Carnegie Mellon University

Carbon Capture through Membranes - Leveraging Multiphysics Modeling, Dimensional Analysis and Machine Learning to Scale up and Optimize Devices and Processes for Decarbonization

Grigorios Panagakos

NAMS, Annual Fall Meeting
Santa Fe, NM, 2024
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Motivation

Problem:
• Pollution, Climate Change
• Industrial CO$_2$ emissions
  • 9.2 Gt out of 36.8 Gt$^1$(2022)

Solution:
• New Technologies
• $2.52$ billion in carbon management programs by DOE Feb 2023$^2$

Goal: Net 0 emission by 2050 to limit average global T to increase by 1.5 deg C$^3$


Membrane Technology: A Lower-Cost Alternative

<table>
<thead>
<tr>
<th>Technology Readiness level (TRL)</th>
<th>Solvent-absorption (e.g., amine scrubbing)</th>
<th>Membrane</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mature</td>
<td></td>
<td>Early adoption</td>
</tr>
</tbody>
</table>

Pros & Cons

- Equipment corrosion
- Massive energy required for solvent regeneration
- High capture rate

- Simple operation
- Low maintenance
- Modularity
- **Small footprint**
- Low Capture rate
- Membrane fouling
Permeance (in Gas Permeation Unit (GPU)) is pressure normalized flux. Permeability (in Barrer) is a material property independent of thickness.

**Permeance** = \( \frac{\text{Permeability (P) of selective material}}{\text{thickness of selective layer}} \)

**Selectivity** (\( > 25 \)) = \( \frac{P(\text{CO}_2)}{P(\text{N}_2)} \)

Thin Film Composite (TFC) membrane development

**Selective layer** (\(< 1 \text{ um})\): \( \text{CO}_2/\text{N}_2 \) separation

**Gutter layer** (\(< 1 \text{ um})\): preventing pore penetration and smoothening porous support

**Porous support** (\( > 20 \text{ µm})\): mechanical reinforcement
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Membrane module

Experimental setup
Model overview

Physics of diffusion on membrane boundary

\[ j_i = \frac{\text{kg}}{m^2 \cdot s} \]

\[ |J_i| = Q_i \cdot M_i \cdot (p_1 \cdot x_{i\text{feed}} - p_2 \cdot x_{i\text{perm}}) \]

Physics of Transport in feed side and Permeate

Navier-Stokes + Convection-diffusion equation

Coupled physics: mass transfer in laminar flow
Model validated using experimental results

1600 CO$_2$ GPU
28 CO$_2$/N$_2$ Selectivity

Feed concentration:
30% CO$_2$, 70% N$_2$

P$_{\text{feed}}$ = 1.21 bar
P$_{\text{perm}}$ = 0.2 bar

max error: 2.33%
max error ret: 2.73%
perm: 4.5%
max error: 5.82%
Model is validated on different membrane

3755 CO₂ GPU
13 CO₂/N₂ Selectivity

Feed concentration:
30% CO₂, 70% N₂

$p_{\text{feed}} = 1.21$ bar
$p_{\text{perm}} = 0.2$ bar
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Designs to analyze

**Design 1A**
- 24 cm² per membrane sheet
- 1 membrane, total area: 24 cm²
- Membrane properties: 1600 GPU and 28 selectivity & 3200 GPU and 32 selectivity

**Design 1B**
- 24 cm² per membrane sheet
- 5 membranes, total area: 120 cm²
- Membrane properties: 1600 GPU and 28 selectivity & 3200 GPU and 32 selectivity

**Design 2A**
- 96 cm² per membrane sheet
- 2 membranes, total area: 192 cm²
- Membrane properties: 1600 GPU and 28 selectivity & 3000 GPU and 25 selectivity

**Design 2B**
- 96 cm² per membrane sheet
- 10 membranes, total area: 960 cm²
- Membrane properties: 1600 GPU and 28 selectivity & 3000 GPU and 25 selectivity
Boundary condition for binary mixtures \((CO_2, N_2)\)

\[
\text{Permeate side} \quad \text{Feed side}
\]

