Optimization Modeling of Energy Processes

L. T. Biegler
Department of Chemical Engineering
Carnegie Mellon University
Pittsburgh, PA 15213

http://capd.cheme.cmu.edu
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Energy Projects with NETL
(Energy Systems Initiative)

• **ROM-based Flowsheet Optimization for IGCC** (Y-D Lang)
  – ROMs developed from CFD models (gasifier and combustion)
  – Kriging models encapsulated within flowsheet optimization
  – High-fidelity optimization with 7% increase in power output

• **Process Synthesis for Oxycombustion** (A. Dowling, Q. Gao, Z. Gao)
  – MPCC models for phase transitions (flash, MHEX, GIBBS)
  – Accurate shortcut models for complex columns (e.g., ASU)
  – MINLP models for Oxycombustion flowsheet synthesis

• **Optimal Design/Operation of PSA Systems for CO2 Capture**
  (A. Agarwal, S. Vetukuri, A. Dowling, Y. Wang)
  – Superstructure developed for PSA system
  – Pre-combustion models analyzed – within DOE benchmark
  – Applied with Sequential optimization strategies
Motivation for CO$_2$ Capture

Existing pulverized coal plants

![Diagram of existing pulverized coal plants]

Post-combustion capture

Pre-combustion capture

Can we use PSA for carbon capture?

Challenges

- Which Sorbent works best for CO$_2$ capture?
- Which PSA cycle for high purity CO$_2$ capture?
- Computationally efficient flowsheet simulation/optimization with PDAE-based PSA model.
Optimization of PSA units for Carbon Capture
(Alex Dowling)
PSA Bed Model

Component mass balance

\[ \varepsilon_b \frac{\partial C_i}{\partial t} + (1 - \varepsilon_b) \rho_s \frac{\partial q_i}{\partial t} + \frac{\partial (vC_i)}{\partial z} = 0 \quad \forall i \]

LDF equation

\[ \frac{\partial q_i}{\partial t} = k_i (q_i^* - q_i) \]

Dual-site LANGmuir Isotherm

\[ q_i^* = \frac{q_i b_i P y_i}{1 + \sum_{j=1}^{nc} b_{ij} P y_j} + \frac{q_i b_{2i} P y_i}{1 + \sum_{j=1}^{nc} b_{2ij} P y_j} \]

Energy Balance

\[ \left( \varepsilon_t \sum_{i=1}^{nc} C_i \left( C_{pg}^i - R \right) + \rho_s C_{ps} + \rho_w C_{pw} \left( \frac{4h_w}{D} \right) \right) \frac{\partial T}{\partial t} - \rho_s \sum_{i=1}^{nc} \Delta H_i \frac{\partial q_i}{\partial t} + \frac{\partial (vh)}{\partial z} + \frac{4h_c (T - T_w)}{D} = 0 \]

\[ C_{pg}^i = a_c^i + b_c^i T + c_c^i T^2 + d_c^i T^3 \quad h = \sum_{i=1}^{nc} C_i C_{pg}^i T \]

Ergun equation

\[ - \frac{\partial P}{\partial z} = \frac{150 \mu (1 - \varepsilon_b)^2}{d_p^2 \varepsilon_b^3} v + \frac{1.75 (1 - \varepsilon_b)}{d_p^3 \varepsilon_b^3} \left( \frac{\sum M_w^i C_i}{1000} \right) v |v| \]

Ideal gas

\[ P = \sum_i C_i R T \]
PSA process for CO₂ separation

- **Applications**
  - Steam/methane reformer off gas
  - Power plant flue gas

What is special about this problem?

- Most commercial cycles designed for light product recovery and purity with heavy product often treated as waste

- Structural changes in cycles to offer CO₂ capture at high purity and recovery

- Need testbed to evaluate new sorbents – dependent on cycle and operating conditions
Two-bed PSA Superstructure

Systematic formulation to develop, evaluate and optimize PSA cycles

Co-current Bed (CoB)

Counter-current Bed (CnB)

Pressure-reducing Valve

Light Product (LP)

Top reflux (TR)

\( C_{a,i}(t), T_a(t), v_a(t), P_{ads}(t) \)

\( b(t) \)

\( P_a(t) \)

Input flux (F)

