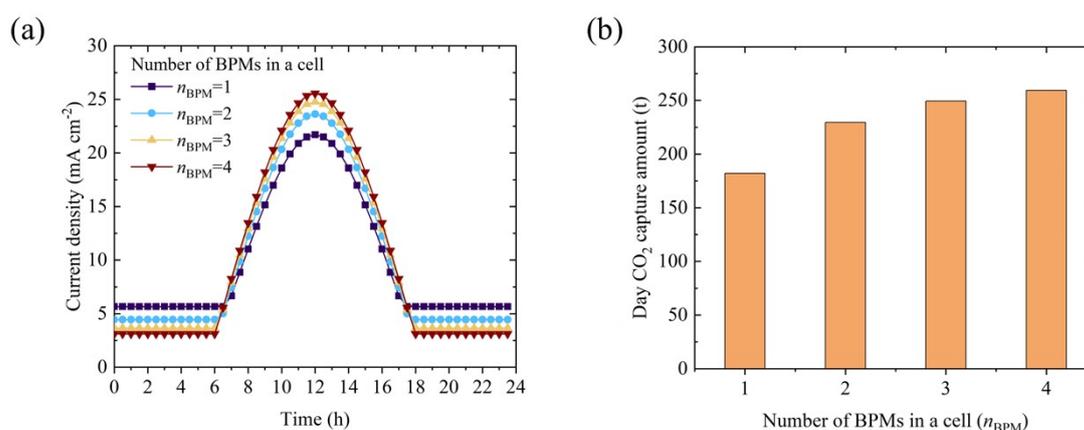


Figure S6. Effect of BPM unit number in HER/OER cells: (a) Current density over the day; (b) System daily DAC capacity.

In all cases, the load of HER/OER cells and the BPMED stack fluctuates with the directly input PV electricity during the daytime, as there is no stand-alone electrolyzer to flexibly absorb excess PV electricity. As the number of BPM units increases, the overall cell voltage and power consumption rise. However, the proportion of energy allocated to HER/OER in the end compartments decreases within the total energy consumption, widening the gap between the available energy during daytime and nighttime. Consequently, the fluctuations in BPMED load and current density throughout the day, shown in Figure S6 (a), become more pronounced with increasing BPM numbers. Conversely, configurations with more BPMs benefit from reduced overall losses in hydrogen generation and utilization, leading to enhanced DAC productivity, as shown in Figure S6 (b). Ultimately, to minimize current density fluctuations and better align with the constant current density of redox-couple cells and

systems, a single BPM unit configuration is chosen for HER/OER cells.

In contrast, systems using redox-couple cells can achieve constant operating loads and current densities. Figure S7 compares the daily CO<sub>2</sub> capture capacity of the BPMED-BS system with varying single-cell BPM numbers.

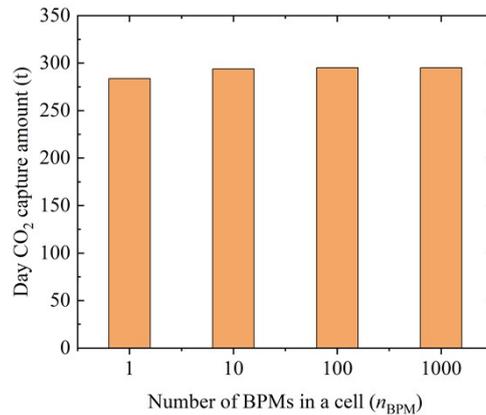


Figure S7. Effect of BPM unit number in redox-couple cells on system DAC performance.

As the number of BPMs in a single cell increases, the potential loss in the end compartments of the redox reaction is distributed across more BPM units, leading to improved energy efficiency and enhanced system DAC performance. However, the gain in daily CO<sub>2</sub> capture becomes negligible beyond 10 BPMs, with only a 0.37% increase observed when the BPM count rises from 10 to 100. This indicates diminishing returns for BPM numbers exceeding 10. Consequently, a configuration with 10 BPM units is selected as the optimal solution for redox-couple cells, balancing energy efficiency and structural simplicity.

