Optimal Membrane Cascade Design for Critical Mineral Recovery through Logic-based Superstructure Optimization

FOCAPD2024
FOUNDATIONS OF COMPUTER-AIDED PROCESS DESIGN

PROMMIS
Process Optimization and Modeling for Minerals Sustainability

Norman Tran, Daniel Ovalle, Ignacio E. Grossmann, Carl D. Laird
Department of Chemical Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, USA

Motivation

- **Critical Minerals (CM)** underpin the clean energy technologies that are central to our climate change initiatives, particularly in applications such as electric vehicles (EVs) and energy storage (ES) [1].
- **Recycling of lithium-ion batteries** is an important approach to address the increased demand for EVs and ES, lack of domestic CM production, and import dependency concerns.
- **Diafiltration Membranes** offer a significant advantage over existing battery recycling pathways, reducing energy and chemical cost.

Generalized Disjunctive Programming

$$\min \quad f(x)$$ — Objective function

$$s.t. \quad g(x) \le 0$$ — Global constraint

$$\underset{i \in D_k}{\veebar} \begin{bmatrix} Y_{ik} \\ r_{ik}(x) \le 0 \end{bmatrix}, \ k \in K$$ — Disjunction

$$\Omega(Y_{i,k}) = \text{True}$$ — Logic proposition

$$x \in \mathbb{R}^n$$

$$Y_{ik} \in \{\text{True}, \text{False}\}, \quad i \in D_k, k \in K$$

- **Generalized Disjunctive Programming (GDP)** allows the modeler to embed logic into optimization formulations.
- Exploits existing implementation and solution approach.
- Avoids the **zero-flow** issue [2].

Formulation

- Includes transport and balance equations for discretized element of the membrane model.
- **Link** *N total membrane elements* using disjunctions resulting in a single membrane stage.
- **Link** *K total stages* forming a membrane cascade system.

$$\max \ F^{r_{out}}_{|N|,1} x^{r_{out}}_{Co,|N|,1}$$ — Maximize extraction of **cobalt**

$$\begin{bmatrix} Y^{ST}_{nk} \\ F^{ST}_{nk} = \hat{F}^{ST} \\ x^{ST}_{cnk} = \hat{x}^{Feed}_{c} \ \ \forall c \in C \end{bmatrix} \veebar \begin{bmatrix} \neg Y^{ST}_{nk} \\ F^{ST}_{nk} = 0 \\ x^{ST}_{cnk} = 0 \ \ \forall c \in C \end{bmatrix},$$
$$\forall n \in N, \ k \in K, ST = \{Feed, Diafiltrate\}$$ — Only one feed/diafiltrate per stage

$$\begin{bmatrix} Y^{Refl}_{nk} \\ F^{Refl}_{nk} = F^{r_{out}}_{|N|,\{k+1\}} \\ x^{Refl}_{cnk} = x^{r_{out}}_{c,|N|,\{k+1\}} \ \ \forall c \in C \end{bmatrix} \veebar \begin{bmatrix} \neg Y^{Refl}_{nk} \\ F^{Refl}_{nk} = 0 \\ x^{Refl}_{cnk} = 0 \ \ \forall c \in C \end{bmatrix},$$
$$\forall n \in N, \ k \in K \setminus \{|K|\}$$ — Only one reflux stream per stage except the last stage

$$F^{p_{out}}_{|N||K|} x^{p_{out}}_{Li,|N||K|} \ge R^{Li}_{min} \left(\hat{F}^{Feed} \hat{x}_{Li}^{Feed} + \hat{F}^{Diaf} \hat{x}_{Li}^{Diaf} \right)$$ — Ensure minimum extraction of **lithium**

$$x^{r_{out}}_{cnk} = x^{r_{in}}_{cnk} \left(\frac{F^{r_{out}}_{nk}}{F^{r_{in}}_{nk}} \right)^{S_c - 1}, \forall c \in C, n \in N, \ k \in K$$ — Flux across membrane. Constant sieving coefficient [3]

python™ · PYOMO GDP

Reformulate as Big M
Solved using SCIP v. 8.0

3 stages 10 elements	• 160 binary variables • 750 continuous variables • 1554 constraints

