Advanced Modeling and Process-Materials Co-Optimization Strategies for Swing Adsorption Based Gas Separations

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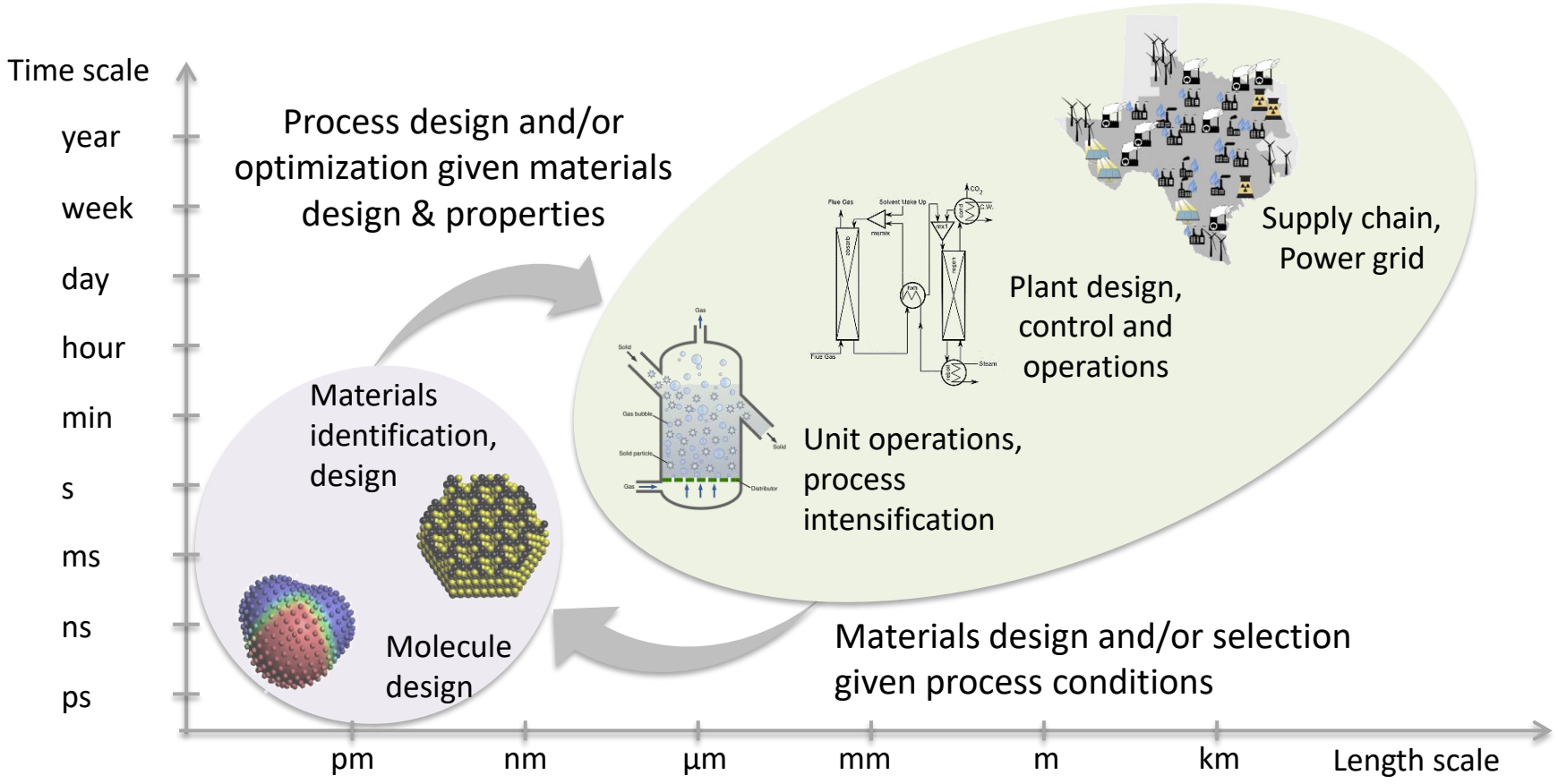
April 20th, 2023

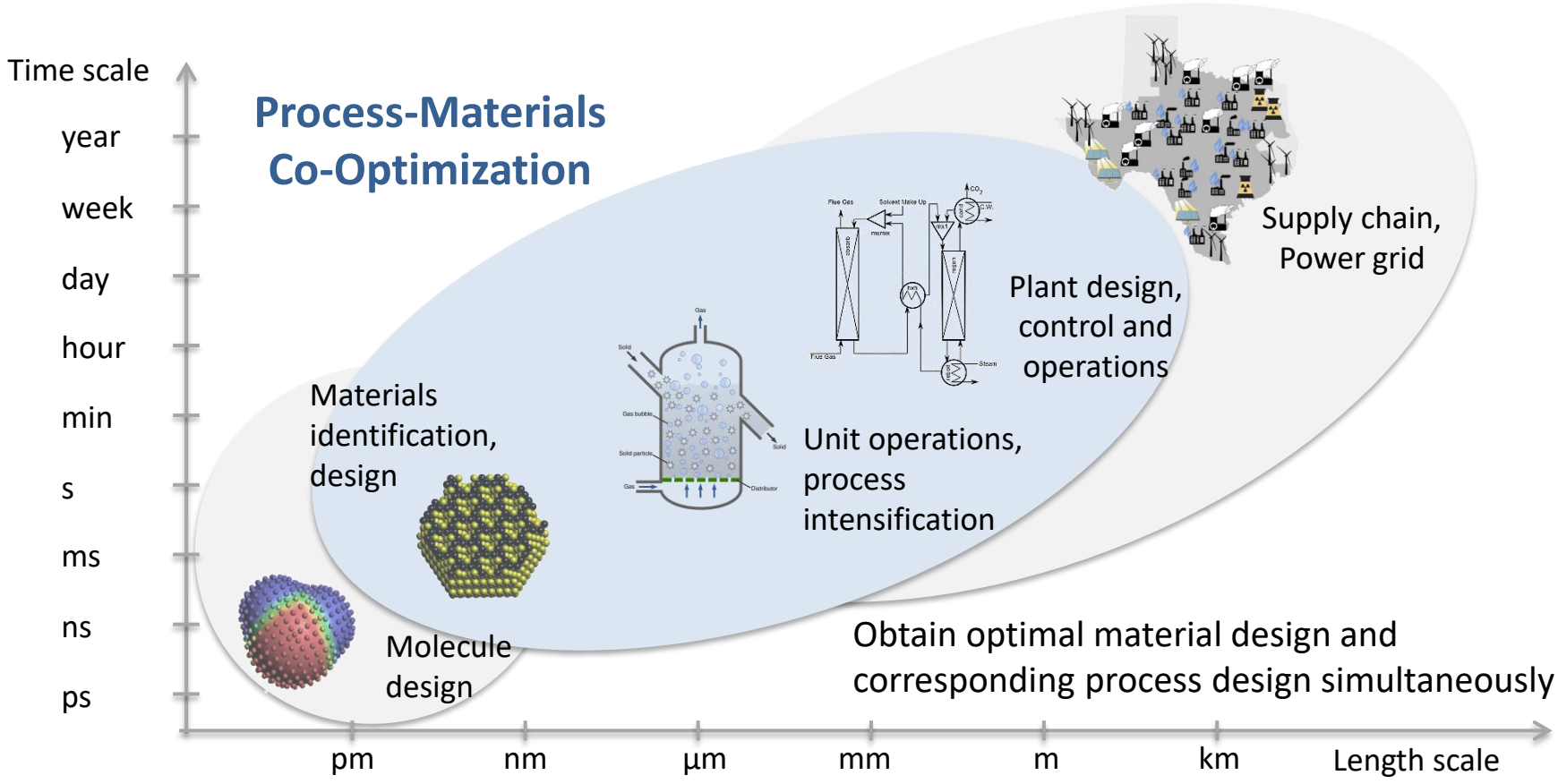
Motivation

- Multi-scale highly integrated energy systems



Motivation

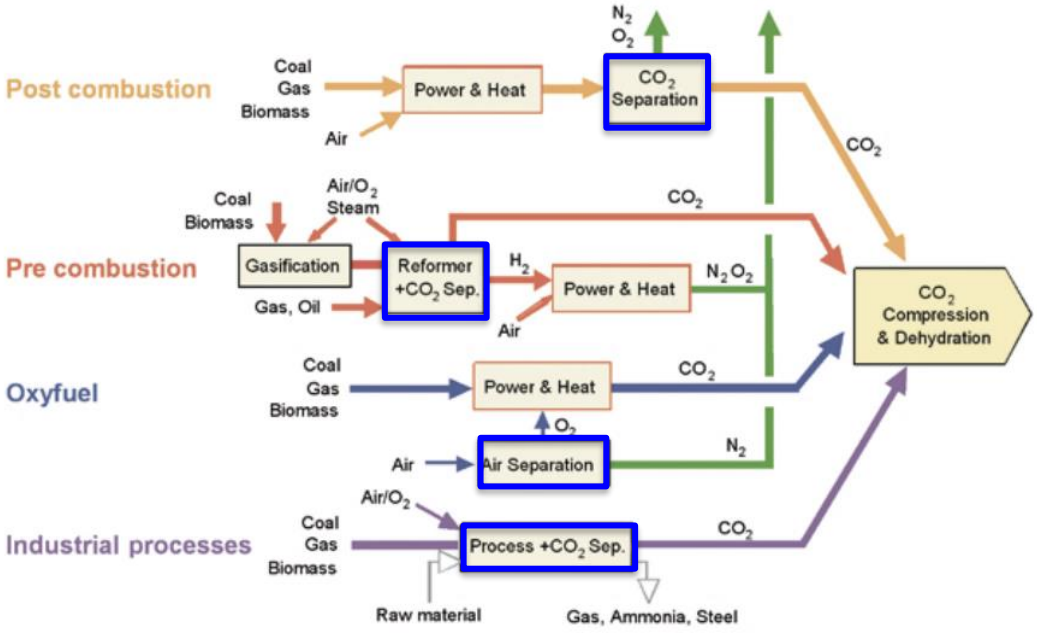
- Multi-scale highly integrated energy systems