**Domain variables**
- \(N_{feed}^{feed}\): molar fraction in the feed side
- \(x_i^{feed}\), \(x_i^{perm}\): molar fractions of \(i\) in the feed and permeate sides
- \(P_{feed}\), \(P_{perm}\): feed and permeate pressures
- \(J_i\): molar flow through the membrane of component \(i\)

**Design variables**
- \(Q_i\): permeance of \(i\)
- \(L\): membrane length
- \(a\): membrane width
Separation equation for binary mixtures \((CO_2, N_2)\)

\[
-L \frac{d(N_{feed} x_{CO_2}^{feed})}{dz} = Q_{CO_2} A_M \left( p_{feed} x_{CO_2}^{feed} - p_{perm} x_{CO_2}^{perm} \right)
\]

\[
-L \frac{d(N_{feed})}{dz} = \sum_i Q_i A_M \left( p_{feed} x_i^{feed} - p_{perm} x_i^{perm} \right) = \eta
\]

\[
x_{CO_2}^{perm} = \frac{Q_{CO_2} A_M \left( p_{feed} x_{CO_2}^{feed} - p_{perm} x_{CO_2}^{perm} \right)}{\eta} = \frac{Q_{CO_2} A_M p_{feed} x_{CO_2}^{feed}}{\eta + Q_{CO_2} A_M p_{perm}}
\]

**Domain variables**
- \(N_{feed}\): molar fraction in the feed side
- \(x_{CO_2}^{feed}, x_i^{perm}\): molar fractions of \(i\) in the feed and permeate sides
- \(p_{feed}, p_{perm}\): feed and permeate pressures
- \(\eta\): total molar flow through the membrane

**Design variables**
- \(Q_i\): permeance of \(i\)
- \(L\): membrane length
- \(A_M\): area of the membrane
Separation equation for binary mixtures: CO\textsubscript{2} balance

\[-L \frac{d(N_{\text{feed}} \chi_{\text{CO}_2}^{\text{feed}})}{dz} = Q_{\text{CO}_2} A_M \left(p_{\text{feed}} \chi_{\text{CO}_2}^{\text{feed}} - p_{\text{perm}} \chi_{\text{CO}_2}^{\text{perm}}\right)\]

\[-L \left(x_{\text{CO}_2}^{\text{feed}} \frac{d(N_{\text{feed}})}{dz} + N_{\text{feed}} \frac{d(x_{\text{CO}_2}^{\text{feed}})}{dz}\right) = Q_{\text{CO}_2} A_M \left(p_{\text{feed}} x_{\text{CO}_2}^{\text{feed}} - p_{\text{perm}} \frac{Q_{\text{CO}_2} A_M p_{\text{feed}} x_{\text{CO}_2}^{\text{feed}}}{\eta + Q_{\text{CO}_2} A_M p_{\text{perm}}}\right)\]

\[-L \frac{1}{x_{\text{CO}_2}^{\text{feed}}} \frac{d(x_{\text{CO}_2}^{\text{feed}})}{dz} = \frac{Q_{\text{CO}_2} A_M p_{\text{feed}}}{N_{\text{feed}}} \left(1 - \frac{Q_{\text{CO}_2} A_M p_{\text{perm}}}{\eta + Q_{\text{CO}_2} A_M p_{\text{perm}}}\right) - \frac{\eta}{N_{\text{feed}}}\]

\[-L \frac{d \left(\ln(x_{\text{CO}_2}^{\text{feed}})\right)}{dz} = \frac{\eta}{N_{\text{feed}}} \left(\frac{Q_{\text{CO}_2} A_M p_{\text{feed}}}{\eta + Q_{\text{CO}_2} A_M p_{\text{perm}}} - 1\right)\]
Dimensionless separation equation for CO$_2$ balance

