Supplementary Materials for
A machine learning-aided equilibrium model of VTSA processes for sorbents screening applied to CO₂ capture from diluted sources

Alexa Grimm⁹ and Matteo Gazzani⁹,‡,⋆

⁹ Universiteit Utrecht, Copernicus Institute of Sustainable Development, Princetonlaan 8a, 3584 CB, Utrecht, The Netherlands
‡ Sustainable Process Engineering, Chemical Engineering and Chemistry, Eindhoven University of Technology, 5612 AP Eindhoven, The Netherlands

⋆ Corresponding author. Email: m.gazzani@uu.nl
1 Modelling details

1.1 Blow-down step: pressure profile and time

The time for the blow-down step is mainly dependent on the vacuum pressure. By fitting simulation data from the 1D model, the following equation was received

\[ t_{BD} = \frac{-10p_{vac}}{b} + a \]  

(S1)

with the parameters listed in Table S1. Since the profile mainly varies with pressure and does not show a notable dependence on the density or temperature of the material, the fitting was carried out for a small dataset for one material (case s2 E-A) and different vacuum pressures over time.

**Table S1:** Fitting parameters for the time of the blow-down step.

| a   | b     |
|-----|-------|
| 20.70 | 20 000 |

The pressure at each sub-step \( p_{vac}^k \) is calculated using the following equation

\[ p_{vac}^k = \frac{1}{10} \left( \frac{a}{p_{vac}^k} \exp \left( c \cdot t_{BD} \right) + p_{vac}^k \exp \left( d \cdot t_{BD} \right) \right) \]  

(S2)

The fitted parameters are listed in Table S2.

**Table S2:** Fitting parameters for the pressure profile of the blow-down step.

| a   | b   | c   | d            |
|-----|-----|-----|--------------|
| 0.18627 | 0.85 | -0.1238 | -7.138 e^{-05} |

1.2 Heating step: temperature profile and time

The training data of the neural network for determining the heating time can be found in Figure S1.

The temperature profile was received by fitting several profiles retrieved from simulations using the 1D model. The profile is dependent on the desorption temperature and the heating time

\[ T_{heat}^k = (a + T_{des} + b \cdot T_{des}^k) \cdot atan \left( \frac{t_{heat}^k - d \cdot 10^9 \cdot T_{des}^k}{f} \right) + (T_{des} - g) \cdot h \]  

(S3)

The fitted parameters are listed in Table S3.

**Table S3:** Fitting parameters for the temperature profile of the heating step.

| a           | b           | c       | d       | e       | f       | g          | h       |
|-------------|-------------|---------|---------|---------|---------|------------|---------|
| -687.31     | 2791.97     | 0.346   | -661.53 | -4.296  | 42.005  | -34162.55  | 0.00867 |
Figure S1: Resulting training data (blue symbols) from the neural network together with the validation data (red) and the testing data (black).

1.3 Cooling step: profile

The time of the cooling step is fixed to 350 seconds, similar to the 1D model simulations. The temperature profile is calculated similar to the heating step, by fitting data from the 1D model. The fitting equation is

$$T_{\text{cooling}}^k = 293 + \left( aT_{\text{des}} \exp \left( b \cdot t_{\text{cool}}^k \right) \right)$$  \hspace{1cm} (S4)

with the parameters listed in Table S4.

Table S4: Fitting parameters for the temperature profile of the cooling step.

| a   | b            |
|-----|--------------|
| 0.17485 | -0.02629    |

1.4 Adsorption step

The total time of the adsorption step can be determined by including the air velocity $u_{\text{air}}$ and the geometry of the considered sorbent

$$t_{\text{ads}} = \frac{N_{\text{feed}} M_{\text{air}}}{u_{\text{air}} A \rho_{\text{air}}}$$  \hspace{1cm} (S5)

with $M_{\text{air}}$ being the molar mass of the air, $A$ the column cross section and $\rho_{\text{air}}$ the density of the air.

1.5 Saturation level

Since we are considering an equilibrium model, the saturation at the end of the adsorption step or rather at the begin of the blow-down step is generally 100%. A more realistic picture of the saturation level gives the simulation with the detailed model, which shows a lower saturation level. Figure S2 shows the saturation level calculated for the Pareto points from the optimization of three different materials (the same optimization as used for the validation). The saturation level $\alpha$ was calculated for each grid point of the bed by calculating the actual loading referred to the full saturation at ambient conditions.
Figure S2: Saturation level from simulations using the 1D model.

\[ \alpha = \sum_{n=1}^{N_{\text{grid}}} \left( q_{\text{CO}_2}^* \left( T_{\text{amb}}, p_{\text{amb}}, y_{\text{CO}_2}, \text{feed} \right) - q_{\text{CO}_2}^0 \left( T, p, y_{\text{CO}_2} \right) \right) \]

with \( q_{\text{CO}_2}^0 \) being the actual loading at the respective grid point and \( q_{\text{CO}_2}^* \) the loading at equilibrium.