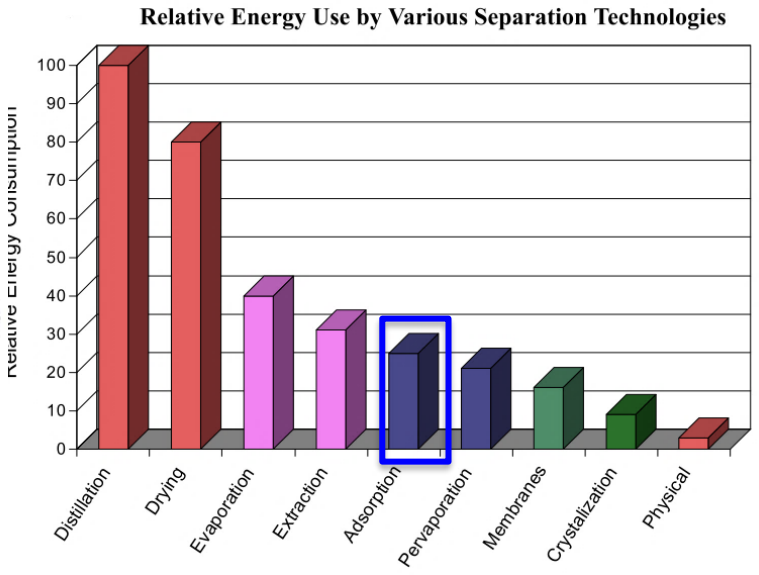
Opportunities exist to optimize materials and processes simultaneously

Adsorption-Based Gas Separation

- Gas separation processes are crucial parts of next-generation energy and environmental technologies
- Adsorption-based separation technologies have been intensively investigated for potential low energy consumption



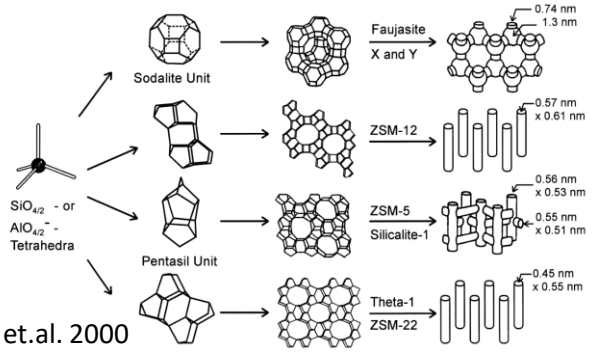
Metz, B., Davidson, O., De Coninck, H.C., Loos, M. and Meyer, L., 2005. IPCC special report on carbon dioxide capture and storage. Cambridge: Cambridge University Press.



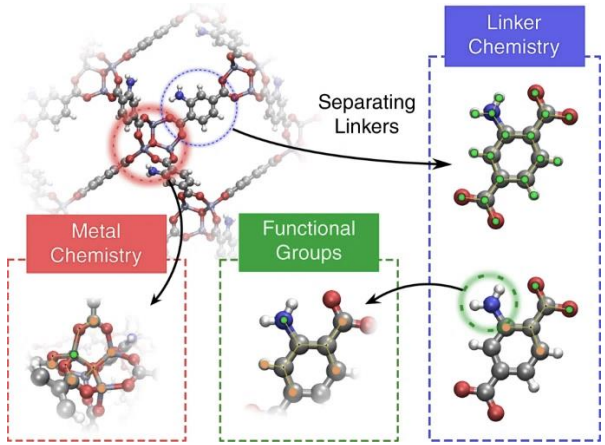
Angelini, P., Armstrong, T., Counce, R., Griffith, W., Klasson, T.L., Muralidharan, G., Narula, C., Sikka, V., Closset, G., Keller, G. and Watson, J., 2005. Materials for separation technologies: Energy and emission reduction opportunities. DOE, EERE Office, Washington, DC, 103.

Microporous Materials as Adsorbents

- Microporous materials (i.e., MOFs, Zeolites) are promising adsorbents for adsorption-based gas separation processes
- Vast design space of microporous materials calls for systematic computational search method



Weitkamp et.al. 2000



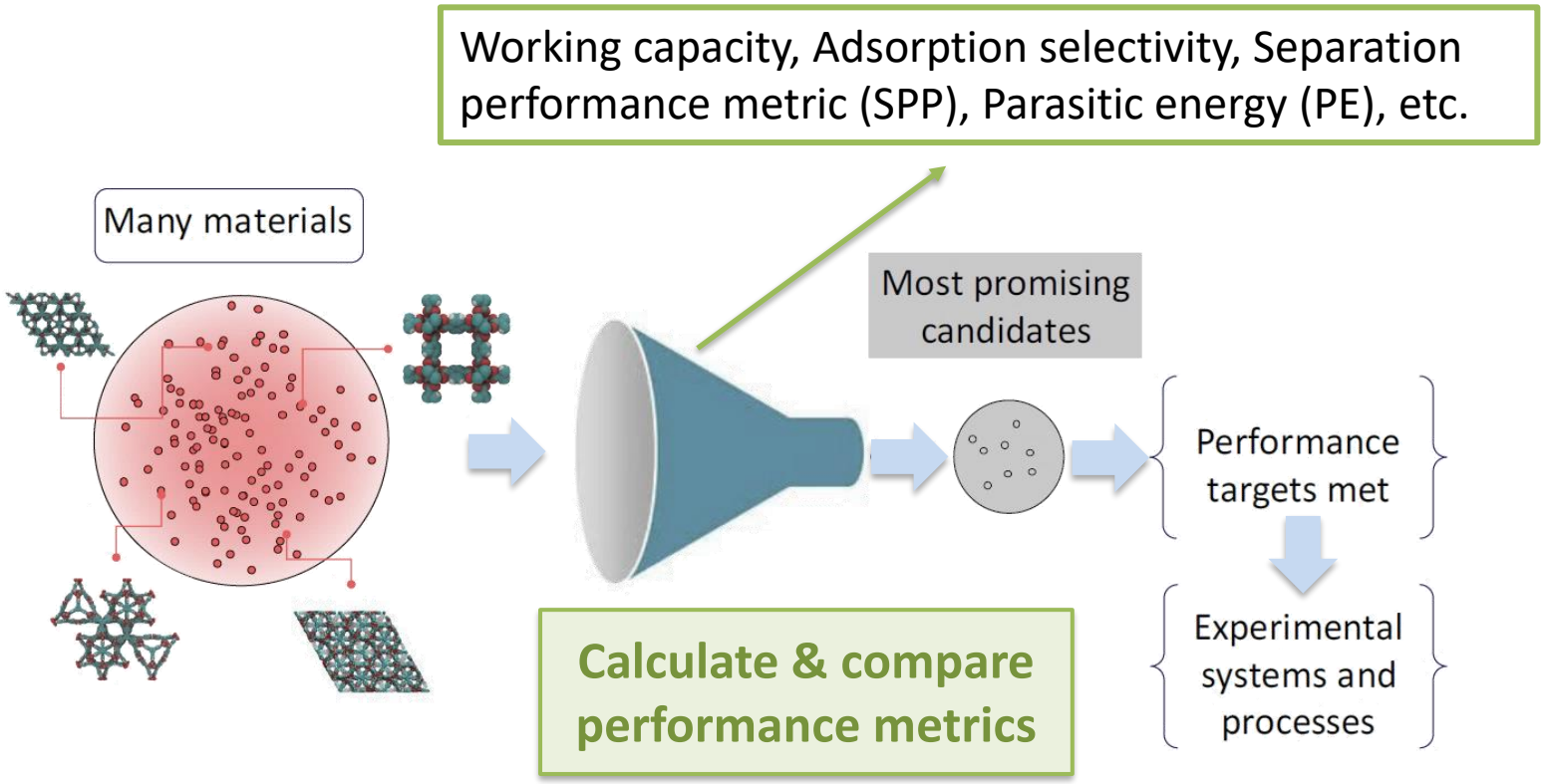
Moosavi et. al. 2016

Database	Number of entries	Origin
CoRE-MOF	14,000+	Experiments
hMOF	137,953	Simulations
ToBaCCo	13,512	Simulations
CSD-MOF	96,000+	Experiments
IZA	252	Experiments
hZeo	2.6M	Simulations
CoRE-COF	449	Experiments
hCOF	69,840	Simulations
CURATED COFs	482	Experiments