Conclusions

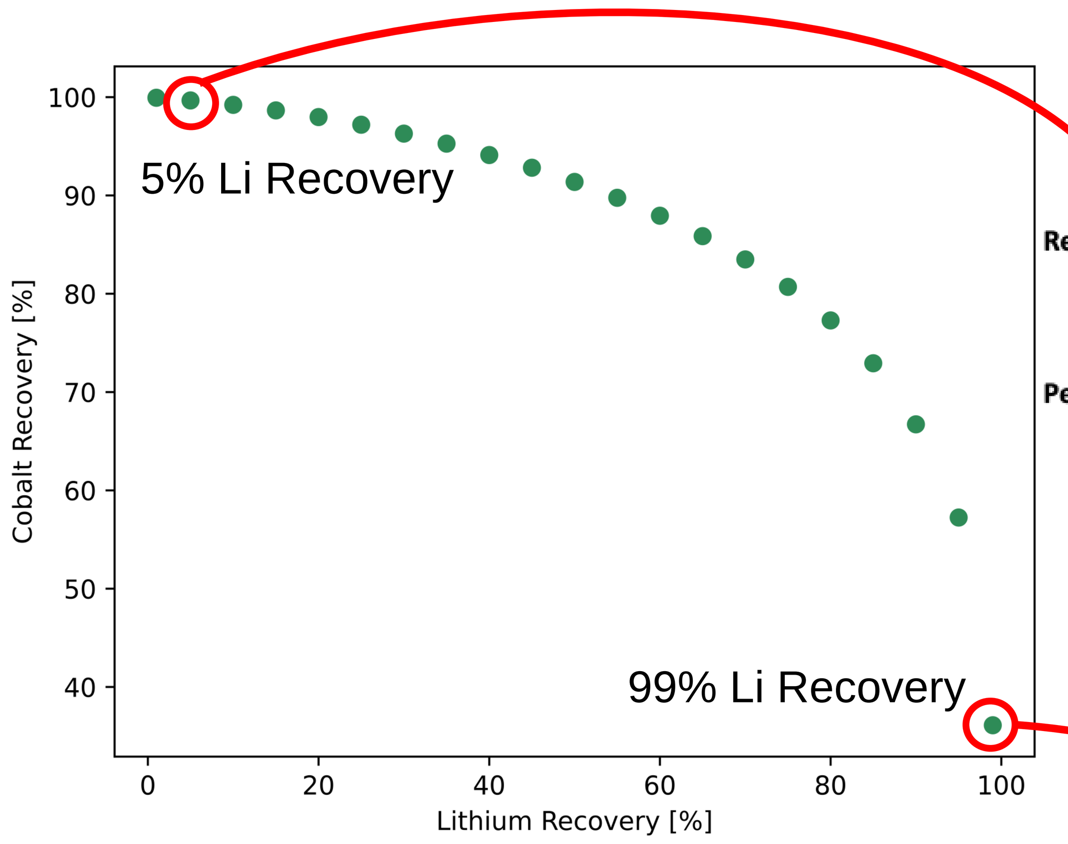
- This work demonstrates the applicability of Generalized Disjunctive Programming in superstructure design of a multi-stage membrane cascade system to separate cobalt and lithium.
- Disjunctions and logic propositions guarantee a single feed, diafiltrate, and reflux flow per stage. This allows numerical stability and avoids singularity in nonlinear expressions, i.e., zero-flow issue.
- Our results show the impact of additional membrane stages on recovery and the available trade-off between recovery of Co or Li.