## 6. Method of Economic Calculation

The DAC total cost comprises capital investment and operation and maintenance (O&M) expenses, and is determined using the following equation<sup>6</sup>:

$$TC = \frac{(TOC \cdot CCF) + C_{\text{O\&M}}^{\text{fix}} + C_{\text{O\&M}}^{\text{var}} h_{\text{eq}}}{R_{\text{CO}_2} h_{\text{eq}}} \quad \text{MERGEFORMAT} \quad (*)$$

(S36)

where  $TOC$  is total overnight cost,  $CCF$  is the capital charge factor and assumed as

12.5%,<sup>6</sup>  $C_{O\&M}^{fix}$  and  $C_{O\&M}^{var}$  are the fixed and variable O&M costs, respectively.  $R_{CO_2}$  stands for the total DAC amount in one full year obtained from system simulation.  $h_{eq}$  represents the percentage of working hours assumed to be 90%.<sup>6</sup>

Table S1 summarizes the calculation method for the TOC. The TOC is estimated based on the bare erected cost (BEC), which represents the overall costs of purchasing the components and materials listed in

Table S2.

Table S1. Calculation of TOC<sup>6</sup>

| Items   | Value      |
|---|------------|
| Total installation cost (TIC)                   | 80% BEC    |
| Total direct plant cost (TDPC)                  | BEC + TIC  |
| Indirect cost (IC)                              | 14% TDPC   |
| Engineering, procurement and construction (EPC) | TDPC + IC  |
| Total contingencies and owner's cost (TCOC)     | 15% EPC    |
| Total overnight cost (TOC)                      | EPC + TCOC |

Table S2. Assumptions of component, material, and other fixed costs

| Items   | Value                                  |
|---|--|
| <b>Fixed operation and maintenance costs</b>    |  |
| Labor   | 1.5 M\$ <sup>6</sup>                   |
| Maintenance                                     | 2.5% TOC <sup>6</sup>                  |
| Insurance                                       | 2.0% TOC <sup>6</sup>                  |
| <b>Variable operation and maintenance costs</b> |  |
| CEM   | 75 \$ m <sup>-2</sup> <sup>6</sup>     |
| BPM   | 750 \$ m <sup>-2</sup> <sup>6</sup>    |
| CEM & BPM lifetime                              | 3 years <sup>6</sup>                   |
| PEMFC   | 2400 \$ kW <sup>-1</sup> <sup>17</sup> |
| PEMFC lifetime                                  | 5 years <sup>18</sup>                  |
| Electrolyzer (alkaline water)                   | 340 \$ kW <sup>-1</sup> <sup>19</sup>  |
| Electrolyzer lifetime                           | 10 years <sup>20</sup>                 |

|   |  |
|---|--|
| Energy storage battery                              | 137 \$ kWh <sup>-1</sup> <sup>21</sup> |
| Battery lifetime                                    | 5 years <sup>22</sup>                  |
| <b>Other purchasing costs</b>                       |  |
| PV panels   | 63 \$ m <sup>-2</sup> <sup>21</sup>    |
| BPMED end electrodes                                | 22 \$ m <sup>-2</sup> <sup>23</sup>    |
| Hydrogen storage tank                               | 577 \$ kg <sup>-1</sup> <sup>24</sup>  |
| Air contactor (scale of 1 Mt-CO <sub>2</sub> /year) | 114.2 M\$ <sup>25</sup>                |

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The entire plant's lifetime is assumed to be 20 years. The lifetimes of critical components, such as electrodes and PV panels, are sufficient to cover this period, with electrodes lasting up to 20 years<sup>16</sup> and PV panels lasting approximately 25–30 years.<sup>26</sup> Additionally, components such as the hydrogen tank and air contactor are generally not expected to require replacement during the plant's lifetime. Consequently, the costs associated with these components are excluded from the variable operation and maintenance costs.