247 trillion MOFs (Lee et. al. 2021)

Process-Materials Co-Design Approaches

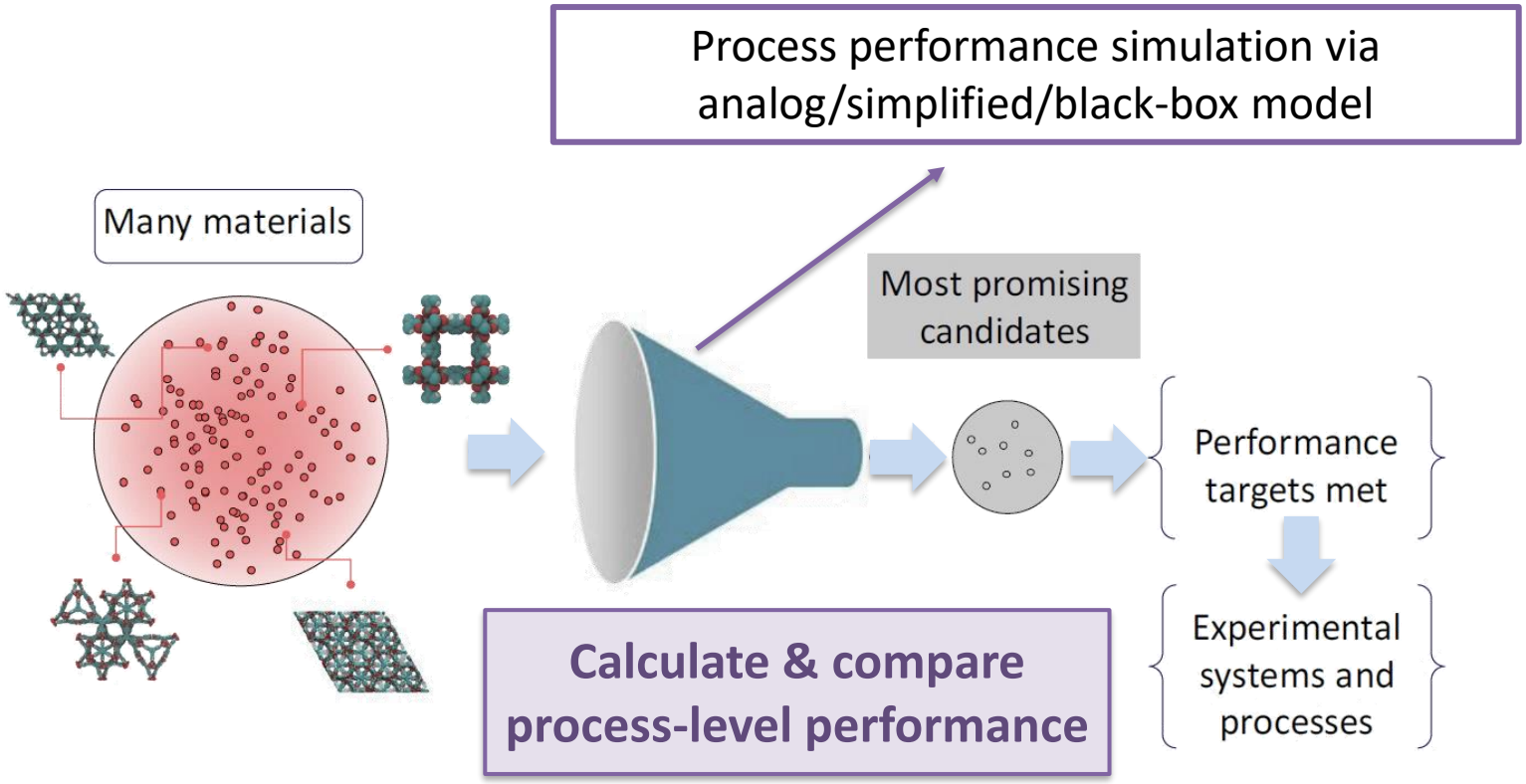
- In silico materials screening



Farmahini, A.H., Krishnamurthy, S., Friedrich, D., Brandani, S. and Sarkisov, L., 2021. Performance-based screening of porous materials for carbon capture. Chemical Reviews, 121(17), pp.10666-10741.

Process-Materials Co-Design Approaches

- Process simulation-based materials screening



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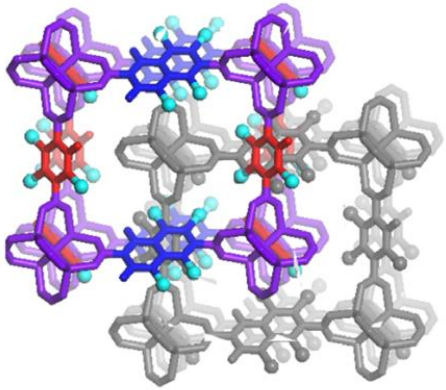
Process-Materials Co-Design Approaches

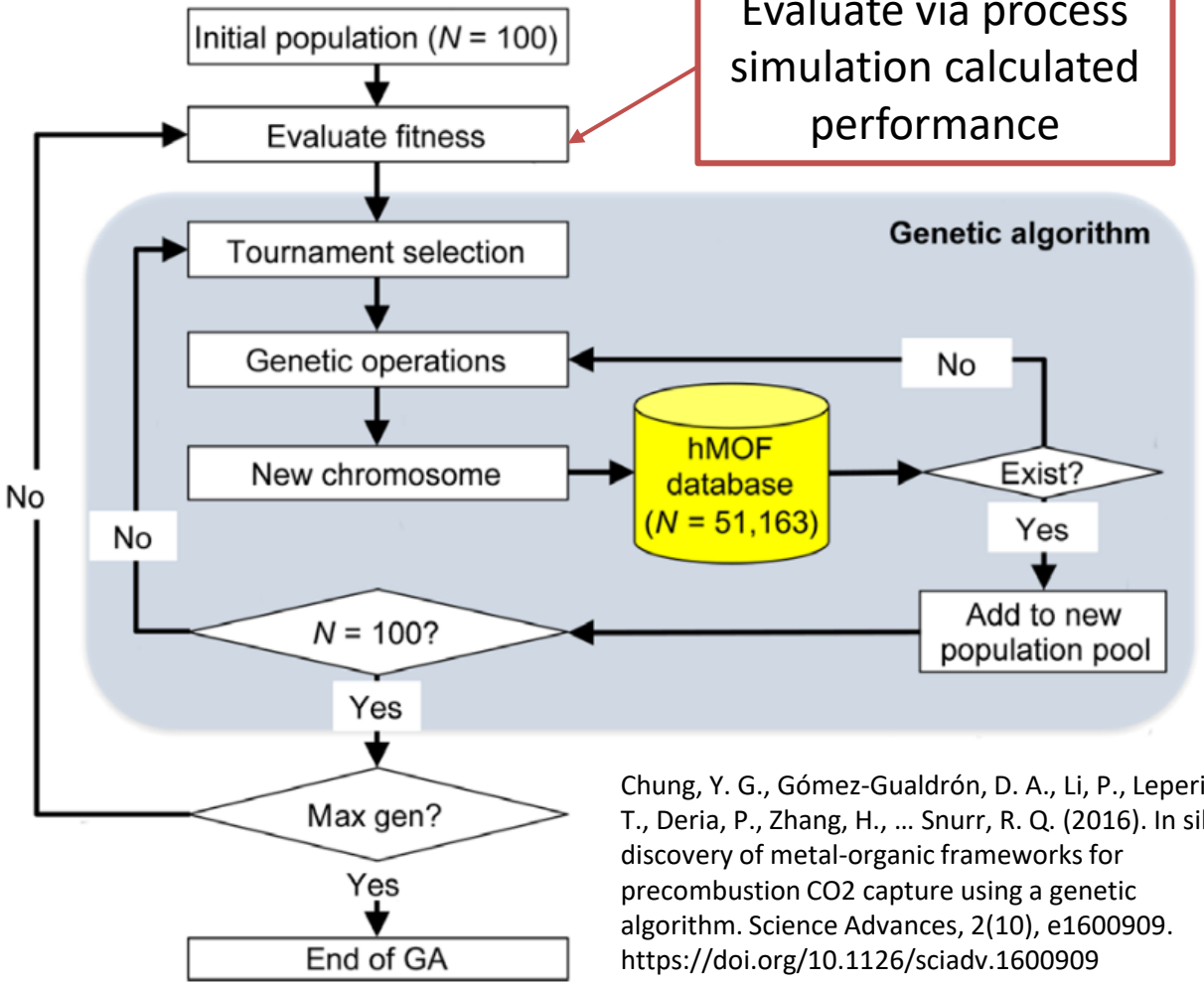
- Descriptor-based guided sampling with process simulator

Describe materials design space via geometrical decomp.



