CFD and systems engineering to minimize membrane-based carbon capture costs

Hector A. Pedrozo, Cheick Dosso, Lingxiang Zhu, Victor Kusuma, Thien Tran, David Hopkinson, Lorenz T. Biegler, and Grigoris Panagakos

2024
Introduction

• Competitive carbon capture technologies based on polymeric membrane with high CO₂ permeance, high CO₂/N₂ selectivity, and stability
• CFD techniques provide the capability to analyze polymeric membranes under different conditions
• Multi-stage membrane configuration to achieve high capture rates and high purity simultaneously
• Membrane Systems Engineering to shed light in the true potential of this technology
CFD model
CFD model

\[ (\mathbf{u} \cdot \nabla) \mathbf{u} = \nu \nabla^2 \mathbf{u} - \frac{1}{\rho} \nabla p \]

- **Convective term**
- **Diffusion term**
- **Pressure gradient**

- **Velocity**
- **Kinematic viscosity**
- **Pressure**

- **Navier-Stokes**

- **Weight fraction**
- **Diffusion coefficient**

- **Velocity** \( \mathbf{u} = \frac{\text{m}}{\text{s}} \)
- **Kinematic viscosity** \( \nu = \frac{\text{kg}}{\text{m}^2 \cdot \text{s}} \)
- **Pressure** \( p = \text{Pa} = \frac{\text{kg}}{\text{m} \cdot \text{s}^2} \)

- **Jacobian** \( j_i = \frac{\text{kg}}{\text{m}^2 \cdot \text{s}} \)
- **Diffusion** \( D = \frac{\text{m}^2}{\text{s}} \)
- **Kinematic viscosity** \( \nu = \frac{\text{m}^2}{\text{s}} \)

- **Carnegie Mellon University**
Boundary condition for the selective layer

\[ j_i \left[ \frac{kg}{m^2 \cdot s} \right] \mid j \mid = Q_i \cdot M_i \cdot \left( p_1 \cdot x_{w_{ret}} - p_2 \cdot x_{w_{perm}} \right) \]

- \( j_i \) = Permeance
- \( Q_i \) = Pressure feed
- \( M_i \) = Molar mass
- \( x_{w_{ret}} \) = Mole fraction retentate side
- \( x_{w_{perm}} \) = Mole fraction permeate side

- \( Q_i \) = [GPU] = \( 3.346 \cdot 10^{-6} \) \[\frac{mol}{Pa \cdot s \cdot m^2}\]
- \( p \) = [Pa] = \[\frac{kg}{m \cdot s^2}\]

\( M_i \) = \[\frac{g}{gmol}\]
Meshing techniques

![Graph showing CO₂ recovery vs. number of degrees of freedom for a free tetrahedral mesh.](image)

- **Graph Details:**
  - **Y-axis:** CO₂ recovery
  - **X-axis:** Number of degrees of freedom
  - **Line:** Red dashed line representing the free tetrahedral mesh
Model validated using experimental results

\[
x_{\text{CO}_2}^{\text{inlet}} : 0.3
\]
\[
p_{\text{feed, out}} : 1.21 \text{ bar}
\]
\[
p_{\text{perm, out}} : 0.2 \text{ bar}
\]
\[
\text{CO}_2/\text{N}_2 \text{ selectivity: 28}
\]
\[
\text{CO}_2 \text{ permeance of 1600 GPU}
\]
Proposed membrane superstructure

Membrane units associated with CFD models
**Optimization formulation**

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

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

- 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
Data from CFD rigorous simulations
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)

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

Predicted variable

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

Covariance matrix of training data

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

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
\( k(x, x') \): covariance function
\( y \): shape factor \( \textbf{vector} \)
\( \sigma \): 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

\[ y^* = \mu + \sum_{i=1}^{N} \lambda_i k(x_i, x) \]

Estimated value

New input variables

Parameters

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

\[ -6.951 + 2.623e^{-0.5(x_s-0.1)^2} + 17.75e^{-0.5(x_s-1.0)^2} - 50.29e^{-2.0(0.5x_s-1)^2} + 56.21e^{-3.125(0.4x_s-1)^2} - 26.29e^{-8.0(0.25x_s-1)^2} \]
Surrogate model for the membrane process

<table>
<thead>
<tr>
<th></th>
<th>Training data</th>
<th>Validation data</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>CO₂ recovery</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( R^2 )</td>
<td>1.0000</td>
<td>0.9992</td>
</tr>
<tr>
<td>Mean squared error</td>
<td>2.6E-06</td>
<td>4.7E-05</td>
</tr>
<tr>
<td>Mean rel. error (%)</td>
<td>0.2819</td>
<td>1.5116</td>
</tr>
<tr>
<td>Max. rel. error (%)</td>
<td>2.1905</td>
<td>5.9386</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>Training data</th>
<th>Validation data</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Purity</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( R^2 )</td>
<td>1.0000</td>
<td>0.9998</td>
</tr>
<tr>
<td>Mean squared error</td>
<td>4.8E-07</td>
<td>8.0E-06</td>
</tr>
<tr>
<td>Mean rel. error (%)</td>
<td>0.1266</td>
<td>0.4301</td>
</tr>
<tr>
<td>Max. rel. error (%)</td>
<td>0.7907</td>
<td>1.9357</td>
</tr>
</tbody>
</table>