Using this data, a neural network was trained to determine the saturation level dependent on the particle density \( \rho_p \), the desorption temperature \( T_{\text{des}} \), the vacuum pressure \( p_{\text{vac}} \) as well as the air volume flow \( \dot{V}_{\text{feed}} \)

\[ \alpha = NN(\rho_p, T_{\text{des}}, p_{\text{vac}}, \dot{V}_{\text{feed}}) \]

The training, testing and validation results can be found in Figure S3.

1.6 Isosteric heat of adsorption

The isosteric heat of adsorption (\( \Delta H_{\text{ads}} \)) represents the strength of the adsorbate-adsorbent interaction and is defined as the difference between the activation energy for adsorption and desorption. It can be calculated from the Clausius-Clapeyron [1] equation

\[ S4 \]
\[
\left( \frac{\partial (\ln p_{\text{CO}_2})}{\partial T} \right)_{\text{qeq}} = -\frac{\Delta H_{\text{ads}}}{RT^2}
\]  

(S8)

where \( p_{\text{CO}_2} \) is the partial pressure of CO\(_2\) (Pa), \( T \) is the absolute temperature (K) and \( R \) is the universal gas constant.

1.7 Equations 0D model
Table S5: Equations 0D model (first part).

**Blow-down step**

Known: \( q_{0\text{CO}_2}, q_{0\text{H}_2\text{O}}, q_{0\text{CO}_2}, q_{0\text{H}_2\text{O}}, y_{\text{N}_2}, p \)

Calculated: \( N_{\text{out}}, y_{\text{CO}_2}, y_{\text{H}_2\text{O}}, y_{\text{N}_2}, T^k \)

\[
\begin{align*}
\left( m_s q_{\text{CO}_2}^{k-1} + \frac{y_{\text{CO}_2}^{k-1} p^{k-1} V_{\epsilon}}{RT^{k-1}} \right) & - y_{\text{CO}_2}^{k} N_{\text{out}}^{k} - \left( m_s q_{\text{CO}_2}^k + \frac{y_{\text{CO}_2}^k p^k V_{\epsilon}}{RT^k} \right) = 0 \\
\left( m_s q_{\text{H}_2\text{O}}^{k-1} + \frac{y_{\text{H}_2\text{O}}^{k-1} p^{k-1} V_{\epsilon}}{RT^{k-1}} \right) & - y_{\text{H}_2\text{O}}^{k} N_{\text{out}}^{k} - \left( m_s q_{\text{H}_2\text{O}}^k + \frac{y_{\text{H}_2\text{O}}^k p^k V_{\epsilon}}{RT^k} \right) = 0
\end{align*}
\] (S9)

\[
\left[ \left( m_s q_{\text{CO}_2}^{k-1} + \frac{y_{\text{CO}_2}^{k-1} p^{k-1} V_{\epsilon}}{RT^{k-1}} \right) + \left( m_s q_{\text{H}_2\text{O}}^{k-1} + \frac{y_{\text{H}_2\text{O}}^{k-1} p^{k-1} V_{\epsilon}}{RT^{k-1}} \right) \right] - N_{\text{out}}^{k} - \left[ \left( m_s q_{\text{CO}_2}^k + \frac{y_{\text{CO}_2}^k p^k V_{\epsilon}}{RT^k} \right) + \left( m_s q_{\text{H}_2\text{O}}^k + \frac{y_{\text{H}_2\text{O}}^k p^k V_{\epsilon}}{RT^k} \right) \right] = 0
\] (S10)

\[
m_s \rho_p \left( T^k - T^{k-1} \right) - \Delta H_{s, \text{CO}_2}^{k-1} \left( m_s q_{\text{CO}_2}^{k-1} - m_s q_{\text{CO}_2}^k \right) - \Delta H_{s, \text{H}_2\text{O}}^{k-1} \left( m_s q_{\text{H}_2\text{O}}^{k-1} - m_s q_{\text{H}_2\text{O}}^k \right) = 0
\] (S11)

\[
1 - \frac{k}{y_{\text{CO}_2}^k - y_{\text{H}_2\text{O}}^k - y_{\text{N}_2}^k = 0}
\] (S12)

**Heating step**

Known: \( q_{0\text{CO}_2}, q_{0\text{H}_2\text{O}}, q_{0\text{CO}_2}, q_{0\text{H}_2\text{O}}, y_{\text{N}_2}, p, T \)