1	→	Interpenetration capacity (0–3)
1	→	Actual interpenetration (0–3)
0	→	Inorganic node (0–4)
9	→	Primary organic linker (0–39)
13	→	Secondary organic linker (0–39)
1	→	Functional group (0–14)

Evaluate via process simulation calculated performance

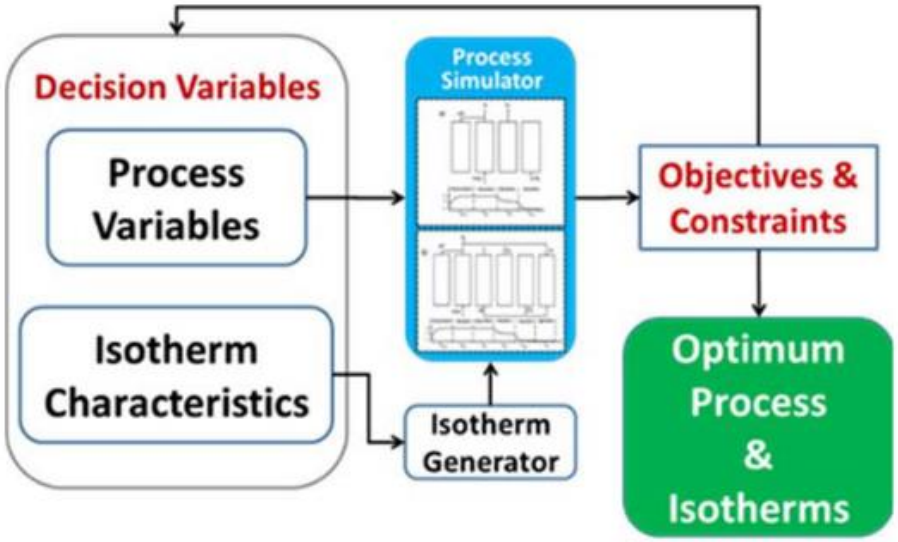


Chung, Y. G., Gómez-Gualdrón, D. A., Li, P., Leperi, K. T., Deria, P., Zhang, H., … Snurr, R. Q. (2016). In silico discovery of metal-organic frameworks for precombustion CO2 capture using a genetic algorithm. Science Advances, 2(10), e1600909. https://doi.org/10.1126/sciadv.1600909

Attempts on Materials-Process Co-optimization

- Process optimization with property model parameters as decision variables

Describe materials design space via parameters in property models (e.g., adsorption isotherm)



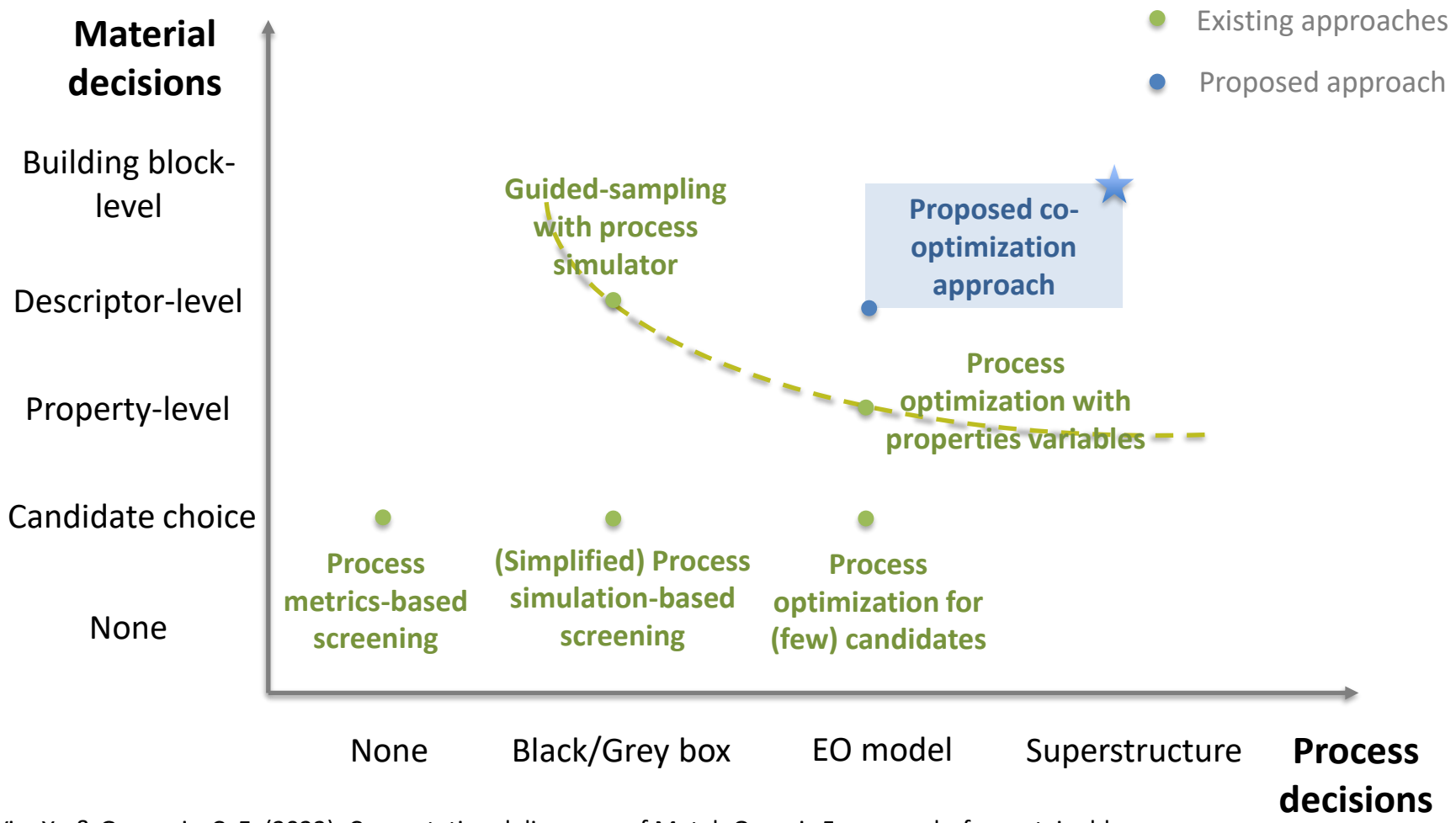
Evaluate via rigorous process optimization

	Four-Step Cycle		Six-Step Cycle	
	Minimum energy (At 0.02 atm)	Maximum Productivity (At 0.01 atm)	Minimum Energy (At 0.05 atm)	Maximum Productivity (At 0.04 atm)
13X Zeolite	164.3	1.30	184.2	2.2
UTSA-16	128.0	2.1	153.8	4.6
Isotherm from integrated optimization	106.0	3.0	116.7	7.4

Khurana, M., & Farooq, S. (2017). Integrated adsorbent-process optimization for carbon capture and concentration using vacuum swing adsorption cycles. AIChE Journal, 63(7), 2987–2995. https://doi.org/10.1002/aic.15602

Co-Optimization Overview

- **Goal:** Push Pareto front of material-process decision fidelity



Yin, X., & Gounaris, C. E. (2022). Computational discovery of Metal–Organic Frameworks for sustainable energy systems: Open challenges. Computers & Chemical Engineering, 167, 108022.

Project Overview

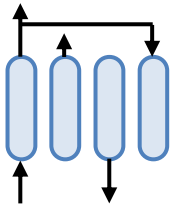
Enable the systematic search of both the materials design space and the process design space within IDAES

Step 1: Optimizable Structure-Function Relationship Surrogate

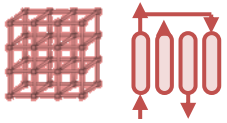
Step 2: Equation-Oriented PSA Process Model

Step 3: Co-Optimization Strategies

Surrogate Model



PSA Process Model

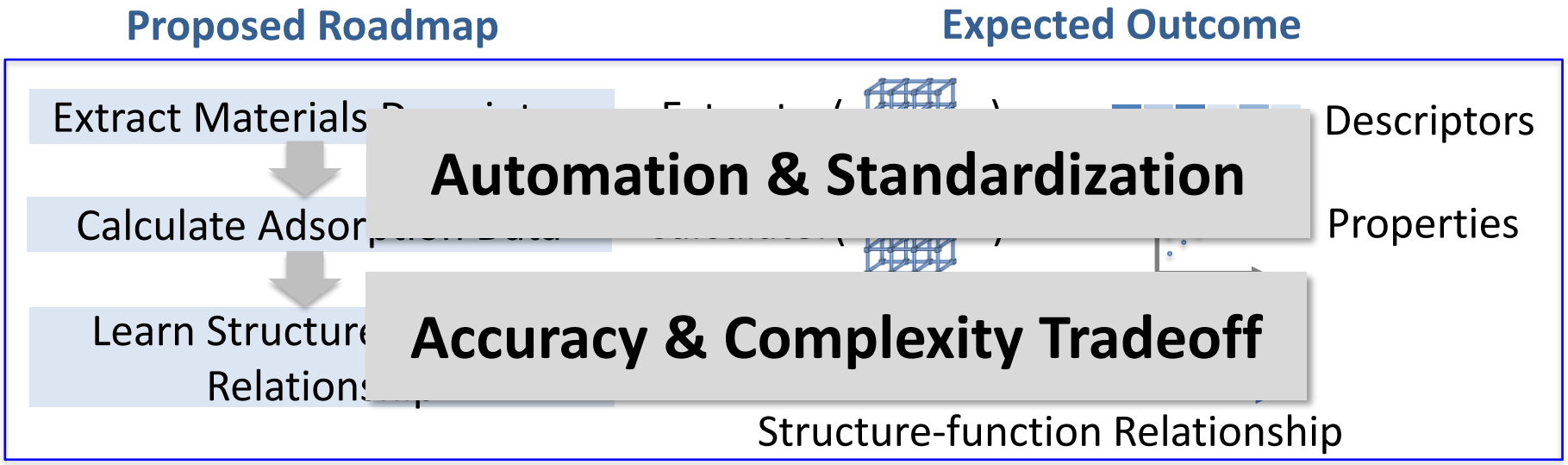


Optimal Process & Material Design

- Focus on the pressure swing adsorption (PSA) and MOFs for post/pre-combustion CO_2 capture applications while keeping methodologies and implementation general