Feed compressor

Inlet gas

\( P_{inlet} \)

Inlet compressor (optional)

\( P_{feed} \)

\( f(t) \)

Heavy-product compressor

Bottom reflux (BR)

\( C_{d,i}(t), T_a(t), v_d(t), P_{des}(t) \)

Vacuum Generator

\( P_{atm} \)

Heavy Product (HP)

Allows all steps to be considered (P-FD-DP-EV-EQ-HP-LP)

Includes 2-bed interacting steps

Stack optimal profiles to obtain multi-bed systems with continuous feed and output
Jacobian of DAEs in Superstructure Model
Binary Separation

Finite Volume Discretization – smoothed flux limiters
Modeling/Designing Complex PSA Cycles

- 5 gases (N₂, CH₄, CO₂, CO, H₂)
- 5 beds, 11 steps
- 5 stages, 3 periods/stage
- Dual-adsorbent layers (APHP, UOP 5A)

### Uni-bed Simulation

**Note:** 2-bed interactions

<table>
<thead>
<tr>
<th>Feed</th>
<th>EQ1</th>
<th>EQ2</th>
<th>Pr purge</th>
<th>EQ3</th>
<th>Blow down</th>
<th>Purge</th>
<th>EQ3</th>
<th>EQ4</th>
<th>Idle</th>
<th>EQ1</th>
<th>Repr.</th>
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<tbody>
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<td>EQ1</td>
<td>Repr.</td>
<td>Feed</td>
<td></td>
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<td>EQ2</td>
<td>Pr purge</td>
<td>EQ3</td>
<td>Blow down</td>
<td>Purge</td>
<td>EQ3</td>
<td>EQ2</td>
</tr>
<tr>
<td>EQ3</td>
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<td>Idle</td>
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<td>Feed</td>
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### Multi-bed Simulation
Minimize specific energy \((\text{kWh}/\text{tonne CO}_2 \text{ captured})\)

- **Objective Function, Constraint Evaluations, & Derivative Info**
- **PSA Superstructure**
  - PSA Bed Model
  - Connectivity Equations
  - Compressor and Turbine Model
  - Valve Equations
  - **Cyclic Steady-State Constraint**
- **Optimization Algorithm**
- **Decision Variable Values**

3 approaches for cyclic-steady state (CSS)
- Embedded fixed point (NDO)
- Fixed Horizon (Small NLP)
- Periodic BCs (Larger NLP)
Implementation Details
(Existing Tools)

Formulation of Bed Equations

- Finite Volume Discretization of Bed Model (11 elements)
- Smoothed van Leer Flux Limiter
- States include gas concentration, solid loading, T, P, T_w

Integration of Bed Equations

- State equations solved with CVODES 2.6.0 (BDF, Sundials)
- Direct and adjoint sensitivity (no checkpointing in CVODES)
- Exact BDF Jacobian using ADOL-C

BOBYQA for Internal Fixed Point Form

- DFO code based on quadratic approximation to objective function
- Accommodates independent variable bounds
- Purity and Recovery constraints handled by $l_2$ penalty functions

IPOPT 3.10 for gradient formulations (equality constrained, finite horizon)

- First derivatives from sensitivity equations
- Second derivatives approximated with LBFGS
Minimize Power for Pre-combustion Capture  
(Dowling, Vetukuri, B., 2012)

Min Total Power \((\text{kWh/tonne CO}_2)\)
\[
\text{CO}_2 \text{ purity } \geq 0.92 \\
\text{CO}_2 \text{ recovery } \geq 0.90 \\
a(t), b(t), f(t) \in [0, 1] \\
\text{P}_{\text{ads}} \geq 5 \text{ bar} \\
\text{P}_{\text{des}} \geq 10^{-4} \text{ bar} \\
\text{P}_{\text{ads}} \geq \text{P}_{\text{d}} \\
\text{P}_{\text{a}} \geq \text{P}_{\text{des}} \\
\text{P}_{\text{feed}} \geq \text{P}_{\text{des}} \\
\text{(CSS constraints)}
\]