Calculated: \( N_{\text{out}}, y_{\text{CO}_2}, y_{\text{H}_2\text{O}}, y_{\text{N}_2}, Q_{\text{ex}} \)

\[
\begin{align*}
\left( m_s q_{\text{CO}_2}^{k-1} + \frac{y_{\text{CO}_2}^{k-1} p^{k-1} V_{\epsilon}}{RT^{k-1}} \right) & - y_{\text{CO}_2}^{k} N_{\text{out}}^{k} - \left( m_s q_{\text{CO}_2}^k + \frac{y_{\text{CO}_2}^k p^k V_{\epsilon}}{RT^k} \right) = 0 \\
\left( m_s q_{\text{H}_2\text{O}}^{k-1} + \frac{y_{\text{H}_2\text{O}}^{k-1} p^{k-1} V_{\epsilon}}{RT^{k-1}} \right) & - y_{\text{H}_2\text{O}}^{k} N_{\text{out}}^{k} - \left( m_s q_{\text{H}_2\text{O}}^k + \frac{y_{\text{H}_2\text{O}}^k p^k V_{\epsilon}}{RT^k} \right) = 0
\end{align*}
\] (S13)

\[
\left[ \left( m_s q_{\text{CO}_2}^{k-1} + \frac{y_{\text{CO}_2}^{k-1} p^{k-1} V_{\epsilon}}{RT^{k-1}} \right) + \left( m_s q_{\text{H}_2\text{O}}^{k-1} + \frac{y_{\text{H}_2\text{O}}^{k-1} p^{k-1} V_{\epsilon}}{RT^{k-1}} \right) \right] - N_{\text{out}}^{k} - \left[ \left( m_s q_{\text{CO}_2}^k + \frac{y_{\text{CO}_2}^k p^k V_{\epsilon}}{RT^k} \right) + \left( m_s q_{\text{H}_2\text{O}}^k + \frac{y_{\text{H}_2\text{O}}^k p^k V_{\epsilon}}{RT^k} \right) \right] = 0
\] (S14)

\[
1 - \frac{k}{y_{\text{CO}_2}^k - y_{\text{H}_2\text{O}}^k - y_{\text{N}_2}^k = 0}
\] (S15)

\[
Q_{\text{ex}}^{k} - m_s \rho_p \left( T^k - T^{k-1} \right) + \Delta H_{s, \text{CO}_2}^{k-1} \left( m_s q_{\text{CO}_2}^{k-1} - m_s q_{\text{CO}_2}^k \right) + \Delta H_{s, \text{H}_2\text{O}}^{k-1} \left( m_s q_{\text{H}_2\text{O}}^{k-1} - m_s q_{\text{H}_2\text{O}}^k \right) = 0
\] (S16)
Table S6: Equations 0D model (second part).

Cooling step

Known: $q_{CO_2}^0, q_{H_2O}^0, y_{CO_2}^0, y_{H_2O}^0, y_{N_2}^0, p, T$

Calculated: $N_m, y_{CO_2}^k, y_{H_2O}^k, y_{N_2}^k, Q_{cool}$

$$
\begin{align*}
\left(m_s q_{CO_2}^{k-1} + \frac{y_{CO_2}^{k-1} p V_\epsilon}{RT^{k-1}}\right) + y_{CO_2}^k N_{in} - \left(m_s q_{CO_2}^k + \frac{y_{CO_2}^k p V_\epsilon}{RT^k}\right) &= 0 \quad (S19) \\
\left(m_s q_{H_2O}^{k-1} + \frac{y_{H_2O}^{k-1} p V_\epsilon}{RT^{k-1}}\right) + y_{H_2O}^k N_{in} - \left(m_s q_{H_2O}^k + \frac{y_{H_2O}^k p V_\epsilon}{RT^k}\right) &= 0 \quad (S20)
\end{align*}
$$

$$
\left[ \left(m_s q_{CO_2}^{k-1} + \frac{y_{CO_2}^{k-1} p V_\epsilon}{RT^{k-1}}\right) + \left(m_s q_{H_2O}^{k-1} + \frac{y_{H_2O}^{k-1} p V_\epsilon}{RT^{k-1}}\right) \right] + N_{in} - \left[ \left(m_s q_{CO_2}^k + \frac{y_{CO_2}^k p V_\epsilon}{RT^k}\right) + \left(m_s q_{H_2O}^k + \frac{y_{H_2O}^k p V_\epsilon}{RT^k}\right) \right] = 0 \quad (S21)
$$

$$
1 - y_{CO_2}^k - y_{H_2O}^k - y_{N_2}^k = 0 \quad (S22)
$$

$$
Q_{cool} - m_s c_p \left(T^k - T^{k-1}\right) + \Delta H_{s, CO_2} (m_s q_{CO_2}^k - m_s q_{CO_2}^{k-1}) + \Delta H_{s, H_2O} (m_s q_{H_2O}^k - m_s q_{H_2O}^{k-1}) = 0 \quad (S23)
$$

Adsorption step

Sub-step 1

Known: $q_{CO_2}^{cool}, q_{H_2O}^{cool}, y_{CO_2}^{cool}, y_{H_2O}^{cool}, p, T$

