From infeasibility certificates towards global optimization of chromatographic processes

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Introduction: Chromatographic processes

True Moving Bed (TMB) process

- **Purpose:** Separation of a mixture $A + B$.
- Based on the different adsorption behaviors of $A$ and $B$ w.r.t. a certain solid $S$.
- **Isotherms** model the relation between liquid-phase and solid-phase concentrations.
- **Key design variables:** Flow-rate ratios $m_i := \frac{\dot{V}_i}{\dot{V}_S}$, $i \in \{1, 2, 3, 4\}$.

Central design question:

Which points $(m_1, m_2, m_3, m_4)$ allow to separate the mixture $A + B$ w.r.t. a given TMB-configuration? → Separation regions
Determination of separation regions

Equilibrium stage model for 4-zone TMB processes

Each zone $i$ is divided into a theoretical number of plates $j$.

- Mass balance equations: $q^k_{i,j+1} + m_i \cdot c^k_{i,j-1} - m_i \cdot c^k_{i,j} - q^k_{i,j} = m_{ext} c^k_{ext}$.
- Purity requirements: $\frac{c^A_i}{c^A_i + c^B_i} \geq \text{pur}^A$.
- Isotherms: $q^k = q^k(c^A, c^B)$.

Second-order isotherms

$$q^A(c^A, c^B) = \frac{q_s c^A(b_{10} + 2b_{20} c^A + b_{11} c^B)}{1 + b_{10} c^A + b_{01} c^B + b_{11} c^A c^B + b_{20} (c^A)^2 + b_{02} (c^B)^2}$$

→ model inflection points.

1. **Scanning techniques** evaluate a finite set of given points.
2. **Global optimization techniques** evaluate all points within a certain domain.

**Aim:** Adopt global optimization techniques for second-order isotherms.
Idea: Exploit convex programming.

1. Consider a possible subdomain $P$.
2. $P = \{ x \in \mathbb{R}^n \mid f(x) \leq 0 \}$ is non-convex.
3. Construct a convex relaxation $P^\ast$.
4. $P^\ast$ infeasible $\Rightarrow$ $P$ infeasible

Optimization: $\max(P) \leq \max(P^\ast)$
Infeasibility certificates and global optimization

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Convexification of real-valued functions

- $P$ is convex if $f(x)$ is convex.
- Use a convex underestimator $f^*(x)$:
  \[ f^*(x) \leq f(x) \leq 0. \]
- The tightest convex underestimator is called convex envelope $\text{vex}_D[f](x)$. 
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Reformulation strategies for second-order isotherms

There is no convex envelope known for second-order isotherms $q_{i,j}^k(c_{A,i,j}, c_{B,i,j})$

$$q^A(c^A, c^B) = \frac{qs c^A(b_{1,0} + 2b_{2,0}c^A + b_{1,1}c^B)}{1 + b_{1,0}c^A + b_{0,1}c^B + b_{1,1}c^A c^B + b_{2,0}(c^A)^2 + b_{0,2}(c^B)^2} =: \frac{r(c^A, c^B)}{s(c^A, c^B)}$$

⇒ Reformulation into known structures.
  - Bilinear terms $xy$ (McCormick [1976]).
  - Trilinear terms $xyz$ (Meyer & Floudas [2003,2004]).
  - Bivariate quadratic terms $a_{20} x^2 + a_{11} xy + a_{02} y^2$ (Jach et al. [2008]).
  - Fractional terms $x/y$ (Tawarmalani & Sahinidis [2001]).