## 7. Variation Coefficient of Daily Solar Irradiance

The original hourly solar irradiance data for different locations were obtained from PVGIS and subsequently aggregated into daily values using 24-hour intervals. The annual total solar irradiance at a given location is calculated as:

$$H_y = \sum_{i=1}^N H_i \quad \backslash * \text{MERGEFORMAT (S37)}$$

where  $H_i$  is the solar irradiance of the  $i$ -th day, and  $N$  is the total number of days in the year. The annual daily average solar irradiance is then defined as:

$$\mu = \frac{1}{N} H_y = \frac{1}{N} \sum_{i=1}^N H_i \quad \backslash * \text{MERGEFORMAT (S38)}$$

Standard deviation is a fundamental statistical measure used to quantify the dispersion of daily solar irradiance around its annual mean value. It is calculated as:

$$\sigma = \sqrt{\frac{1}{N-1} \sum_{i=1}^N (H_i - \mu)^2} \quad \backslash * \text{MERGEFORMAT (S39)}$$

While the standard deviation reflects the absolute magnitude of daily solar variability, it depends on the scale and units of the data, which limits its applicability for comparing variability across datasets with different magnitudes. To overcome this limitation, the coefficient of variation of daily solar irradiance is introduced as:

$$CV = \frac{\sigma}{\mu} \quad \backslash * \text{MERGEFORMAT (S40)}$$

## 8. Regional Solar Irradiance and DAC Cost Data for China and the UK

The data from the 50 sites in China and the UK used in Figure 5 are summarized as follows:

Table S3. Site data of China

| Site number | Longitude (°) | Latitude (°) | DAC total cost (\$ t <sup>-1</sup> -CO <sub>2</sub> ) | Annual total solar irradiance (kWh m <sup>-2</sup> ) | Variation coefficient of daily solar irradiance |
|-------------|---------------|--------------|---|--|---|
| 1           | 109.81        | 19.046       | 4733.652859   | 985.27068  | 0.563559524                                     |
| 2           | 120.709       | 22.74        | 4947.748504   | 1125.97812   | 0.463374051                                     |
| 3           | 108.844       | 22.902       | 5698.414985   | 819.29906  | 0.709629526                                     |
| 4           | 104.976       | 23.629       | 5773.557251   | 715.11834  | 0.759735781                                     |
| 5           | 113.853       | 23.629       | 4277.687372   | 890.69043  | 0.600752636                                     |
| 6           | 121.236       | 24.591       | 6827.080622   | 869.61253  | 0.646314224                                     |
| 7           | 100.494       | 24.911       | 4687.326204   | 1189.69785   | 0.539474984                                     |
| 8           | 117.721       | 25.547       | 4378.849226   | 924.52735  | 0.635109179                                     |
| 9           | 108.524       | 26.612       | 7210.474446   | 540.52742  | 1.026290411                                     |
| 10          | 111.129       | 27.59        | 6035.139652   | 691.26489  | 0.907093951                                     |
| 11          | 104.185       | 27.668       | 5790.486578   | 649.03168  | 0.961209284                                     |
| 12          | 115.26        | 27.902       | 4831.709506   | 881.54996  | 0.735917083                                     |
| 13          | 119.918       | 29.214       | 4351.798809   | 844.84553  | 0.815837799                                     |
| 14          | 91.002        | 29.673       | 2740.034957   | 1686.93029   | 0.32667056                                      |
| 15          | 85.377        | 29.902       | 2595.32043  | 1926.67233   | 0.275286213                                     |
| 16          | 98.033        | 30.812       | 3383.493338   | 1630.18901   | 0.375622359                                     |
| 17          | 103.043       | 31.114       | 5726.634494   | 675.74372  | 0.713292253                                     |
| 18          | 114.557       | 31.264       | 3752.520556   | 930.58274  | 0.700320157                                     |
| 19          | 109.019       | 31.414       | 5059.121332   | 759.32249  | 0.79920564                                      |
| 20          | 119.83        | 32.384       | 4087.691334   | 950.89958  | 0.713454964                                     |
| 21          | 85.201        | 33.123       | 2199.500154   | 1929.76025   | 0.274457775                                     |
| 22          | 94.078        | 33.564       | 2805.125516   | 1635.01265   | 0.375659863                                     |
| 23          | 107.086       | 33.564       | 4496.610225   | 853.09005  | 0.764086302                                     |
| 24          | 89.288        | 33.82        | 2520.510411   | 1834.44145   | 0.30518694                                      |
| 25          | 112.183       | 34.002       | 4010.455272   | 980.73091  | 0.666346237                                     |