Calculated: $N_{waste}, N_{feed}, y_{H_2O}, y_{N_2}$

$$
\begin{align*}
\left(m_s q_{CO_2}^{cool} + \frac{y_{CO_2}^{cool} p V_\epsilon}{RT_{amb}}\right) + y_{CO_2}^{cool} N_{feed} - \left(m_s q_{CO_2} + \frac{y_{CO_2} p V_\epsilon}{RT_{amb}}\right) - y_{CO_2} N_{waste} &= 0 \quad (S24) \\
\left(m_s q_{H_2O}^{cool} + \frac{y_{H_2O}^{cool} p V_\epsilon}{RT_{amb}}\right) + y_{H_2O}^{cool} N_{feed} - \left(m_s q_{H_2O} + \frac{y_{H_2O} p V_\epsilon}{RT_{amb}}\right) - y_{H_2O} N_{waste} &= 0 \quad (S25)
\end{align*}
$$

$$
\left(y_{N_2}^{cool} p V_\epsilon + y_{N_2}^{cool} N_{feed} - y_{N_2} p V_\epsilon - y_{N_2} N_{waste}\right) = 0 \quad (S26)
$$

$$
1 - y_{CO_2}^k - y_{H_2O} - y_{N_2} = 0 \quad (S27)
$$
Table S7: Equations 0D model (third part).

| Sub-step 2 |
|------------|
| **Known:** \( q_{\text{CO}_2} \), \( y_{\text{CO}_2}^{\text{sat}} \), \( y_{\text{H}_2\text{O}} \), \( y_{N_2}^1 \), \( p, T \) |
| **Calculated:** \( N_{\text{waste}} \), \( N_{\text{feed}} \), \( y_{N_2}^2 \) |

\[
\left( m_s q_{\text{CO}_2} + \frac{y_{\text{CO}_2}^{\text{sat}} p_{\text{amb}} V \epsilon}{RT_{\text{amb}}^s} \right) + \frac{y_{\text{CO}_2}^{\text{feed}} N_{\text{feed}} - \left( m_s q_{\text{CO}_2} + \frac{y_{\text{CO}_2}^{\text{feed}} p_{\text{amb}} V \epsilon}{RT_{\text{amb}}^s} \right) - y_{\text{CO}_2} N_{\text{waste}}}{y_{\text{CO}_2} N_{\text{waste}}} = 0 \quad (S28) \\
\frac{y_{N_2}^{\text{feed}} p_{\text{amb}} V \epsilon}{RT_{\text{amb}}^s} + \frac{y_{N_2}^{\text{feed}} N_{\text{feed}} - \left( \frac{y_{N_2}^{\text{feed}} p_{\text{amb}} V \epsilon}{RT_{\text{amb}}^s} \right) - y_{N_2} N_{\text{waste}}}{y_{N_2} N_{\text{waste}}} = 0 \quad (S29) \\
1 - y_{\text{CO}_2}^{\text{feed}} - y_{\text{H}_2\text{O}} - y_{N_2}^1 = 0 \quad (S30) 
\]
1.8 Details 1D model

Table S8: Equations for 1D adsorption model [2]

Total mass balance

\[ \epsilon_t \frac{\partial ( \rho p T )}{\partial t} + \frac{\partial ( \rho p u T )}{\partial x} + \rho_b R \sum_{j=1}^{N} \frac{\partial n_j}{\partial t} = 0 \quad (S31) \]

Component mass balance

\[ \epsilon \frac{\partial ( \rho_b u p y_i p T )}{\partial t} + \rho_b u \frac{\partial n_i}{\partial x} + \rho_b R \frac{\partial n_i}{\partial t} = 0 \quad i = 1, ..., N \quad (S32) \]

Mass transfer (Linear driving force model)

\[ \frac{\partial n_i}{\partial t} = k_i (n_i^e - n_i) \quad i = 1, ..., N \quad (S33) \]

Energy balance for the fixed bed

\[ (\epsilon_t C_g + \rho_b C_s + \rho_b C_{ads}) \frac{\partial p}{\partial t} + \rho_b C_g \frac{\partial T}{\partial x} - \rho_b \sum_{j=1}^{N} (-\Delta H_j) \frac{\partial n_j}{\partial t} + 4h_{wall-bed} \left( T - T_W \right) = 0 \quad (S34) \]

Energy balance for the bed wall

\[ \frac{\partial T_W}{\partial t} = \frac{4}{C_{p,W} (d^2_0 - d^2_i)} \left( d_i h_{wall-bed} (T - T_W) + d_i h_{fluid-wall} (T_{fluid} - T_w) \right) \quad (S35) \]

Momentum balance (Ergun equation)

\[ \frac{\partial p}{\partial x} = -150 \mu \left( 1 - \epsilon_b \right)^2 \frac{u}{\epsilon_b d^2_p} - \frac{1.75 (1 - \epsilon_b) \rho}{\epsilon_b^2 d^2_p} |u| \quad u \quad (S36) \]

The decision variables for the detailed process simulation are listed in Table S9.
Table S9: Design variables for optimizing the process, including lower and upper bound. The upper bound for the air volume stream is calculated using the minimum fluidization velocity [3]. For the 0D model only $T_{\text{des}}$, $p_{\text{vac}}$ and $V_{\text{air}}$ are considered as decision variables.

| Variable     | Range                        |
|--------------|------------------------------|
| $t_{\text{ads}}$ (s) | 100-10000                   |
| $t_{\text{des}}$ (s)  | 50-2200                     |
| $t_{\text{purge}}$ (s) | 1-400                      |
| $p_{\text{vac}}$ (bar) | 0.1-0.9                    |
| $T_{\text{des}}$ (K)  | 363-373                     |
| $\Delta T_{\text{purge}}$ (K) | 1-60                  |
| $V_{\text{air}}$ (m$^3$/s) | $2.0 \times 10^{-7}$ - $2.0 \times 10^{-5}$ |

Details regarding the modeling of the solid sorbent process are shown in Table S10.