Reformulation Strategies:

- **RS1:** $q^A + b_{1,0} q^A c^A + \ldots + b_{1,1} q^A c^A c^B = q_s b_{1,0} c^A + \ldots + b_{1,1} q_s c^A c^B$
- **RS2:** $q^A s' = r', \quad s' = s(c^A, c^B), \quad r' = r(c^A, c^B)$
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Computational experiments

Statistical isotherms of second degree

\[
q_{i,j}^A = q_{i,j}^A(c_{i,j}^A, c_{i,j}^B) = \frac{5c_{i,j}^A(1 + 4c_{i,j}^A + 1c_{i,j}^B)}{1 + 1c_{i,j}^A + 2c_{i,j}^B + 2(c_{i,j}^A)^2 + 3(c_{i,j}^B)^2 + 1c_{i,j}^A c_{i,j}^B},
\]

\[
q_{i,j}^B = q_{i,j}^B(c_{i,j}^A, c_{i,j}^B) = \frac{5c_{i,j}^B(2 + 6c_{i,j}^B + 1c_{i,j}^A)}{1 + 1c_{i,j}^A + 2c_{i,j}^B + 2(c_{i,j}^A)^2 + 3(c_{i,j}^B)^2 + 1c_{i,j}^A c_{i,j}^B}.
\]

Test instances

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<th>test</th>
<th>(m_1)</th>
<th>(m_2)</th>
<th>(m_3)</th>
<th>(m_4)</th>
<th>(\text{pur}^k)</th>
<th>(c_F^k)</th>
<th>(M_i)</th>
<th>(c_{i,j}^A)</th>
<th>(c_{i,j}^B)</th>
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<tr>
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<td>[15,16]</td>
<td>[3,5]</td>
<td>[8,10]</td>
<td>[0.25,1.25]</td>
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<td>30</td>
<td>[0.0, 0.2]</td>
<td>[0.0, 0.1]</td>
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<tr>
<td>T2</td>
<td>[15,16]</td>
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<td>0.950</td>
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<tr>
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<td>[8,10]</td>
<td>[0.25,1.25]</td>
<td>0.995</td>
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<td>60</td>
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</tr>
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Each test instance represents a possible subproblem of a certain TMB process.
Computational experiments (2) : $\max(m_3 - m_2)$

Results (on SUN FireV440 - UltraSPARC-IIIi, 16GB RAM with Cplex V9.10)

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Problem sizes of the LP-relaxations

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Lifting technique

Idea (cf. Padberg [1976], Gangadwala et al. [2006]):

- Consider \( q^A = q^A(c^A, c^B) \) on \([l^A, u^A] \times [l^B, u^B]\).
- Fix one variable to one of its bounds, e.g. \( c^A = l^A \).
- Find a \( \mu \in \mathbb{R} \) such that
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  q^A(l^A, c^B) + \mu(c^A - l^A) \leq q^A(c^A, c^B), \quad \text{for all } c^B \in [l^B, u^B], \ c^A = l^A
  \]

The best possible value \( \mu^* \) can be determined by an optimization problem:

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<td>750</td>
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Behavior of the separation regions w.r.t. three factors

$c_F^k = 0.001$

$c_F^k = 0.150$

$c_F^k = 0.250$

Chromatographic aspects

- Influence of different feed concentrations, number of stages and purity requirements.
- Separation regions are more elaborate compared to linear and Langmuir isotherms.
- Observation: Maximal feed stream is not necessarily achieved at the turning point of the region.

#stages
- $=204$
- $=404$
- $=404$

$pur^k$
- $=0.995$
- $=0.995$
- $=0.900$
Conclusion

Aim 1: Adopt global optimization techniques for second-order isotherms

- Analysis of three reformulation strategies
  - **RS1** “standard” reformulation
    → Good relaxation vs. slow computation.
  - **RS2** and **RS3** exploit larger structures
    → Weaker relaxation vs. fast computation.

⇒ Choice of reformulation strategy is not clear.

- **Lifting** technique
  - General formula for an underestimator for interesting domains
    → Strong relaxation & fast computation.

Aim 2: Separation regions for TMB processes with second-order isotherms

- Determination of **feasible operating points** and **infeasible regions**.
- More elaborated separation regions compared to standard isotherm models.