|    |         |        |             |            |             |
|----|---------|--------|-------------|------------|-------------|
| 26 | 99.879  | 34.293 | 3283.412502 | 1371.78858 | 0.47851309  |
| 27 | 117.193 | 34.655 | 3850.561861 | 1082.86411 | 0.60808108  |
| 28 | 81.422  | 36.089 | 2257.731144 | 1737.18591 | 0.314434539 |
| 29 | 116.842 | 37.287 | 3323.270755 | 1250.1121  | 0.48123101  |
| 30 | 93.99   | 37.426 | 2364.941868 | 1903.48352 | 0.287737519 |
| 31 | 101.9   | 37.983 | 2869.514267 | 1430.71686 | 0.412191252 |
| 32 | 109.635 | 38.466 | 3018.832291 | 1431.5115  | 0.434172364 |
| 33 | 87.223  | 38.535 | 2597.192452 | 1766.81959 | 0.303079509 |
| 34 | 105.711 | 38.535 | 2841.70145  | 1561.3341  | 0.365958615 |
| 35 | 79.137  | 39.423 | 2586.136779 | 1677.97379 | 0.322786066 |
| 36 | 116.578 | 40.568 | 3103.619251 | 1309.55703 | 0.426211264 |
| 37 | 87.223  | 41.1   | 2708.950223 | 1737.59715 | 0.282280138 |
| 38 | 107.701 | 41.496 | 2571.625687 | 1559.65411 | 0.327828948 |
| 39 | 122.73  | 41.496 | 3052.694684 | 1325.42662 | 0.452044607 |
| 40 | 98.912  | 41.627 | 2485.860037 | 1711.51288 | 0.291605795 |
| 41 | 112.008 | 42.605 | 2584.625159 | 1505.70588 | 0.342552802 |
| 42 | 91.09   | 42.67  | 2445.491382 | 1754.62476 | 0.26095949  |
| 43 | 125.982 | 42.799 | 3447.189821 | 1195.47774 | 0.523467307 |
| 44 | 84.146  | 43.886 | 2845.61635  | 1489.39176 | 0.419064951 |
| 45 | 130.113 | 44.953 | 3302.073156 | 1178.68035 | 0.507999649 |
| 46 | 116.666 | 45.139 | 2660.356906 | 1382.909   | 0.381000577 |
| 47 | 87.223  | 47.327 | 4167.574253 | 1466.42017 | 0.442607099 |
| 48 | 120.885 | 47.327 | 3307.750557 | 1263.04956 | 0.45800515  |
| 49 | 127.213 | 47.92  | 3599.283165 | 1225.56131 | 0.513149636 |
| 50 | 123.258 | 52.526 | 4546.812179 | 1169.36562 | 0.544894293 |

Table S4. Site data of the UK

| Site number | Longitude (°) | Latitude (°) | DAC total cost (\$ t <sup>-1</sup> -CO <sub>2</sub> ) | Annual total solar irradiance (kWh m <sup>-2</sup> ) | Variation coefficient of daily solar irradiance |
|-------------|---------------|--------------|---|--|---|
| 1           | -4.965        | 50.367       | 5934.359526   | 838.04843  | 0.743999523                                     |
| 2           | -3.891        | 50.739       | 5989.158359   | 881.0781   | 0.716845793                                     |
| 3           | -3.012        | 50.94        | 6263.303135   | 825.957  | 0.76187912                                      |
| 4           | -2.108        | 50.955       | 6094.681362   | 861.05971  | 0.732130214                                     |
| 5           | -0.302        | 51.047       | 6335.949605   | 879.43412  | 0.721646613                                     |
| 6           | 0.675         | 51.185       | 6309.531568   | 974.64142  | 0.681740865                                     |
| 7           | -1.278        | 51.201       | 6065.46742  | 834.85333  | 0.74680013                                      |
| 8           | -1.464        | 51.46        | 6839.103761   | 650.7384   | 0.973330054                                     |
| 9           | 0.187         | 51.582       | 6441.963987   | 825.70782  | 0.731790426                                     |
| 10          | -2.67         | 51.778       | 6478.622124   | 708.7764   | 0.831455203                                     |
| 11          | -2.123        | 51.824       | 8036.692198   | 575.17503  | 1.087336071                                     |
| 12          | -4.281        | 51.854       | 6325.389444   | 852.14801  | 0.764207101                                     |
| 13          | -0.375        | 51.914       | 6533.101292   | 753.11677  | 0.779336297                                     |
| 14          | -1.571        | 51.929       | 6141.962254   | 735.70157  | 0.805593381                                     |
| 15          | 0.431         | 51.959       | 6434.163106   | 832.29614  | 0.72529659                                      |
| 16          | -1.244        | 52.304       | 6425.946318   | 644.57533  | 0.956065103                                     |
| 17          | -3.622        | 52.379       | 6522.876875   | 702.11332  | 0.870701088                                     |
| 18          | -0.229        | 52.423       | 6294.768844   | 780.4598   | 0.757275065                                     |