Table S10: Parameters for modeling the adsorption unit using the 1D model.

| Parameter                          | Value                        | Source            |
|------------------------------------|------------------------------|-------------------|
| Plate containing sorbent           |                              |                   |
| Length                             | 0.05 m                       |                   |
| Internal radius                    | 0.005 m                      |                   |
| External radius                    | 0.005001 m                   |                   |
| Heat capacity wall                 | $2.457 \times 10^6$ J/(K m$^3$) |                   |
| Heat transfer fluid/wall/bed       | $6.7$ W/(m$^2$K)             | Modeled using [4] |
| Contactor geometry                 |                              |                   |
| LxWxH                              | $1.5 \times 1.5 \times 1.5$ m | [4]               |
| Void fraction, $\epsilon = 1 - V_{\text{sorbent}}/V_{\text{aircontator}}$ | 0.6                          | [4]               |

2 Validation

Table S11: Different combinations for CO$_2$ and H$_2$O isotherms.

| CO$_2$ isotherm | H$_2$O isotherm |
|----------------|-----------------|
| APDES-NFC      | s1: A-A         |
| Exemplary      | s2: E-A         |
| MIL-101(Cr)-PEI-800 | s5: MP-A  |
| Lewatit VP OC 106 | s6: MP-M  |
|                | s3: E-M         |
|                | s4: E-L         |
|                | s7: MP-L        |
|                | s8: L-L         |
**Figure S4**: Composition of CO$_2$ and H$_2$O over the pressure for the whole cycle for case s2/E-A (a) and b), and Cr-MIL(101) (c) and d)).

**Figure S5**: Profiles of the adsorbed amount during one cycle, corresponding to Figure 4 in the main text. a) shows case s2/E-A and b) Cr-MIL(101).
2.1 Parity plots varying $\text{CO}_2$ concentration
Figure S6: Parity plot for $y_{\text{CO}_2} = 0.1\%$.

Figure S7: Parity plot for $y_{\text{CO}_2} = 1.0\%$. 

S13
The range of the resulting purity and recovery for both the 0D and the 1D model are shown in Figure S8.

Figure S8: a) Purity and b) capture rate from optimizing the materials during the validation for the 0D and 1D model.
3 Screening

Table S12: Equations of the different isotherm models.

| Model       | Equation                                                                 |
|-------------|--------------------------------------------------------------------------|
| Toth-cp     | \[ q_{CO_2} = n_s \cdot \frac{b_{y_{CO_2}}}{\left(1 + b_{y_{CO_2}}\right)^{1/t_c}} + n_s \cdot \frac{b_{y_{CO_2}}}{\left(1 + b_{y_{CO_2}}\right)^{1/t_p}} \] |
|             | \[ n_{s,c/p} = n_{a,c/p} \cdot \exp \left( X_{l,c/p} \cdot \left(1 - \frac{T}{T_0}\right) \right) \] |
|             | \[ b_{c/p} = b_{0,c/p} \cdot \exp \left( \frac{\Delta H_{c/p}}{R T_0} \right) \cdot \left( \frac{T}{T_0} - 1 \right) \] |
| DSL         | \[ q_{CO_2} = n_1 \cdot \frac{b_{y_{CO_2}}}{\left(1 + b_{y_{CO_2}}\right)^{1/d_c}} + n_2 \cdot \frac{d_{y_{CO_2}}}{\left(1 + d_{y_{CO_2}}\right)^{1/t_p}} \] |
| Toth        | \[ q_{CO_2} = n_s \cdot \frac{b_{y_{CO_2}}}{\left(1 + b_{y_{CO_2}}\right)^{1/t_l}} \] |
|             | \[ n_s = n_{a_0} \cdot \exp \left( X_l \cdot \left(1 - \frac{T}{T_0}\right) \right) \] |
|             | \[ b = b_0 \cdot \exp \left( \frac{\Delta H_{c}}{R T_0} \right) \cdot \left( \frac{T}{T_0} - 1 \right) \] |
| Langmuir-Freundlich | \[ q_{CO_2} = n_s \cdot \frac{b_{y_{CO_2}}}{\left(1 + b_{y_{CO_2}}\right)^{1/t_l}} \] |
|             | \[ n_s = n_{a_0} \cdot \exp \left( X_l \cdot \left(1 - \frac{T}{T_0}\right) \right) \] |
|             | \[ b = b_0 \cdot \exp \left( \frac{\Delta H_{c}}{R T_0} \right) \cdot \left( \frac{T}{T_0} - 1 \right) \] |
| S-shaped    | \[ q_{CO_2} = q_L \cdot \left(1 - w\right) + q_V \cdot w \] |
|             | \[ q_L = \frac{q_L \cdot b_L(y_{CO_2})}{(1+b_L(y_{CO_2}))} \] |
|             | \[ b_L/U/H = b_{L/U/H,0} \cdot \exp \left( \frac{\Delta U_{L/U/H}}{R T_0} \right) \] |
|             | \[ w = \exp \left( \frac{-\log(y_{CO_2}) - \log(p_{step})}{\sigma} \right) \cdot \left(1 + \exp \left( \frac{\log(y_{CO_2}) - \log(p_{step})}{\sigma} \right) \right) \] |
|             | \[ p_{step} = p_{step,0} \cdot \exp \left( \frac{-\Delta H_{step}}{R T_0} \left(1 - \frac{T}{T_0}\right) \right) \] |
|             | \[ \sigma = X_{i1} \cdot \exp \left( X_{i2} \left(\frac{T}{T_0} - 1\right) \right) \] |
|             | \[ q_V = \frac{q_{L,U} \cdot b_L(y_{CO_2})}{1+b_L(y_{CO_2})} + b_H(y_{CO_2}) \] |