$$-L \frac{d\left(\ln(x_{CO_2}^{feed})\right)}{dz} = \frac{\eta}{N^{feed}} \left( \frac{Q_{CO_2}A_MP^{feed}}{\eta + Q_{CO_2}A_MP^{perm}} - 1 \right)$$

$$F^d = \frac{N^{inlet}}{Q_{CO_2}A_MP^{feed}}; \quad S^{cut} = 1 - \frac{N^{feed}}{N^{inlet}}$$

$$p^{ratio} = \frac{p^{feed}}{p^{perm}}; \quad \eta^* = \frac{\eta}{N^{inlet}} = \frac{dS^{cut}}{dz^*}$$

$$\frac{d\left(\ln(x_{CO_2}^{feed})\right)}{dz^*} = \frac{\eta^*}{1 - S^{cut}} \left( \frac{p^{ratio}}{1 + p^{ratio}\eta^*F^d} - 1 \right)$$

$$x_{CO_2}^{feed} \bigg|_{z=L} = f_1 \left( p^{ratio}, F^d, S^{cut} \right)$$

This is an output variable
Separation equation for feed flowrate

\[ \eta = Q_{CO_2} A_M P_{feed} \left[ \left( x_{CO_2}^{feed} - \frac{P_{perm}}{P_{feed}} x_{CO_2}^{perm} \right) + \frac{Q_{N_2}}{Q_{CO_2}} \left( (1 - x_{CO_2}^{feed}) - \frac{P_{perm}}{P_{perm}} (1 - x_{CO_2}^{perm}) \right) \right] \]

\[ x_{CO_2}^{perm} = \frac{Q_{CO_2} A_M P_{feed} x_{CO_2}^{feed}}{\eta + Q_{CO_2} A_M P_{perm}} \]

\[ \eta = Q_{CO_2} A_M P_{feed} \left[ \left( x_{CO_2}^{feed} - \frac{P_{perm}}{P_{feed}} \frac{Q_{CO_2} A_M P_{feed} x_{CO_2}^{feed}}{\eta + Q_{CO_2} A_M P_{perm}} \right) + \frac{Q_{N_2}}{Q_{CO_2}} \left( (1 - x_{CO_2}^{feed}) - \frac{P_{perm}}{P_{perm}} \left( 1 - \frac{Q_{CO_2} A_M P_{feed} x_{CO_2}^{feed}}{\eta + Q_{CO_2} A_M P_{perm}} \right) \right) \right] \]
Dimensionless separation equation for feed flowrate

\[ \eta = x_{CO_2}^{feed} Q_{CO_2} A_M^p_{feed} \left[ \left( 1 - \frac{Q_{N_2}}{Q_{CO_2}} \right) \left( 1 - \frac{p_{perm}}{p_{feed}} \eta + Q_{CO_2} A_M^p_{perm} \right) + \frac{Q_{N_2}}{x_{CO_2}^{feed} Q_{CO_2} (1 - \frac{p_{perm}}{p_{feed}})} \right] \]

\[ \eta^* = \frac{x_{CO_2}^{feed}}{F^d} \left( 1 - \frac{1}{Sel} \right) \left( 1 - \frac{1}{Pratio F^d \eta^* + 1} \right) + \frac{1}{F^d Sel} \left( 1 - \frac{1}{Pratio} \right) \]

Dimensionless form of the equation

\[ Sel: \text{selectivity } \frac{Q_{CO_2}}{Q_{N_2}} \]

\[ \eta^* = \frac{\eta}{N_{inlet}} = \frac{dS^{cut}}{dz^*} \]

\[ S^{cut} \bigg|_{z=L} = f_2 \left( Pratio, F^d, Sel, x_{CO_2}^{feed} \bigg|_{z=0} \right) \]
Dimensionless numbers for membrane separation

Pressure ratio

\[ \text{pratio} = \frac{p_{\text{feed}}}{p_{\text{perm}}} \]