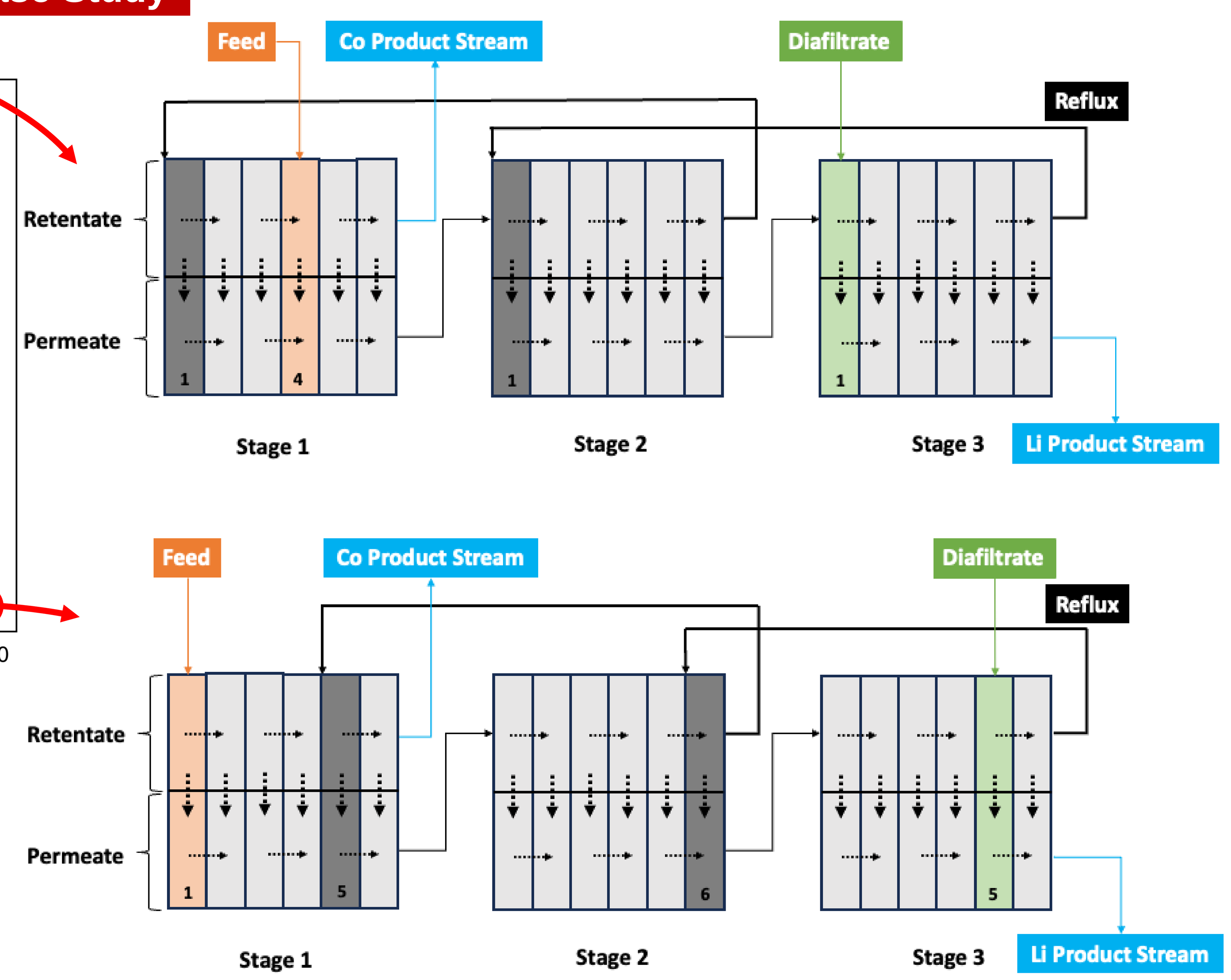
Future Work

- Extend the GDP modeling approach to solvent extraction system.
- Apply GDP algorithm and reformulation approach to determine stage existence and separation sequence for multi-component system.
- Integrate recycling scheme into solvent extraction to determine the impact on fresh solvent usage.