The NIST/ARPA-E database, the database of novel and emerging adsorbents by the National Institute of Standards and Technology (NIST) includes data from published studies, both real and hypothetical. Since the database includes thousands of isotherm data files, not only for CO$_2$ adsorption, but also many other gases, it is important to first filter the materials. We use a script written in MATLAB to run through the materials filter the suitable adsorbents. The approach is graphically shown in Figure S9. During the filtering process, we are also important to first filter the materials. We use a script written in MATLAB to run through the materials filter the database includes thousands of isotherm data files, not only for CO$_2$ Standards and Technology (NIST) includes data from published studies, both real and hypothetical. Since the database includes thousands of isotherm data files, not only for CO$_2$ adsorption, but also many other gases, it is important to first filter the materials. We use a script written in MATLAB to run through the materials filter the suitable adsorbents. The approach is graphically shown in Figure S9. During the filtering process, we are also excluding all data sets for which only one temperature is available. This is necessary for the following step, where the remaining isotherms are fitted by temperature dependent isotherm models. The fitting process itself includes several steps, which are shown in the flowchart in Figure S10. For the objective function during the fitting, the normalized standard deviation was applied, which is commonly used to fit isotherm models to experimental data [4–7]. It includes the adsorbed amount determined experimentally $q_{exp}$, the amount adsorbed as predicted by the model $q_{fit}$ and the total number of experimental points $N$, and is calculated in the form of

$$\text{err} = \sqrt{\frac{\sum \left[ (q_{fit} - q_{exp})^2 \right]}{N - 1}} \cdot 100$$  \hspace{1cm} (S37)

which is minimized using the inbuilt Matlab optimization routine fmincon together with the sequential quadratic programming (SQP) algorithm.
Figure S9: Flowchart describing the filtering process of the adsorbents from the NIST/ARPA-E database.
Input data from database (qCO2 and p)

Fitting using Toth-cp model

Fitting using Langmuir-Freundlich model

$R^2_{Toth-cp}$ or $R^2_{Langm-fr} < 2\%$

Choose fitting with Toth-cp model

Choose fitting with Langmuir-Freundlich model

$r^2_{s-shaped} > R^2_{Toth-cp}$ or $R^2_{Langm-fr}$

Choose fitting with s-shaped model

$r^2_{Toth-cp} < R^2_{Langm-fr}$

Choose fitting with Toth-cp model

$r^2_{Toth-cp} < 0.8$

Exclude this fitting

$R^2_{Toth-cp} < 0.8$

Save the fitting parameters

Figure S10: Flowchart fitting.
The error is expressed by

$$R^2 = \frac{\sum (q_{\text{exp}} - q_{\text{fit}})^2}{\sum (q_{\text{exp}} - \bar{q}_{\text{exp}})^2}$$  \hspace{1cm} (S38)

with $\bar{q}_{\text{exp}}$ being the mean of the experimental data.
Table S13: Resulting ranking showing the 10 best performing materials for $\gamma_{\text{CO}_2} = 0.4\%$. The values of the maximum productivity and the corresponding thermal energy consumption are given for the best performing adsorbent. For the remaining materials, the deviation to the best performing one is given in percentage.

| Adsorbent            | Pr    | $Q_{\text{th}}$ |
|----------------------|-------|-----------------|
| Cr-MIL(101)          | 20.6 kg/m$^3$/h | 14.4 MJ/kg     |
| MIL-101              | -5%   | -4%             |
| CuBTC                | -24%  | +9%             |
| MIL-53(Al)           | -10%  | +42%            |
| Zn-DABCO             | -24%  | +49%            |
| MIL-101(Cr)-PEI-800  | -66%  | -11%            |
| Lewatit              | -74%  | +9%             |
| Exemplary            | -77%  | +18%            |
| Zeolite Na-LSX       | -21%  | +61%            |
| Ca-X                 | -27%  | +52%            |