Dimensionless feed flow

\[ F^d = \frac{N_{\text{inlet}}}{Q_{CO_2} A_M p_{\text{feed}}} \]

\[ F^d = \frac{b L U V_{CO_2}}{R \tau_{m} \tau_{s} \tau_{f}} \]

\[ \begin{align*}
\text{pratio} & : \text{inlet molar flow} \\
p_{\text{feed}} & : \text{feed pressure} \\
p_{\text{perm}} & : \text{permeate pressure} \\
Q_{CO_2} & : \text{permeance of CO}_2 \\
A_M & : \text{area of the membrane} \\
a & : \text{feed membrane width} \\
b & : \text{feed membrane height} \\
F^d & : \text{dimensionless feed flow} \\
L & : \text{membrane length} \\
N_{\text{inlet}} & : \text{inlet total molar flow} \\
R & : \text{gas superficial velocity} \\
V_{CO_2} & : \text{mass transfer coefficient (}Q_{CO_2}RT\text{)} \\
\tau_{m} & : \text{time scale of mass transfer} \\
\tau_{s} & : \text{time scale of fluid to exit the feed} \\
\end{align*} \]
Key points of equations for the simplified process

\[-L \frac{d(N^{feed}x_{CO_2}^{feed})}{dz} = Q_{CO_2}A_M (p^{feed}x_{CO_2}^{feed} - p^{perm}x_{CO_2}^{perm})\]

\[-L \frac{d(N^{feed})}{dz} = \sum_i Q_i A_M (p^{feed}x_{i}^{feed} - p^{perm}x_{i}^{perm}) = \eta\]

Then, the separation performance can be described by:

\[S_{cut}^{\|L} = f_1 \left( F^d, Pratio, Sel, x_{CO_2}^{feed} \right) ; \]

\[x_{CO_2}^{feed} \bigg|_{z=L} = f_2 \left( F^d, Pratio, Sel, x_{CO_2}^{feed} \right) ; \]

\[CO_2 \text{ recovery} = 1 - (1 - S_{cut}^{\|L}) \]

\[CO_2 \text{ purity} = \frac{x_{CO_2}^{feed} \bigg|_{z=0} - (1 - S_{cut}^{\|L})x_{CO_2}^{feed} \bigg|_{z=L}}{S_{cut}^{\|L}}\]

\(N^{feed}\): molar fraction in the feed

\(S_{cut}^{\|L}\): Stage-cut

\(x_{i}^{feed}, x_{i}^{perm}\): molar fractions of \(i\) in the feed and permeate sides

\(p^{feed}, p^{perm}\): feed and permeate pressures

\(Sel\): selectivity \(Q_{CO_2}/Q_{N_2}\)

\(A_M\): area of the membrane
## Consistency test using the Buckingham Pi theorem

<table>
<thead>
<tr>
<th>Variable</th>
<th>units</th>
<th>Type</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N_{inlet}$</td>
<td>mol/s</td>
<td>input</td>
</tr>
<tr>
<td>$p_{feed}$</td>
<td>kg/(s$^2$m)</td>
<td>input</td>
</tr>
<tr>
<td>$A_M$</td>
<td>m$^2$</td>
<td>input</td>
</tr>
<tr>
<td>$V_{CO_2}$</td>
<td>m/s</td>
<td>input</td>
</tr>
<tr>
<td>$V_{N_2}$</td>
<td>m/s</td>
<td>input</td>
</tr>
<tr>
<td>$R$</td>
<td>kg m$^2$/(s$^2$mol K)</td>
<td>constant</td>
</tr>
<tr>
<td>$T$</td>
<td>K</td>
<td>input</td>
</tr>
<tr>
<td>$p_{perm}$</td>
<td>kg/(s$^2$m)</td>
<td>input</td>
</tr>
<tr>
<td>$x_{CO_2}^{feed} \bigg</td>
<td>_{z=0}$</td>
<td>–</td>
</tr>
<tr>
<td>$N_{feed} \bigg</td>
<td>_{z=L}$</td>
<td>mol/s</td>
</tr>
<tr>
<td>$x_{CO_2}^{feed} \bigg</td>
<td>_{z=L}$</td>
<td>–</td>
</tr>
</tbody>
</table>

- 11 dimensional variables
- 5 base units
- Therefore, 6 dimensionless variables can be built
- 4 input dimensionless vars. and 2 output dimensionless vars.