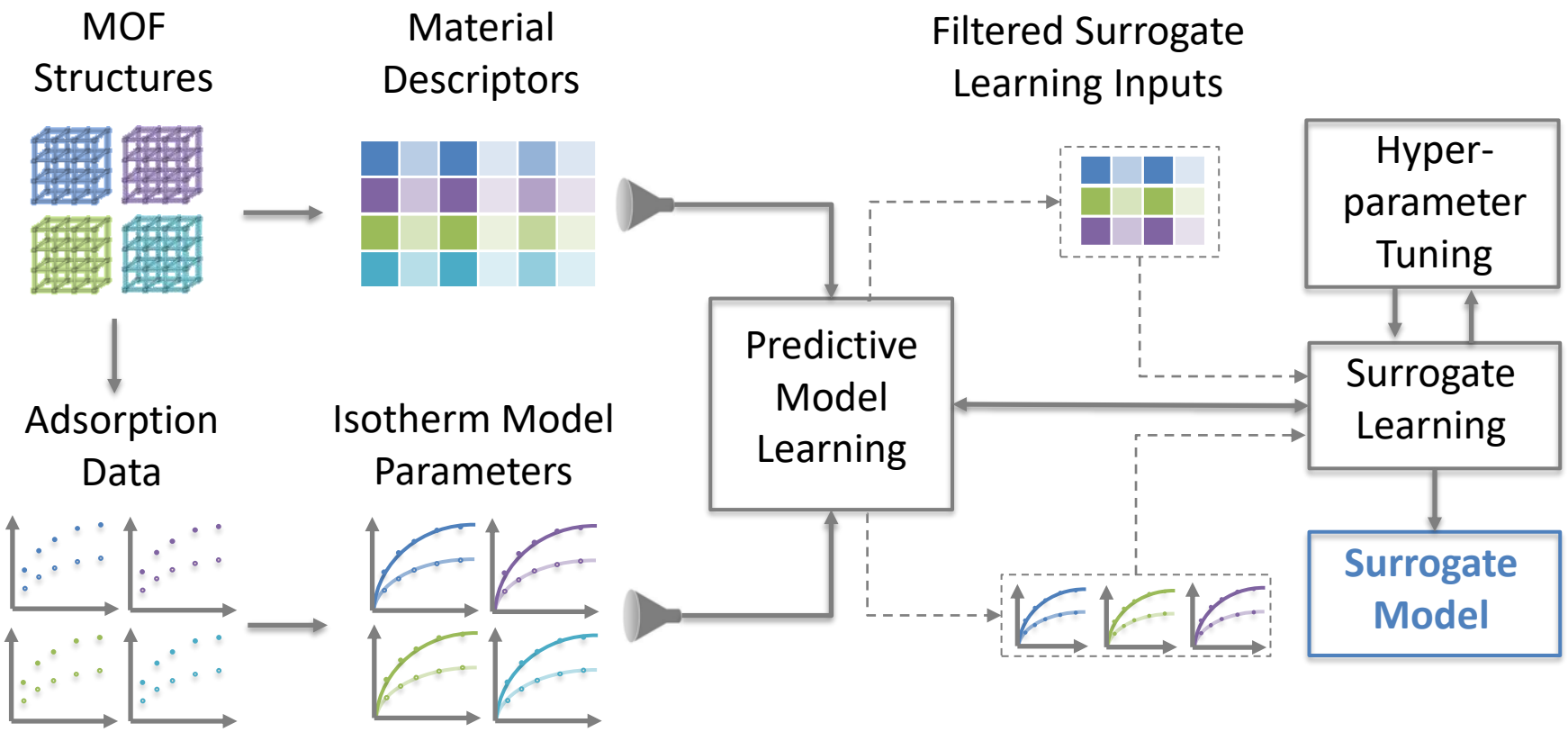
University Coal Research Program
Award DE-FE0032069

Proposed Process-Materials Co-Optimization Roadmap



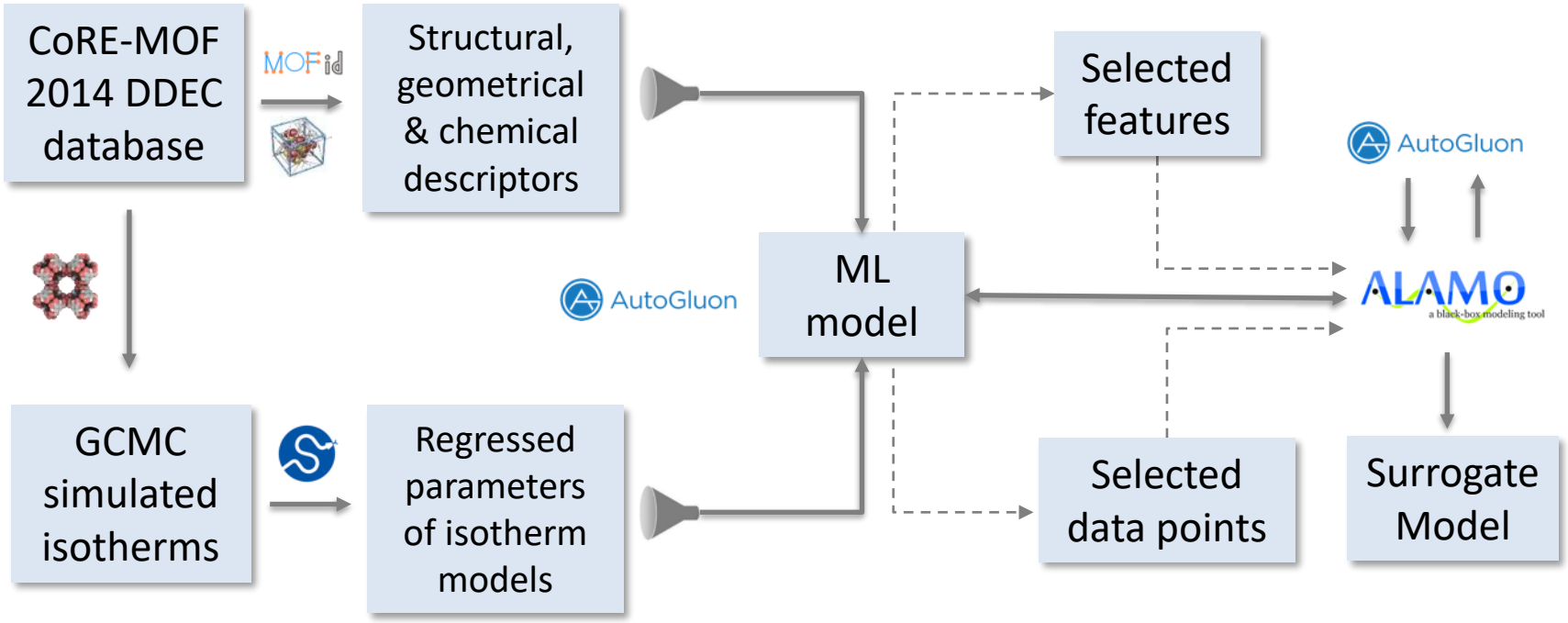
ML-Assisted Surrogate Learning

- Automatic surrogate learning pipeline developed
- Predictive ML model improves surrogate modeling efficiency and helps learn better surrogate via feature selection and data filtration