Results

3 Stages 6 Elements Optimal Design Case Study

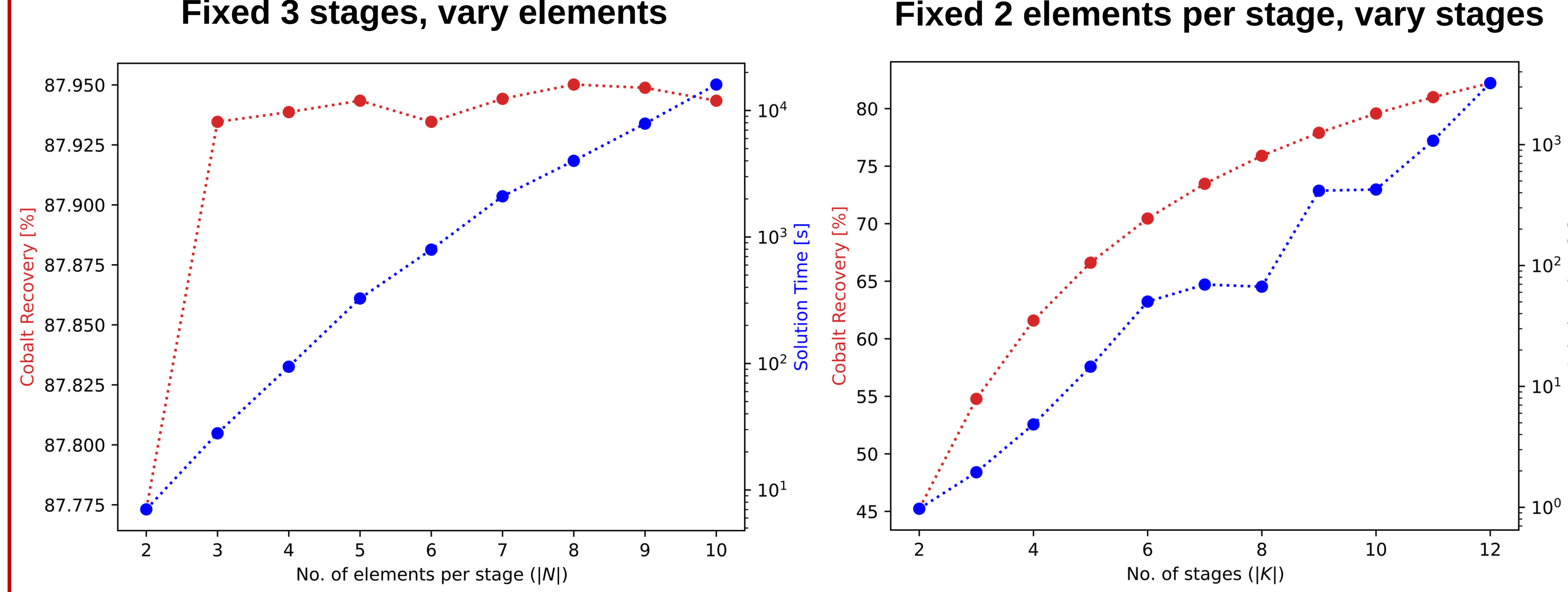




- 96 binary variables
- 462 continuous variables
- 946 constraints

Solves to global optimality with 0.001% gap

Computational Performance– 60% Li Recovery



- Varying the discretized element offers minimal increase (~0.2%) in cobalt recovery.
- Increase in number of stages correlates with an increase in cobalt recovery.
- Potential for trade-off analysis between the focus on cobalt recovery and the cost to install additional membrane stages

References

[1] - IEA (2021), The Role of Critical Minerals in Clean Energy Transitions, IEA, Paris

[2] - Chen Q, Liu Y, Seastream G, Siirola JD, Grossmann IE. Pyosyn: a new framework for conceptual design modeling and optimization. Computers & Chemical Engineering 153 (2021)

[3] - Wamble NP, Eugene EA, Phillip WA, Dowling AW. Optimal diafiltration membrane cascades enable green recycling of spent lithium-ion batteries. ACS Sustainable Chemistry & Engineering 10.37 (2022)

Acknowledgement

This effort was funded by the US Department of Energy's Process Optimization and Modeling for Minerals Sustainability (PrOMMiS) Initiative, supported by the Office of Fossil Energy and Carbon Management's Office of Resource Sustainability.

Disclaimer