- Input vars.: $F^d$, $P_{ratio}$, $Sel$, $x_{CO_2}^{feed} \bigg|_{z=0}$
- Output vars.: $CO_2$ recovery, $CO_2$ purity
### Designs to analyze

<table>
<thead>
<tr>
<th>Design</th>
<th>Details</th>
</tr>
</thead>
</table>
| **1A**   | • 24 cm² per membrane sheet  
          | • 1 membrane, total area: 24 cm²  
          | • Membrane properties: 1600 GPU and 28 selectivity & 3200 GPU and 32 selectivity |
| **1B**   | • 24 cm² per membrane sheet  
          | • 5 membranes, total area: 120 cm²  
          | • Membrane properties: 1600 GPU and 28 selectivity & 3200 GPU and 32 selectivity |
| **2A**   | • 96 cm² per membrane sheet  
          | • 2 membranes, total area: 192 cm²  
          | • Membrane properties: 1600 GPU and 28 selectivity & 3000 GPU and 25 selectivity |
| **2B**   | • 96 cm² per membrane sheet  
          | • 10 membranes, total area: 960 cm²  
          | • Membrane properties: 1600 GPU and 28 selectivity & 3000 GPU and 25 selectivity |
Dimensionless feed flow \( (F_d^d) \) to predict the separation performance

\[
F_d^d = \frac{\text{Total molar flow in the feed}}{\text{CO}_2 \text{ permeance} \times \text{Membrane area} \times \text{Pressure}}
\]

1600 GPU, 28 selectivity
- Design 0: 72 cm\(^2\) (3 flat sheets)
- Design 1A: 24 cm\(^2\) (1 flat sheet)
- Design 1B: 120 cm\(^2\) (5 flat sheets)
- Design 2A: 192 cm\(^2\) (2 flat sheets)
- Design 2B: 960 cm\(^2\) (10 flat sheets)

3200 GPU, 32 selectivity
- Design 1A\(_n\): 24 cm\(^2\) (1 flat sheet)
- Design 1B\(_n\): 120 cm\(^2\) (5 flat sheets)

3000 GPU, 25 selectivity
- Design 2A\(_n\): 192 cm\(^2\) (2 flat sheets)
- Design 2B\(_n\): 960 cm\(^2\) (10 flat sheets)
Simultaneous effect of changing $F^d$ and $P^{ratio}$

- Data include the combination two levels for the CO$_2$ permeance (1600 and 3200 GPU), two levels for the feed pressure (1 and 2 atm), five values of $F^d$, and five values of $P^{ratio}$
- The input flowrate and the permeate pressure are calculated variables.
- Input CO$_2$ molar fraction: 0.3; Selectivity: 32; Membrane area: 24 cm$^2$
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Power trendline function based on $F^d$

\[ S^{cut}_{|z=L} = 0.126(F^d)^{-0.743} \]

\[ R^2 = 0.99 \]

\[ -\ln \left( \frac{x_{CO2}^{feed}}{x_{CO2}} \right)_{|z=L} = 0.306(F^d)^{-0.896} \]

\[ R^2 = 0.99 \]
Kriging-based surrogate models

\[ f(x) \sim GP(m(x), k(x, x)) \]

Gaussian process

\[ k(x^i, x^j) = \exp \left( -\sum_{d=1}^{D} \frac{\gamma_d^2}{2} |x^i_d - x^j_d|^2 \right) \]

Kernel function (Gaussian kernel)

\[ \bar{y}_* = m(x) + k_\star^T (K + \sigma^2 I)^{-1} y \]