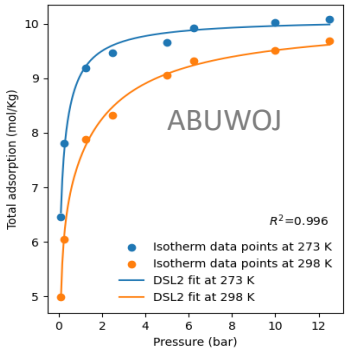
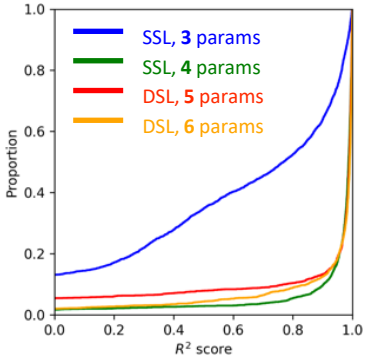
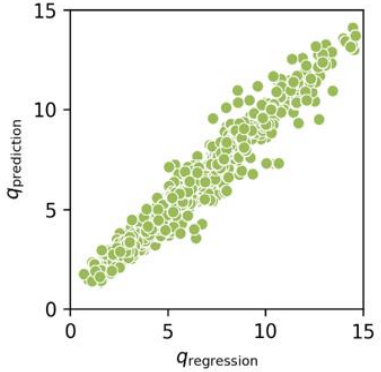
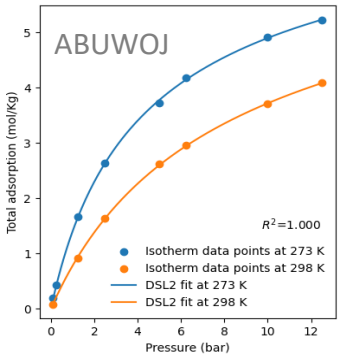
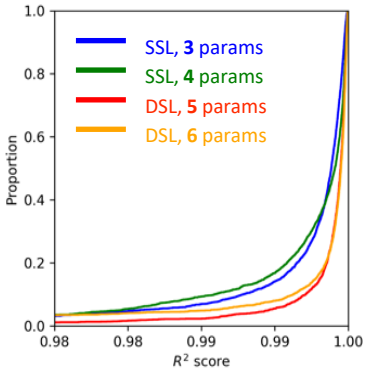
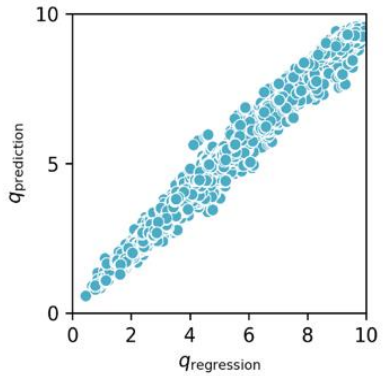
ML-Assisted Surrogate Learning

- Case study: the CoRE-MOF 2014 DDEC database
  - Compute adsorption data with Grand Canonical Monte Carlo simulations
  - Compute various types of materials descriptors
  - Learn predictive regression model with the AutoML tool
  - Learn surrogate model with the IDAES-PSE surrogate tool



ML-Assisted Surrogate Learning

- Case study: the CoRE-MOF 2014 DDEC database

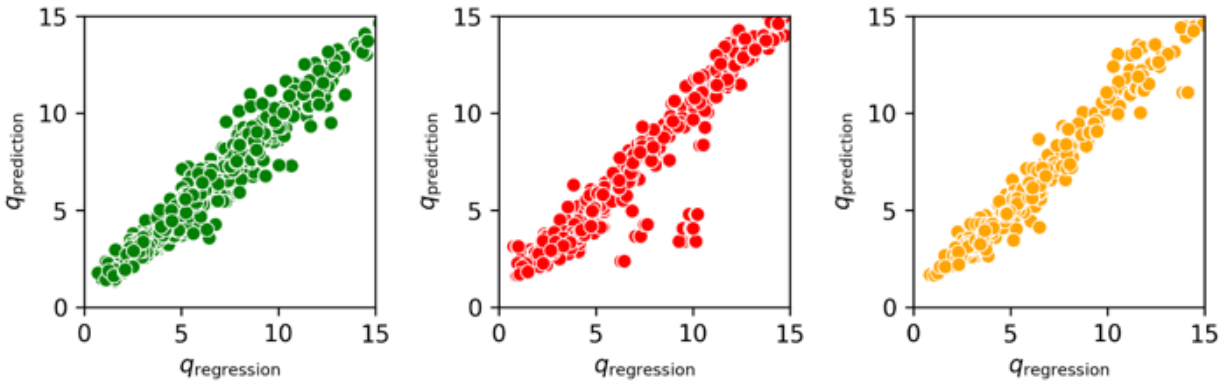
	Example DSL isotherm fitting	Cumulative distribution of regression R2 scores	Simulated and ML model predicted loadings parity plots
CO_2			
N_2			

ML-Assisted Surrogate Learning

- Case study: the CoRE-MOF 2014 DDEC database

Simulated and surrogate model predicted loadings parity plots

CO_2



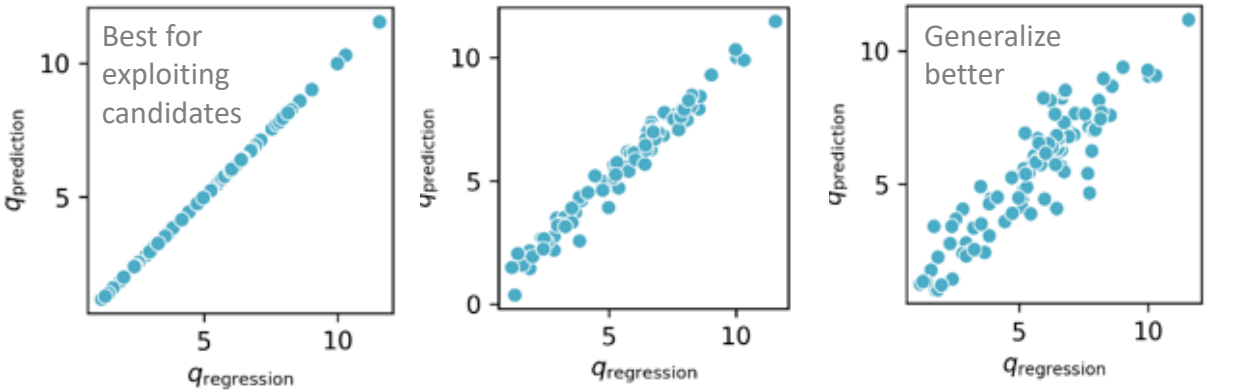
SSL, **4** params

DSL, **5** params

DSL, **6** params

Surrogate models for various isotherm forms

N_2



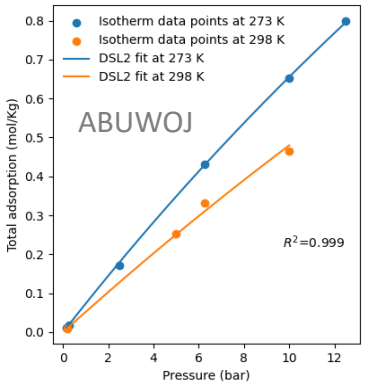
Sum of square errors | Mallow's Cp | Bayesian information criterion

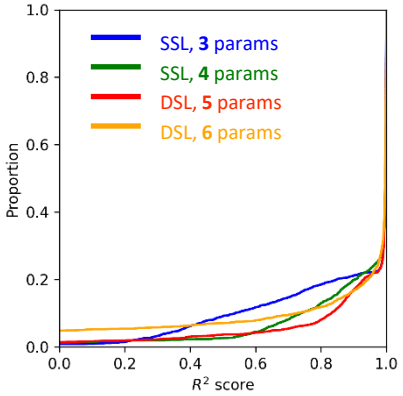
Surrogate models for different learning objectives

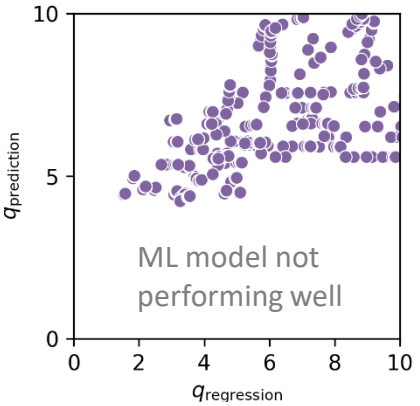
ML-Assisted Surrogate Learning

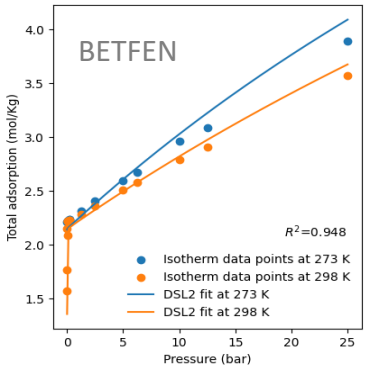
- Case study: the CoRE-MOF 2014 DDEC database