2D Scatter Plot
# Optimal results

<table>
<thead>
<tr>
<th></th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Capture cost ($/t-\text{CO}_2)</td>
<td>45.69</td>
</tr>
<tr>
<td>Total annual cost (MM$/y)</td>
<td>13.89</td>
</tr>
<tr>
<td>Capital cost (MM$)</td>
<td>46.25</td>
</tr>
<tr>
<td>Operating cost (MM$/y)</td>
<td>4.00</td>
</tr>
<tr>
<td>Purity (%)</td>
<td>90.00</td>
</tr>
<tr>
<td>\text{CO}_2 recovery (%)</td>
<td>80.00</td>
</tr>
</tbody>
</table>
Optimal results
### Output variables for the membrane stages

<table>
<thead>
<tr>
<th></th>
<th>M1</th>
<th>M2</th>
<th>M3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Membrane area (m²)</td>
<td>30849</td>
<td>107800</td>
<td>54887</td>
</tr>
<tr>
<td>Inlet flow (mol/s)</td>
<td>565</td>
<td>1465</td>
<td>901</td>
</tr>
<tr>
<td>Inlet CO₂ molar fraction</td>
<td>0.70</td>
<td>0.36</td>
<td>0.15</td>
</tr>
<tr>
<td>Retentate pressure (atm)</td>
<td>1.02</td>
<td>1.02</td>
<td>1.02</td>
</tr>
<tr>
<td>Permeate pressure (atm)</td>
<td>0.40</td>
<td>0.11</td>
<td>0.10</td>
</tr>
<tr>
<td>CO₂ recovery</td>
<td>0.61</td>
<td>0.74</td>
<td>0.55</td>
</tr>
<tr>
<td>CO₂ purity</td>
<td>0.90</td>
<td>0.70</td>
<td>0.42</td>
</tr>
<tr>
<td>Dimensionless feed flow</td>
<td>0.33</td>
<td>0.25</td>
<td>0.30</td>
</tr>
<tr>
<td>Retentate recovery</td>
<td>0.53</td>
<td>0.62</td>
<td>0.81</td>
</tr>
</tbody>
</table>

#### Results with the rigorous model

<table>
<thead>
<tr>
<th></th>
<th>M1</th>
<th>M2</th>
<th>M3</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO₂ recovery</td>
<td>0.608</td>
<td>0.740</td>
<td>0.551</td>
</tr>
<tr>
<td>CO₂ purity</td>
<td>0.906</td>
<td>0.701</td>
<td>0.445</td>
</tr>
</tbody>
</table>
**Profile of CO₂ molar fraction**

**M1**
- CO₂ recovery: 61%
- CO₂ purity: 90%

**M2**
- CO₂ recovery: 74%
- CO₂ purity: 70%

**M3**
- CO₂ recovery: 55%
- CO₂ purity: 42%

Input flow: 133.12 mg/s; $x_{CO₂}:0.70$; $p_{feed}: 1.02$ atm; $p_{perm}: 0.40$ atm

Input flow: 86.06 mg/s; $x_{CO₂}:0.36$; $p_{feed}: 1.02$ atm; $p_{perm}: 0.11$ atm

Input flow: 94.20 mg/s; $x_{CO₂}:0.15$; $p_{feed}: 1.02$ atm; $p_{perm}: 0.10$ atm
Effect of the inlet CO$_2$ concentration

<table>
<thead>
<tr>
<th>Inlet CO$_2$ molar fraction</th>
<th>Capture cost ($/t-CO$_2$)</th>
<th>Membrane area (m$^2$)</th>
<th>Energy demand (kWh/t-CO$_2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2</td>
<td>73.13</td>
<td>371880</td>
<td>284.5</td>
</tr>
<tr>
<td>0.225</td>
<td>64.77</td>
<td>320059</td>
<td>252</td>
</tr>
<tr>
<td>0.25</td>
<td>57.90</td>
<td>277819</td>
<td>231.4</td>
</tr>
<tr>
<td>0.275</td>
<td>52.01</td>
<td>244103</td>
<td>206.51</td>
</tr>
<tr>
<td>0.3</td>
<td>45.69</td>
<td>193536</td>
<td>182.8</td>
</tr>
</tbody>
</table>
Conclusions

• The computational framework is flexible and versatile
  • It can be adapted to accommodate various inlet and operating conditions to simulate any industrial plant and real-life gas stream compositions

• The optimal design includes the three membrane stages and the capture cost of the process is 45.69 $/t-CO₂

• Current work focus on including additional design variables associated with the membrane module through dimensionless numbers
Thank you!

Cheick Dosso
Lingxiang Zhu
Victor Kusuma
Thien (James) Tran
David Hopkinson
Grigorios Panagakos
Lorenz Biegler
Thank you for your attention!