Table S14: Resulting ranking showing the 10 best performing materials for $\gamma_{\text{CO}_2} = 0.1\%$. The values of the maximum productivity and the corresponding thermal energy consumption are given for the best performing adsorbent. For the remaining materials, the deviation to the best performing one is given in percentage.

| Adsorbent            | Pr    | $Q_{\text{th}}$ |
|----------------------|-------|-----------------|
| PCN-11               | 43.8 kg/m$^3$/h | 14.9 MJ/kg     |
| MIL-101              | -8%   | -2%             |
| Cr-MIL(101)          | 0%    | +33%            |
| Ca-X                 | -25%  | +12%            |
| Zeolite Na-LSX       | -32%  | +32%            |
| Cu-BTC powder        | -31%  | +52%            |
| CuBTC                | -29%  | +54%            |
| MIL-53(Al)           | -15%  | +96%            |
| Zn-DABCO             | -26%  | +53%            |
| MIL-101(Cr)-PEI-800  | -71%  | -30%            |
Table S15: Resulting ranking showing the 10 best performing materials for y\textsubscript{CO}_2 = 1.0%. The values of the maximum productivity and the corresponding thermal energy consumption are given for the best performing adsorbent. For the remaining materials, the deviation to the best performing one is given in percentage.

| Adsorbent                  | Pr    | Q\textsubscript{th}   |
|----------------------------|-------|-----------------------|
| Mg-MOF-74                  | 207.7 kg/m\textsuperscript{3}/h | 4.72 MJ/kg     |
| Ca-X                       | -32%  | +68%                  |
| MIL-101(Cr)-250nm-PEI-300  | -36%  | +88%                  |
| Zeolite Na-LSX              | -47%  | +118%                 |
| Zeolite 13X                | -49%  | +106%                 |
| PCN-11                     | -41%  | +205%                 |
| MIL-101                    | -52%  | +206%                 |
| Carbon                     | -54%  | +139%                 |
| Cr-MIL(101)                | -43%  | +318%                 |
| Mg-X                       | -60%  | +164%                 |

Figure S11: a) Purity and b) recovery for resulting materials from screening with 0.04% CO\textsubscript{2} in the feed.
Figure S12: a) Purity and b) recovery for resulting materials from screening with 0.1% CO₂ in the feed.

Figure S13: a) Purity and b) recovery for resulting materials from screening with 1.0% CO₂ in the feed.
Figure S14: Resulting objectives humid using Lewatit H₂O isotherm for a) 0.04%, b) 0.1% and c) 1.0% CO₂ in the feed.

Figure S15: Resulting objectives with dry feed for a) 0.04%, b) 0.1% and c) 1.0% CO₂ in the feed.
The following three Tables S16, S17, and S18 list the resulting materials from the screening with different CO₂ concentrations for the case with the APDES-NFC isotherm for water.

**Table S16:** Resulting materials with their DOI from screening with \( \gamma_{CO_2} = 40Pa \) and the APDES-NFC isotherm for the water.

| Material         | DOI                           |
|------------------|-------------------------------|
| Ca-X             | 10.1039/c2ee23337a            |
| Carbon           | 10.1021/la4004998             |
| APDES-NFC        | 10.1021/es404430g             |
| Cr-MIL(101)      | 10.1039/C2ra20641b            |
| CuBTC            | 10.1016/j.cej.2015.07.020    |
| Exemplary        | 10.1016/j.joule.2021.05.023  |
| Lewatit          | 10.1016/j.ijggc.2015.07.014  |
| MIL-101(Cr)-PEI-800 | 10.1021/acssuschemeng.6b01692 |
| MIL-101          | 10.1021/jp8111418r            |
| MIL-53(Al)       | 10.1021/ie5006146             |
| Zeolite Na-LSX   | 10.1016/S0167-2991(02)80202-6 |
| Zn-DABCO         | 10.1016/j.seppur.2011.09.041 |
Table S17: Resulting materials with their DOI from screening with $y_{CO_2} = 100 Pa$ and the APDES-NFC isotherm for the water.

| Material               | DOI                           |
|------------------------|-------------------------------|
| AC-Chitosan            | 10.1016/j.physe.2013.10.024  |
| Ca-X                   | 10.1039/t2ee23337a            |
| Carbon                 | 10.1021/la4004998             |
| Cr-MIL(101)            | 10.1039/C2ra20641b            |
| CuBTC                  | 10.1016/jcej.2015.07.020     |
| Exemplary              | 10.1016/j.joule.2021.05.023  |
| Lewatit                | 10.1016/j.ijgge.2015.07.014  |
| MIL-101(Cr)-PEI-800    | 10.1021/acssuschemeng.6b01692|
| MIL-101                | 10.1021/Jp811418r             |
| MIL-101(Cr)-150nm-PEI-1800 | 10.1039/C4ta01174k         |
| MIL-53(Al)             | 10.1021/fe5006146             |
| PCN-11                 | 10.1039/C0ee00700e            |
| Zeolite Na-LSX         | 10.1016/S0167-2991(02)80202-6|
| Zn-DABCO               | 10.1016/j.seppur.2011.09.041|
Table S18: Resulting materials with their DOI from screening with $y_{CO_2} = 1000 Pa$ and the APDES-NFC isotherm for the water.