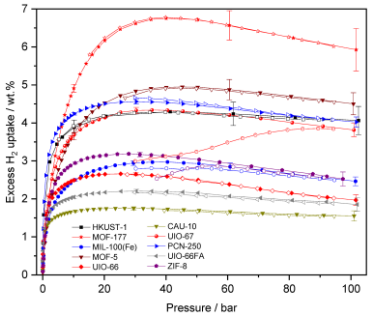
H_2



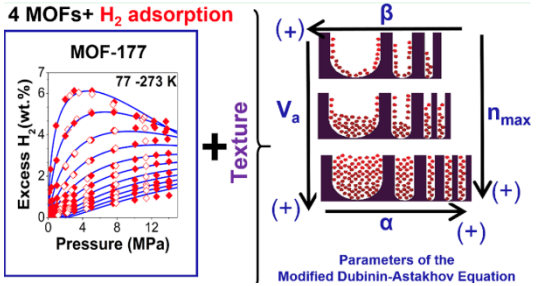








Villajos et. al. 2009



Ramirez-Vida et. al. 2022

Sharp turning point observed

Very steep slope at low pressure region

Alternative temperature dependent isotherm equation needed

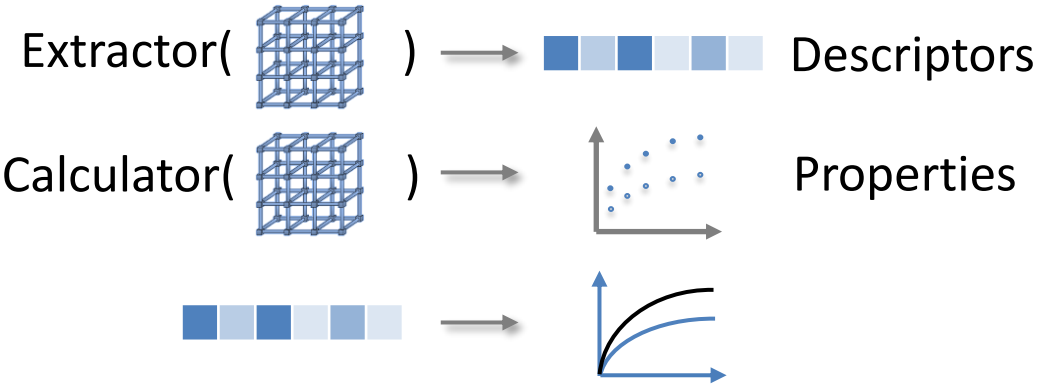
Proposed Process-Materials Co-Optimization Roadmap

Proposed Roadmap

- Extract Materials Descriptors
- Calculate Adsorption Data
- Learn Structure-function Relationship
- Model Adsorption Column
- Desi…

IDAES standards

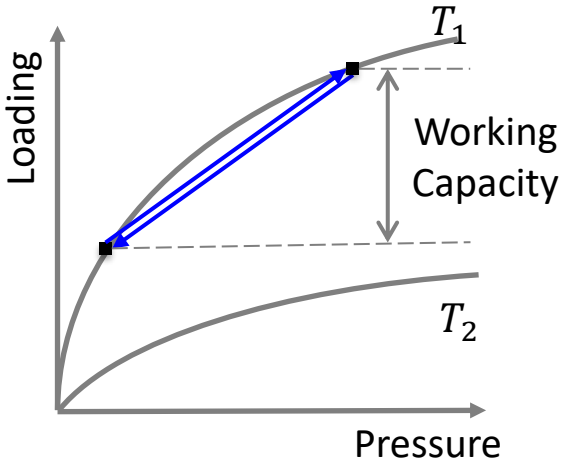
Expected Outcome



Fidelity-tunable

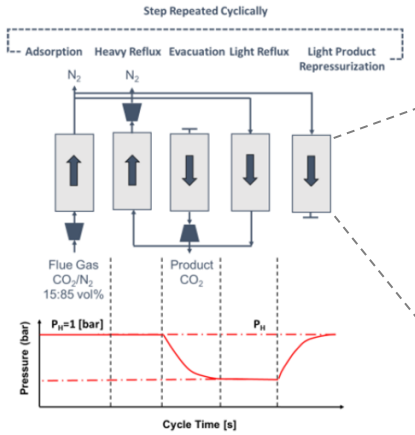
Pressure Swing Adsorption

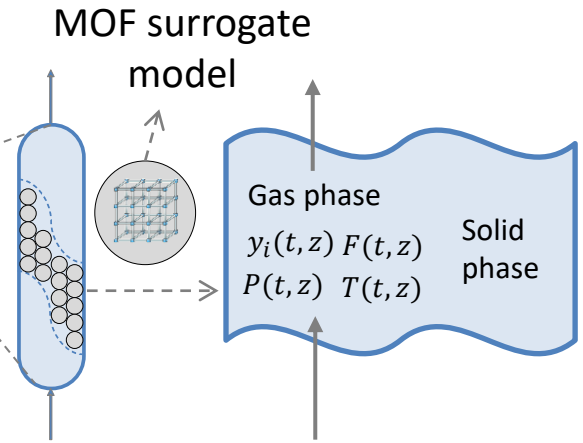
- Pressure difference drives the adsorption
- Continuous adsorption gas separation relies on cyclic operations
- Model columns as 1D dynamic packed-beds
  - No radial gradients → 1D phenomena
  - Rigid adsorbents → fixed solid phase
  - Equilibrium-controlled adsorption → isotherm models