|    |        |        |             |           |             |
|----|--------|--------|-------------|-----------|-------------|
| 19 | 0.724  | 52.423 | 6525.353614 | 810.66725 | 0.733949984 |
| 20 | -2.06  | 52.512 | 6259.076215 | 728.4711  | 0.816054703 |
| 21 | -0.131 | 52.72  | 6263.92679  | 784.79787 | 0.75090047  |
| 22 | -3.573 | 52.927 | 7095.181178 | 676.15674 | 0.906669504 |
| 23 | -1.389 | 53.12  | 6838.052455 | 595.07526 | 1.014890332 |
| 24 | -0.497 | 53.147 | 6229.229208 | 773.50225 | 0.760057948 |
| 25 | -2.499 | 53.468 | 6494.81585  | 736.05002 | 0.827691114 |
| 26 | -1.536 | 53.775 | 6856.549416 | 609.16854 | 1.004445815 |
| 27 | -0.814 | 53.844 | 6260.575054 | 794.36368 | 0.748699125 |
| 28 | -2.523 | 54.045 | 7542.850333 | 723.51214 | 0.873484812 |
| 29 | -1.01  | 54.188 | 6437.241641 | 770.6999  | 0.771702254 |
| 30 | -6.176 | 54.331 | 7840.437021 | 542.31395 | 1.076054188 |
| 31 | -2.073 | 54.376 | 10633.58262 | 468.80248 | 1.406347236 |
| 32 | -7.494 | 54.444 | 8153.896066 | 477.03456 | 1.173207189 |
| 33 | -2.89  | 54.501 | 7843.318519 | 658.00157 | 0.939427207 |
| 34 | -1.645 | 54.756 | 6778.482227 | 753.22718 | 0.804908253 |
| 35 | -6.444 | 54.91  | 8128.554425 | 509.26859 | 1.109920558 |
| 36 | -3.915 | 55.022 | 7014.462492 | 736.64388 | 0.854719394 |
| 37 | -2.283 | 55.03  | 7722.791261 | 560.42292 | 1.089283312 |
| 38 | -4.403 | 55.26  | 6893.722243 | 718.15805 | 0.856095447 |
| 39 | -3.207 | 55.371 | 6773.296193 | 716.97284 | 0.849616155 |
| 40 | -2.182 | 55.564 | 7357.306775 | 754.68367 | 0.832183425 |
| 41 | -4.574 | 56.099 | 6958.845388 | 701.89754 | 0.855871203 |
| 42 | -3.402 | 56.371 | 7567.429367 | 713.739   | 0.874237094 |
| 43 | -4.452 | 56.667 | 7844.765808 | 611.38468 | 0.977311821 |
| 44 | -3.134 | 56.801 | 7178.817102 | 733.98905 | 0.833765083 |
| 45 | -2.938 | 57.253 | 6642.841651 | 758.3082  | 0.790998423 |
| 46 | -4.11  | 57.292 | 8812.331565 | 600.44965 | 1.012592016 |
| 47 | -5.282 | 57.292 | 9663.892415 | 534.49378 | 1.11780315  |
| 48 | -4.818 | 57.894 | 8403.638211 | 604.56045 | 0.979919666 |
| 49 | -6.649 | 58.178 | 6731.11556  | 784.23901 | 0.820681546 |
| 50 | -3.793 | 58.345 | 7258.177182 | 765.84186 | 0.783823131 |

## 9. Grid-Assisted Electricity Supply

In grid-assisted operating scenarios, PV electricity remains the primary energy source for BPMED operation. Grid electricity is introduced only when instantaneous PV generation is insufficient to meet the electrical demand required by the BPMED stack and associated subsystems. Grid electricity therefore serves as a supporting or transitional supply, rather than a continuous primary power source. This operating strategy reflects realistic deployment conditions for renewable-powered industrial systems and enables systematic identification of the lower bound of electricity cost under constrained renewable availability.