Predicted variable

\[ [K]_{i,j} = k(x^i, x^j) \]

Covariance matrix of training data

\[ [k_\star]_i = k(x^i, x^\star) \]

Covariance vector with the new point

\[ m(x) = \mu = \frac{1^T (K + \sigma^2 I)^{-1} y}{1^T (K + \sigma^2 I)^{-1} 1} \]

mean value

\[ m(x) \]: mean function
\[ \mu \]: mean value
\[ k(x, x') \]: covariance function
\[ \gamma \]: shape factor (vector)
\[ \sigma \]: Noise parameter
\[ x \]: input variables
\[ y \]: output variables
\[ N \]: number of points
\[ D \]: number of input variables

\[ y = [y_0, \ldots, y_{N-1}]^T, 1 = [1, \ldots, 1]^T \]
Kriging-based surrogate models

\[-6.906 + 8.025e^{-0.5(x_s-0.1)^2} + 7.717e^{-2.0(0.5x_s-1)^2} - 15.74e^{-8.0(0.25x_s-1)^2}\]

![Graph 1](image1.png)

![Graph 2](image2.png)

\[-6.951 + 2.623e^{-0.5(x_s-0.1)^2} + 17.75e^{-0.5(x_s-1.0)^2} - 50.29e^{-2.0(0.5x_s-1)^2} + 56.21e^{-3.125(0.4x_s-1)^2} - 26.29e^{-8.0(0.25x_s-1)^2}\]
Kriging-based surrogate models

**Input** dimensionless variables: \( F^d, P_{\text{ratio}}, Sel, x_{CO_2}^{\text{feed, initial}} \)

**Output** variables: \( CO_2 \) recovery and \( CO_2 \) purity in the permeate

\[ R^2: 0.99996, \text{ Mean relative error: 0.373 \% } \]

\[ R^2: 0.99985, \text{ Mean relative error: 0.377 \% } \]
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5. **Super-structure optimization for membrane-based separations.**
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Proposed membrane superstructure

**Given a superstructure**
- Determine the optimal process configuration
- The operating condition of separation stages
  - Feed pressure
  - Permeate pressure
  - Inlet concentration
  - Membrane area
  - Inlet flowrate
  - $\text{CO}_2$ permeance
  - Selectivity
Optimization formulation

\[ \min_x f(x) \]
\[ s.t. \ h(x) = 0 \]
\[ g(x) \leq 0 \]
\[ x^L \leq x \leq x^U \]

\[ CapCost = \frac{(\phi + \omega) \cdot CAPEX + OPEX + M_{REP}}{F_{CO_2}} \]

- Mass balances
- Units equipment models
- Cost correlations
- Pressure drop constraints
- Minimum purity target
- Minimum CO\(_2\) recovery target
- Minimum \(\Delta T\) in heat exchangers
- Pressure limits
- Mass flowrate per module
- CO\(_2\) concentrations
Membrane properties

Blue line: theoretical limit based on Robeson diagrams; Green line: assumed constraint (80 % of theoretical limit)


Fixed input flow of 200 sccm, \( x_{CO2}^{inlet} : 0.3; P^{fd, out} : 1.21 \text{ bar}; P^{pd, out} : 0.2 \text{ bar} \)
## Optimal results

<table>
<thead>
<tr>
<th></th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Capture cost ($/t-CO$_2$)</td>
<td>23.62</td>
</tr>
<tr>
<td>Total annual cost (MM$/y)</td>
<td>8.08</td>
</tr>
<tr>
<td>Capital cost (MM$)</td>
<td>26.31</td>
</tr>
<tr>
<td>Operating cost (MM$/y)</td>
<td>2.68</td>
</tr>
<tr>
<td>Purity (%)</td>
<td>95.00</td>
</tr>
<tr>
<td>CO$_2$ recovery (%)</td>
<td>90.00</td>
</tr>
</tbody>
</table>
Optimal results
### Optimal operating conditions for the membrane stages