| Material                              | DOI                          |
|---------------------------------------|------------------------------|
| AC-Chitosan                           | 10.1016/j.physe.2013.10.024 |
| Activated Carbon-600-0.5              | 10.1039/C2cp44436d          |
| Ca-X                                  | 10.1039/c2ee23337a          |
| Carbon                                | 10.1021/ia4004998          |
| Carbon KNC-A-K                        | 10.1021/Cm303072n          |
| Carbon SBA-NC                         | 10.1021/Cm303072n          |
| Cr-MIL(101)                           | 10.1039/C2ra20641b          |
| CuBTC                                 | 10.1016/j.cej.2015.07.020  |
| Exemplary                             | 10.1016/j.joule.2021.05.023|
| Graphite Oxide Cu-BTC (3rd cycle)    | 10.1021/am404952z          |
| Graphite Oxide/Urea Cu-BTC 2 (4th cycle) | 10.1021/am404952z    |
| Lewatit                               | 10.1016/j.ijgge.2015.07.014|
| MIL-101(Cr)-PEI-800                  | 10.1021/acssuschemeng.6b01692|
| MIL-101                               | 10.1021/jp811418r          |
| MIL-101(Cr)-150nm-PEI-1800            | 10.1039/C4ta01174k         |
| MIL-101(Cr)-250nm-PEI-300             | 10.1039/C4ta01174k         |
| MIL-53(Al)                            | 10.1021/je5006146          |
| M-DNL-6                               | 10.1002/cssc.201200907     |
| Mg-MOF-74                             | 10.1016/j.ijgge.2013.01.009|
| Mg-X                                  | 10.1039/c2ee23337a         |
| PCN-11                                | 10.1039/C0ee00700e         |
| SAPO-34 zeolite                       | 10.1080/01496395.2013.812118|
| SNS52-20                              | 10.1039/c5ta01776a         |
| UTSA-16-GO (19 mg)                    | 10.1039/c4ta04770b         |
| UiO-66-EA                             | 10.1039/c5ta05997f         |
| Zeolite 13X                           | 10.1021/Je800900a         |
| Zeolite Na-LSX                        | 10.1016/S0167-              |
|                                       | 2991(02)80202-6            |
| Zn-DABCO                              | 10.1016/j.seppur.2011.09.041|
**Nomenclature**

\( \rho \)  
Density, kg/m\(^3\)  

\( \alpha \)  
Saturation level  

\( \Delta H_{\text{ads}} \)  
Isosteric heat of adsorption, J/mol  

\( \Delta H_{\text{ads}} \)  
Isosteric heat of adsorption, kJ/mol  

\( \dot{Q} \)  
Specific heat, kJ/kg  

\( \dot{V}_{\text{feed}} \)  
Feed volume stream, m\(^3\)/s  

\( \epsilon \)  
Void fraction  

\( \mu \)  
Dynamic viscosity, Pas  

\( \Phi \)  
Purity  

\( c_p \)  
Specific heat capacity at constant pressure, kJ/kg/K  

\( C_{\text{ads}} \)  
Heat capacity of the adsorbed phase, J/(Kkg)  

\( C_g \)  
Heat capacity of the gas, J/(Km\(^3\))  

\( C_s \)  
Heat capacity of the solid, J/(Kkg)  

\( d_p \)  
Particle diameter, m  

\( e^{\text{th}} \)  
Specific thermal energy, kJ/kg\(\text{CO}_2\)  

\( h \)  
Heat transfer coefficient, J/(m\(^2\)sK)  

\( k \)  
Overall mass transfer coefficient, 1/s  

\( N_{\text{grid}} \)  
Number of grid points  

\( r \)  
Capture rate  

\( u_{\text{air}} \)  
Air velocity, m/s  

\( N \)  
Amount of moles, mol  

\( R^2 \)  
Error  

a,b,c,d,e,f,g,h  
Fitting parameters  

ads  
Adsorption  

amb  
Ambient  

BD  
Blow-down  

des  
Desorption  

err  
Error  

exp  
Experimental
f fluid, final
fit fitted
i component
m mass, kg
MM Molar mass, g/mol
p pressure, bar
Pr Productivity, kg/m³/h
q solid phase concentration, mol/kg
q* equilibrium adsorbed amount, mol/kg
R universal gas constant, J/mol/K
s solid, sorbent
T temperature, K
t time, s
Vc column volume, m³
vac vacuum
y mole fraction

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