Two distinct metrics are defined to characterize grid-assisted operation: Grid support

fraction,  $f_{\text{grid}}^{\text{support}}$ , defined as the fraction of BPMED electrical demand supplied by the grid during time steps when PV generation is insufficient. Annual grid electricity share,  $\varphi_{\text{grid}}$ , defined as the ratio of grid-supplied electricity to the total annual electricity consumption of the BPMED system. The grid support fraction is a prescribed operational parameter, whereas the annual grid electricity share emerges as a system-level outcome from time-resolved simulation.

At each simulation time step, the electrical demand of the BPMED system is compared with available PV power. When BPMED demand power is higher than PV output power, the grid power is calculated as:

$$P_{\text{grid}} = f_{\text{grid}}^{\text{support}} (P_{\text{BPMED}} - P_{\text{PV}}) \quad \backslash * \text{MERGEFORMAT (S41)}$$

The remaining demand is met by energy storage where applicable. Over an annual period, the total grid electricity input is integrated to compute annual grid electricity share:

$$\varphi_{\text{grid}} = \frac{\sum P_{\text{grid}}}{\sum P_{\text{BPMED}}} \times 100\% \quad \backslash * \text{MERGEFORMAT (S42)}$$

Grid-assisted scenarios represent transitional or hybrid deployment strategies and are not intended to describe fully decarbonized end states. Sensitivity to grid carbon intensity is not explicitly modeled here but does not affect the structural conclusions regarding cost reduction pathways and electricity cost lower bounds.

In the system economic calculations, the price of grid electricity is assumed to be constant at 80 \$ MWh<sup>-1</sup> to simplify the analysis and enable consistent comparison across different scenarios.

## 10. Levelized Cost of PV Electricity

Table S5 summarizes recent reports on the levelized cost of electricity (LCOE) from PV power generation. Three primary strategies have emerged to address PV intermittency and enable more competitive LCOEs: integration with complementary energy sources; deployment of advanced energy storage systems; connection to the power grid as a large-scale buffer. Reported LCOEs generally range from 40 to 600 \$ MWh<sup>-1</sup>, with the majority falling between 50 and 400 \$MWh<sup>-1</sup>. This range can be

considered a representative benchmark for PV-based electricity generation.

Table S5. Summary of LCOEs of PV electricity

| Power generation   | Energy storage                    | Off-grid | LCOE (\$ MWh <sup>-1</sup> ) | Reference |
|--------------------|-----------------------------------|----------|------------------------------|-----------|
| PV                 | Battery                           | Yes      | 160                          | 27        |
| PV                 | Battery                           | No       | 193-273                      | 28        |
| PV and wind        | Thermal                           | Yes      | 122                          | 29        |
| PV and wind        | Battery                           | Yes      | 181                          | 29        |
| PV and wind        | Hydrogen                          | Yes      | 122                          | 29        |
| PV and wind        | Thermal                           | Yes      | 223                          | 29        |
| PV and wind        | Battery, hydrogen,<br>and thermal | Yes      | 313                          | 30        |
| PV, wind and CSP   | Battery and thermal               | No       | 53-160                       | 31        |
| PV and diesel      | Battery                           | Yes      | 569-595                      | 32        |
| PV and SOFC        | N.A.                              | Yes      | 40-110                       | 27        |
| PV and natural gas | Liquid air                        | No       | 91                           | 33        |
| PV and biogas      | Battery                           | No       | 280                          | 34        |

## 11. Reported Performance of DAC Systems

Table S6 summarizes the reported energy consumption and cost ranges of major commercial and pilot-scale DAC systems. These data represent the most widely cited benchmarks for assessing the techno-economic status of currently deployed or announced DAC technologies.