<table>
<thead>
<tr>
<th>Input variables</th>
<th>M1</th>
<th>M2</th>
<th>M3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dimensionless feed flow ($F^d$)</td>
<td>0.21</td>
<td>0.41</td>
<td>0.29</td>
</tr>
<tr>
<td>Inlet CO$_2$ molar fraction</td>
<td>0.73</td>
<td>0.31</td>
<td>0.12</td>
</tr>
<tr>
<td>Pressure ratio ($P_{ratio}$)</td>
<td>2.42</td>
<td>8.65</td>
<td>11.90</td>
</tr>
<tr>
<td>Selectivity</td>
<td>33.02</td>
<td>37.39</td>
<td>35.61</td>
</tr>
<tr>
<td>Membrane area (m$^2$)</td>
<td>10216</td>
<td>18342</td>
<td>16933</td>
</tr>
<tr>
<td>Feed pressure (atm)</td>
<td>1.04</td>
<td>1.04</td>
<td>1.04</td>
</tr>
<tr>
<td>Permeate pressure (atm)</td>
<td>0.43</td>
<td>0.12</td>
<td>0.09</td>
</tr>
<tr>
<td>CO$_2$ permeance (GPU)</td>
<td>6000</td>
<td>4470</td>
<td>5018</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Output variables</th>
<th>M1</th>
<th>M2</th>
<th>M3</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO$_2$ recovery</td>
<td>0.813</td>
<td>0.715</td>
<td>0.709</td>
</tr>
<tr>
<td>CO$_2$ purity</td>
<td>0.950</td>
<td>0.839</td>
<td>0.510</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>CFD model results</th>
<th>M1</th>
<th>M2</th>
<th>M3</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO$_2$ recovery</td>
<td>0.815</td>
<td>0.711</td>
<td>0.706</td>
</tr>
<tr>
<td>CO$_2$ purity</td>
<td>0.945</td>
<td>0.848</td>
<td>0.517</td>
</tr>
</tbody>
</table>

High accuracy of the kriging-based model!
Dimensionless profiles of CO$_2$ molar fraction

M1
CO$_2$ recovery: 81 %
CO$_2$ purity: 95 %

M2
CO$_2$ recovery: 72 %
CO$_2$ purity: 84 %

M3
CO$_2$ recovery: 71 %
CO$_2$ purity: 51 %

$F^d$: 0.21; $x_{CO_2}$: 0.73; $pratio$: 2.42; Sel: 33.02

$F^d$: 0.41; $x_{CO_2}$: 0.31; $pratio$: 8.65; Sel: 37.39

$F^d$: 0.29; $x_{CO_2}$: 0.12; $pratio$: 11.90; Sel: 35.61
Conclusions

- CFD model for **fluid flow** and **diffusion**
- **Validated** bench scale model
  - Low relative error compared to experimental
- **Dimensional Analysis (DA)** can provide **four** dimensionless variables for the membrane separations: $F^d$, $p_{ratio}$, $Sel$, $x_{CO_2}^{feed,initial}$

- The dimensionless feed flow presents a relevant **physical meaning** associated with the time scales of mass transfer through the membrane and time scale of fluid to exit the feed side

- **Kriging-based surrogate models** were built to determine the CO$_2$ recovery and CO$_2$ purity in the retentate for a given combination of dimensionless variables

- Optimal design with three membrane stages shows a capture cost of **23.62 \$/t-CO_2**
Thank you!