Adsorbent: Activated Carbon
Feed: \( \text{H}_2 \ (58\%) \) and \( \text{CO}_2 \ (42\%) \)
Feed: \( \text{H}_2 \ (56.5\%), \text{CO}_2 \ (42\%), \) \( \text{CO}(0.8\%), \text{CH}_4(0.6\%), \text{N}_2\) (trace)
\[
\text{P}_{\text{feed}}: \ 51 \text{ bar}, \ \text{T}_{\text{feed}}: \ 308 \text{ K} \\
\text{P}_{\text{out}} \text{ for } \text{H}_2 \text{ Product}: \ 31 \text{ bar} \\
\text{P}_{\text{out}} \text{ for CO}_2 \text{ Product}: \ 150 \text{ bar}
\]
Designed Pre-Combustion Cycle

Step 1
Step 2
Step 3
Step 4
Step 5

Switch Beds and Repeat

Legend: CO₂
Sorbent Loading
High
Low

Best 5 Component Solution

Adsorbing Bed (produces H₂)

Desorbing Bed (produces CO₂)
86.8 kWh/tonne CO$_2$ captured

3.2 MPa

Light Product Compressor

13.0 kWh/tonne

Top Reflux Fraction $b(t)$

< 2.8 MPa

Co-current Bed

12.6 kWh/tonne

Heavy Reflux Compressor

-35.4 kWh/tonne

Bottom Reflux Fraction $a(t)$

-3.3 kWh/tonne

Heavy Product Compressor

15 MPa

> 0.02 MPa

Counter-current Bed

0.02 MPa

Vacuum Generator

99.9 kWh/tonne

Carbon Dioxide Rich Stream to Pipeline

5.1 MPa

Feed from WSR, $f(t)$

3.2 MPa

Hydrogen Rich Stream to Turbine

> 0.02 MPa

Feed Turbine

5.1 MPa

Feed from WSR, $f(t)$

> 0.02 MPa

Feed Turbine

3.2 MPa

Light Product Compressor

13.0 kWh/tonne

Top Reflux Fraction $b(t)$

< 2.8 MPa

Co-current Bed

12.6 kWh/tonne

Heavy Reflux Compressor

-35.4 kWh/tonne

Bottom Reflux Fraction $a(t)$

-3.3 kWh/tonne

Heavy Product Compressor

15 MPa

> 0.02 MPa

Vacuum Generator

99.9 kWh/tonne

Carbon Dioxide Rich Stream to Pipeline

5.1 MPa

Feed from WSR, $f(t)$

> 0.02 MPa

Feed Turbine
Computational Performance

Part A: Binary System (CO$_2$, H$_2$)

<table>
<thead>
<tr>
<th>Approach</th>
<th>Obj. Func kWh/tonne CO$_2$</th>
<th>CPU Time/Iter h:mm:ss</th>
<th>Iter</th>
</tr>
</thead>
<tbody>
<tr>
<td>Periodic Bnd. Cnd. (1)</td>
<td>83.51</td>
<td>0:10:37</td>
<td>82</td>
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<tr>
<td>Fixed Horizon (3)</td>
<td>86.46</td>
<td>0:21:09</td>
<td>56</td>
</tr>
</tbody>
</table>

Part B: Five Components (CO$_2$, H$_2$, CH$_4$, N$_2$, CO)

<table>
<thead>
<tr>
<th>Approach</th>
<th>Obj. Func kWh/tonne CO$_2$</th>
<th>CPU Time/Iter h:mm:ss</th>
<th>Iter</th>
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</thead>
<tbody>
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<td>470</td>
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<tr>
<td>Derivative Free (2)</td>
<td>109.04</td>
<td>0:11:29</td>
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<tr>
<td>Fixed Horizon (3)</td>
<td>86.81</td>
<td>1:27:52</td>
<td>260</td>
</tr>
</tbody>
</table>

Multiple common starting points

Internal Fixed Point approach terminates at infeasible solution
  • Local minima, effect of noise, poor scaling

Derivative based approaches: KKT conditions not always satisfied
  • Terminate due to resource limits, accumulation points
  • Noisy first derivatives, approximate (L-BFGS) second derivatives
PSA Optimization Conclusions

• Compared three PSA optimization formulation

• Developed novel application of adjoint sensitivity equations to PSA optimization

• Demonstrated potential cost competitiveness of PSA for $\text{H}_2$-$\text{CO}_2$ separation in IGCC power plant with an activated carbon sorbent

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