Table S6. Summary of main deployed DAC technologies, energy consumption, and cost.

| Company/Plant                          | Method                      | Energy consumption<br>(kJ mol <sup>-1</sup> -CO <sub>2</sub> ) | Cost (\$ t <sup>-1</sup> -CO <sub>2</sub> ) |
|--|-----------------------------|--|---|
| Carbon Engineering <sup>25,35</sup>    | Liquid wet-<br>scrubbing    | 388 (natural gas)  | 94-232                                      |
| Climeworks Orca <sup>35-38</sup>       | Solid sorbent-<br>scrubbing | 317 (heat and<br>electricity)                                  | 500-600                                     |
| Climeworks 2030<br>target <sup>a</sup> | Solid sorbent-<br>scrubbing | ~159 <sup>b</sup>  | 250-350                                     |

|                   |                           |      |                   |
|-------------------|---------------------------|------|-------------------|
| Global Thermostat | Solid amine-<br>scrubbing | N.A. | <100 <sup>c</sup> |
|-------------------|---------------------------|------|-------------------|

a, Publicly claimed by Climeworks in 2024. b, Estimated as approximately half of the current energy consumption. c, Claimed by Global Thermostat, but not yet validated by publicly disclosed data.

## 12. Electrochemical Cells for CO<sub>2</sub> Capture

Table S7 summarizes representative electrochemical systems that have been explored for CO<sub>2</sub> capture and release. The four main categories include BPMED, proton-coupled electrolysis, capacitive deionization, and redox-active carrier systems. These approaches differ substantially in reaction mechanisms, operational potentials, and cost-determining components.

Table S7. Overview of electrochemical cells for CO<sub>2</sub> capture.

| Electrochemical cells                        | Voltage range (V)    | Energy consumption (kJ mol <sup>-1</sup> -CO <sub>2</sub> ) | Key costs                                   |
|--|----------------------|---|---|
| BPMED <sup>a</sup>                           | 1.0-2.4              | 375-937   | Membranes                                   |
| Proton-coupled electrolysis <sup>39-43</sup> | 0.5-2.0              | 100-300 <sup>b</sup>  | Electrode, catalyst                         |
| Capacitive deionization <sup>44-46</sup>     | <1 <sup>c</sup>      | 30-60 <sup>b</sup>  | Electrode, membrane, current collector      |
| Redox-active carriers <sup>47-49</sup>       | 0.2-1.2 <sup>d</sup> | ≈100  | Electrode, redox-absorption medium, solvent |

a, Simulation results based on Shenzhen case. b, The lower values are normally for rich CO<sub>2</sub> sources (flue gas). c, Operation voltage of charging and discharging. d, Varies significantly among different redox systems.

Overall, these electrochemical pathways offer promising routes for low-temperature, electrically driven carbon capture. However, the actual energy performance depends strongly on CO<sub>2</sub> concentration (e.g., flue gas vs. DAC), system design, and material durability.

### 13. Sensitivity Analysis of Electrochemical Cells

The baseline values and limits for the evaluated electrochemical DAC systems are summarized in Table S8. The simulations are conducted under the Shenzhen case with a constant electrochemical device load (i.e., fixed current density). Assuming a fixed LCOE of 50 \$ MWh<sup>-1</sup> and based on Figure 7B, the corresponding optimal current density for the BPMED system is 80 mA cm<sup>-2</sup>. Accordingly, the baseline values are derived from the BPMED simulation and are listed in Table S8. To account for the potentially wide performance range of other electrochemical technologies, the lower and upper limits of these parameters are defined.

Table S8. Parameters for electrochemical DAC sensitivity analysis.

| Parameters                                | Baseline value | Lower limit | Upper limit |
|---|----------------|-------------|-------------|
| Cell voltage (V)                          | 2.13           | 0.5         | 4           |
| Current density (mA cm <sup>-2</sup> )    | 80             | 20          | 100         |
| Current efficiency (%)                    | 52.29          | 20          | 80          |
| Cell lifetime (year)                      | 5              | 1           | 10          |
| Cell unit area cost (\$ m <sup>-2</sup> ) | 836.90         | 100         | 1000        |

Here, the current efficiency is defined as the ratio of charge associated with captured CO<sub>2</sub> to the total charge transferred through the cell:

$$\eta_F = \frac{2n_{\text{CO}_2}F}{iA_{\text{cell}}} \times 100\% \quad \backslash * \text{MERGEFORMAT (S43)}$$

where  $n_{\text{CO}_2}$  is the DAC rate derived from a single cell/single BPM unit.

The cell unit area cost represents the total material cost required to fabricate a cell of a given area. For example, in the case of BPMED, it is calculated as the sum of the costs of the BPM and the stack-averaged unit costs of the CEM and the electrodes.

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