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Hector Pedrozo
Cheick Dosso
Lorenz Biegler
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1. Motivation.
2. CFD model strategy and validation.
3. Dimensional Analysis for membrane separation.
6. **Optimizing Carbon Capture Plants with Hollow Fiber Membrane Technology: Integration of CFD Rigorous Models through Trust-Region Method**
Multi-scale optimization linked through reduced models

Mathematical formulation

Original problem

\[
\min_{z, w} f(z, w, y)
\]
\[
s.t. \quad h(z, w, y) = 0
\]
\[
g(z, w, y) \leq 0
\]
\[
y = t(w)
\]

\(t(w):\) truth model

Trust-region subproblem

\[
\min_x f(x)
\]
\[
s.t. \quad h(x) = 0
\]
\[
g(x) \leq 0
\]
\[
y = r_k(w)
\]
\[
\|x - x_k\| \leq \Delta_k
\]
\[
x^T = [z^T, w^T, y^T]
\]

\(t(w):\) truth model
Reduced models

**Challenge with Reduced Models:** Accuracy may be compromised in the vicinity of the optimal solution

**Enhanced accuracy through derivatives:** The use of derivative information improved the reduced models.
Key concepts

• Trust region methods have offered some **guaranteed convergence properties** for optimization with reduced models
• Trust region methods guarantee to find a locally optimal solution
• The trust region approach combines features of the **successive quadratic programming (SQP)** method with **model-based DFO** concepts
• Whereas SQP solves a nonlinear program through a **sequence of quadratic programming subproblems**, the **trust region method** solves the hybrid optimization problem through a **sequence of NLP subproblems**
• The motivation for this approach is to exploit the **cheap derivative information** that the model provides while reducing the black box calls
Case study 3: Trust region problem in Pyomo
Rigorous models

Hollow fiber membrane contactor
(CFD model in Comsol Multiphysics®)

Regenerator column
(Aspen Plus model)

Models for simple units (heat exchangers and pumps) are implemented in Pyomo directly.
Solution strategy

Objective function: CO₂ avoided cost of the capture process

Simple model units and economic models

Process specifications

Rigorous models

Trust-region constraints
Main economic indicators for the base case

<table>
<thead>
<tr>
<th></th>
<th>Base Case*</th>
<th>Optimal Design</th>
</tr>
</thead>
<tbody>
<tr>
<td>Capital cost (MM$)</td>
<td>105.80</td>
<td>40.87</td>
</tr>
<tr>
<td>Operating cost (MM$/y)</td>
<td>12.52</td>
<td>9.38</td>
</tr>
<tr>
<td>Total annual cost (MM$/y)</td>
<td>35.23</td>
<td>17.78</td>
</tr>
<tr>
<td>Reboiler demand (GJ/t-CO₂)</td>
<td>4.68</td>
<td>3.46</td>
</tr>
<tr>
<td>CO₂ avoided cost ($/t-CO₂)</td>
<td>118.01</td>
<td>55.33</td>
</tr>
<tr>
<td>CO₂ recovery (%)</td>
<td>92.43</td>
<td>90.00</td>
</tr>
</tbody>
</table>

Performance of the algorithm

33 Iterations of the trust-region filter method

θ: infeasibility metric
Δ: trust region size
CO₂ molar concentration (mol/m³) profile for a single hollow fiber

$Re_G: 16.56; x_{CO_2}: 0.15; Re_L: 4.55; w_{CO_2}: 5.31 \text{ wt\%}; w_{MEA}: 40 \text{ wt\%}; T_S: 320 \text{ K};$

$r_1: 0.375 \text{ mm}; r_3 - r_1: 0.130 \text{ mm}; r_4 - r_3: 0.359 \text{ mm}; l_f: 1.46 \text{ m}; \varepsilon: 0.55$
Conclusions

- The **trust-region filter method** is a powerful strategy to formulate optimization processes including rigorous CFD models
  - The optimization algorithm converged in 33 iterations, and the total CPU time was 1260 s
  - The optimal design for the capture plant with hollow fiber membrane contactors presents a CO$_2$ avoided cost of **55.33 $/t$-CO$_2$**, and a capture cost of 40 $/t$-CO$_2$
  - We find optimal values for the variables associated with the design of hollow fiber contactors, including the inner fiber radius, the membrane thickness, the hypothetical radius, the fiber length, and the porosity of the membrane
Thank you!

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