https://www.linde-engineering.com/en/images/

5-bed 5-step cycle

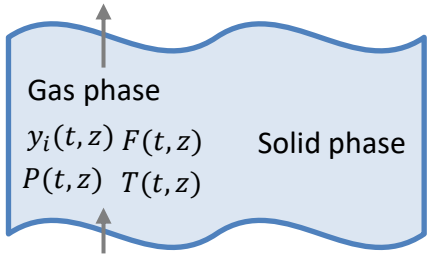


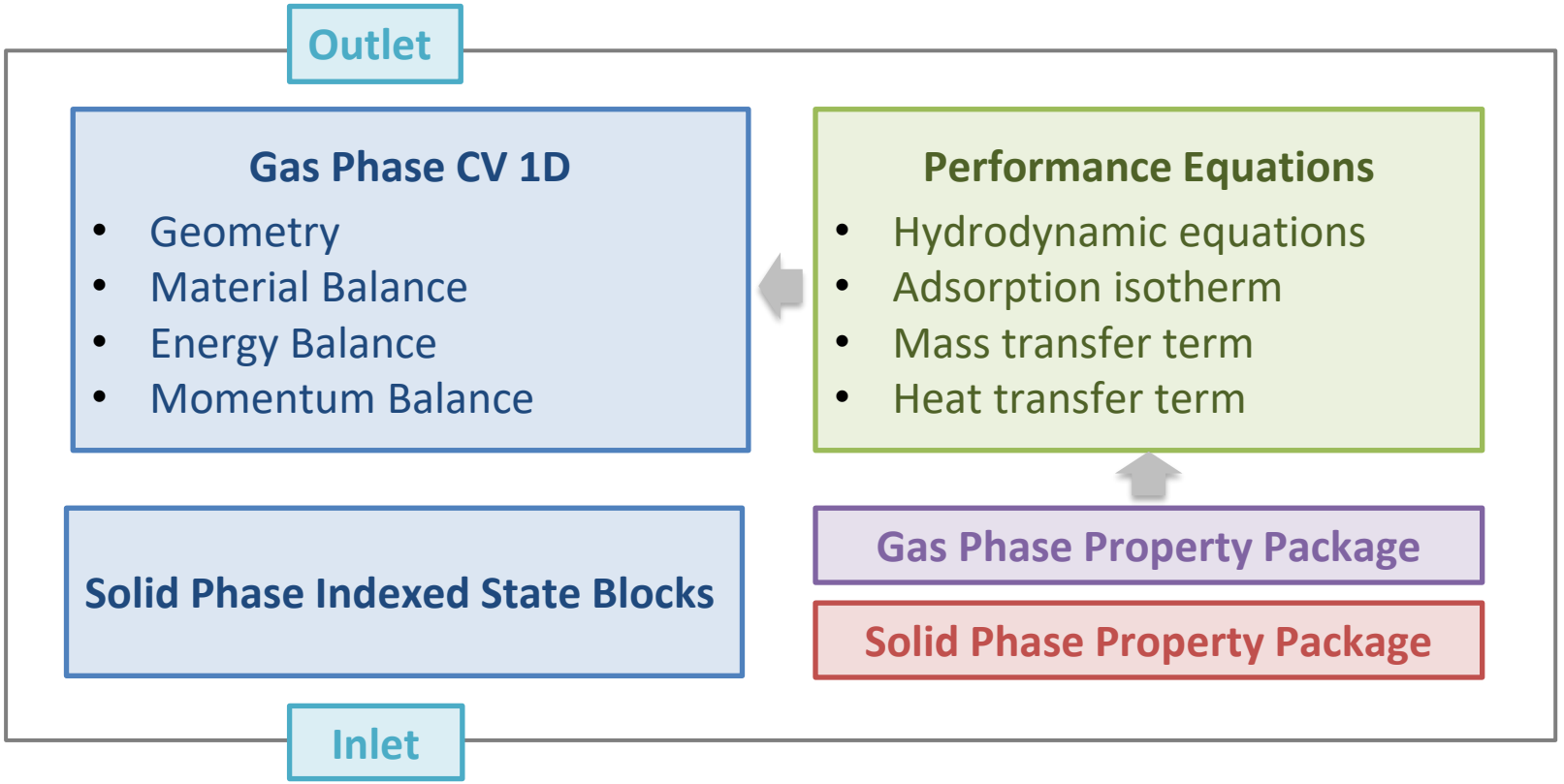


Column conceptual model

IDAES-PSE Standard Process Model

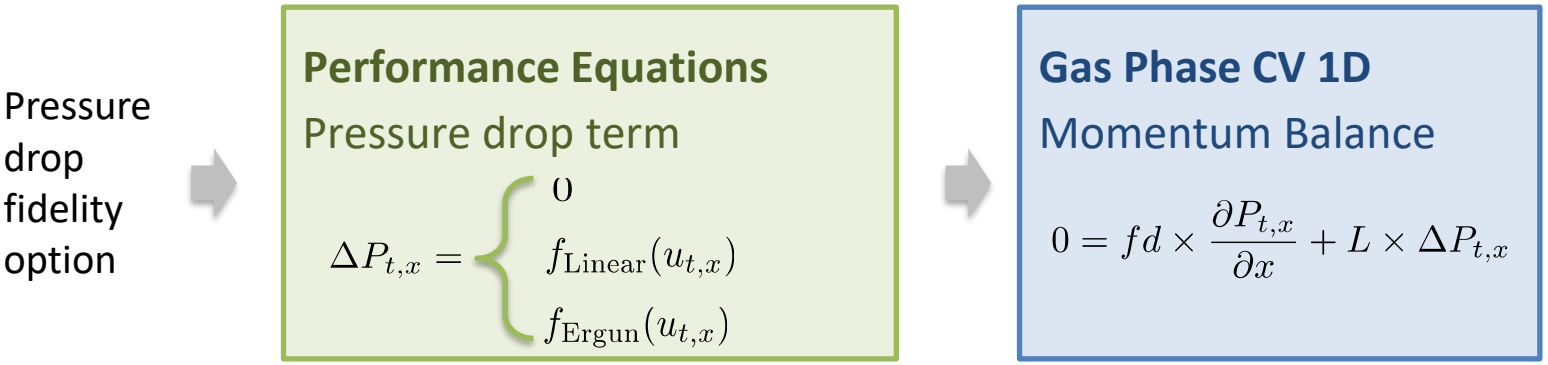
- Custom 1D packed bed unit model developed
- Implementation follows IDAES standards





IDAES-PSE Standard Process Model

- Give users control over the fidelity of the model
- Implementation takes advantage of the built-in configuration options and the modular architecture of IDAES-PSE

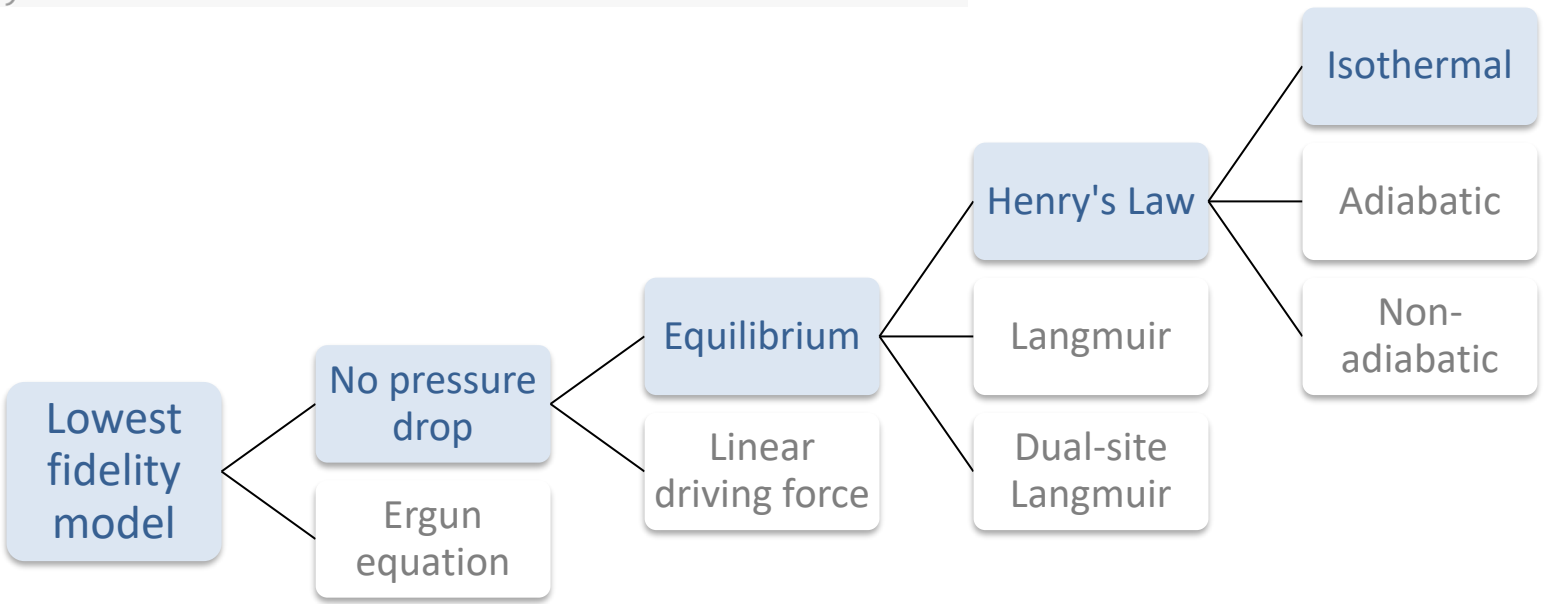


- 5 fidelity options result in 36 column fidelity levels
  - **Momentum**: No pressure drop/linear pressure drop/Ergun equation
  - **Heat transfer**: isothermal/Adiabatic/Diabatic with heat loss
  - **Mass transfer**: Immediate adsorption equilibrium/Linear driving force
  - **Isotherm**: Henry's law/Single-site Langmuir/Dual-site Langmuir

IDAES-PSE Standard Process Model

- Lowest fidelity model

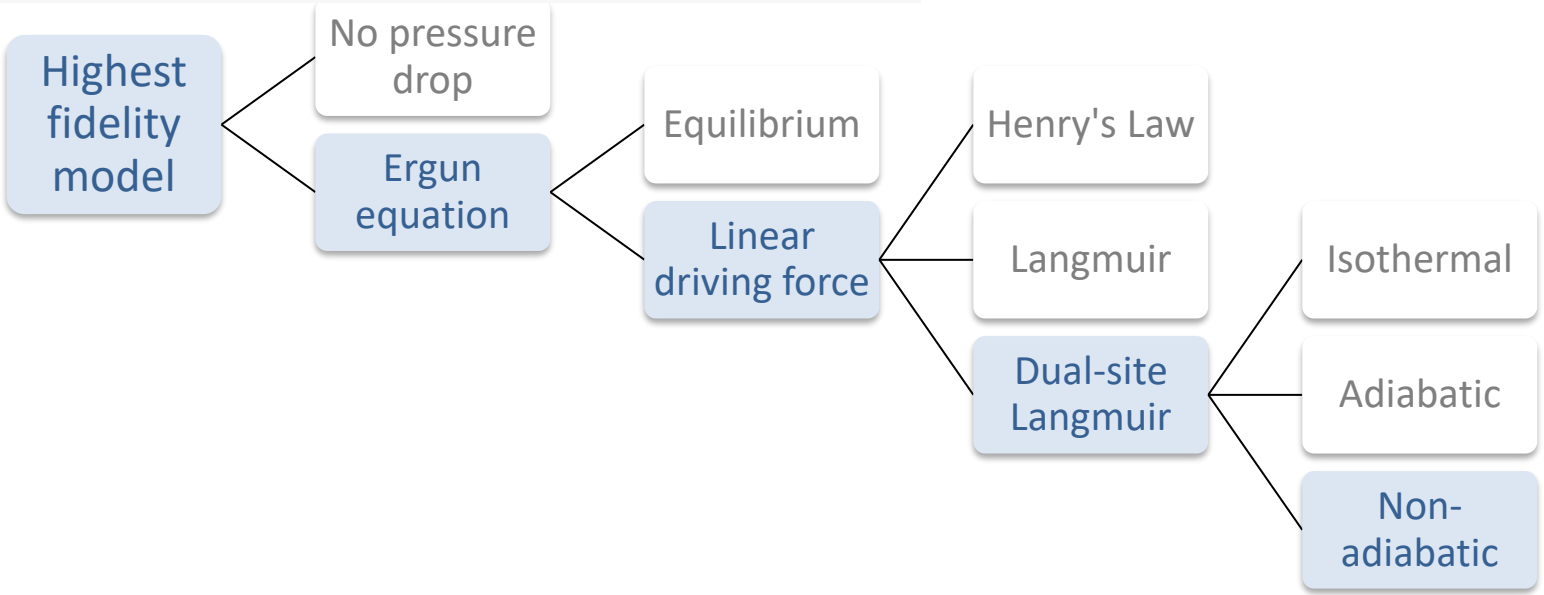
```
m.fs.PB = PackedBed1D(
    finite_elements=nxfe,
    step=step_name,
    has_heat_generation=False,
    has_heat_loss=False,
    pressure_drop_type=None,
    mass_transfer_type=None,
    isotherm_type="henry",
    gas_phase_config={"property_package": m.fs.gas_properties},
    solid_phase_config={"property_package": m.fs.solid_properties},
)
```



IDAES-PSE Standard Process Model

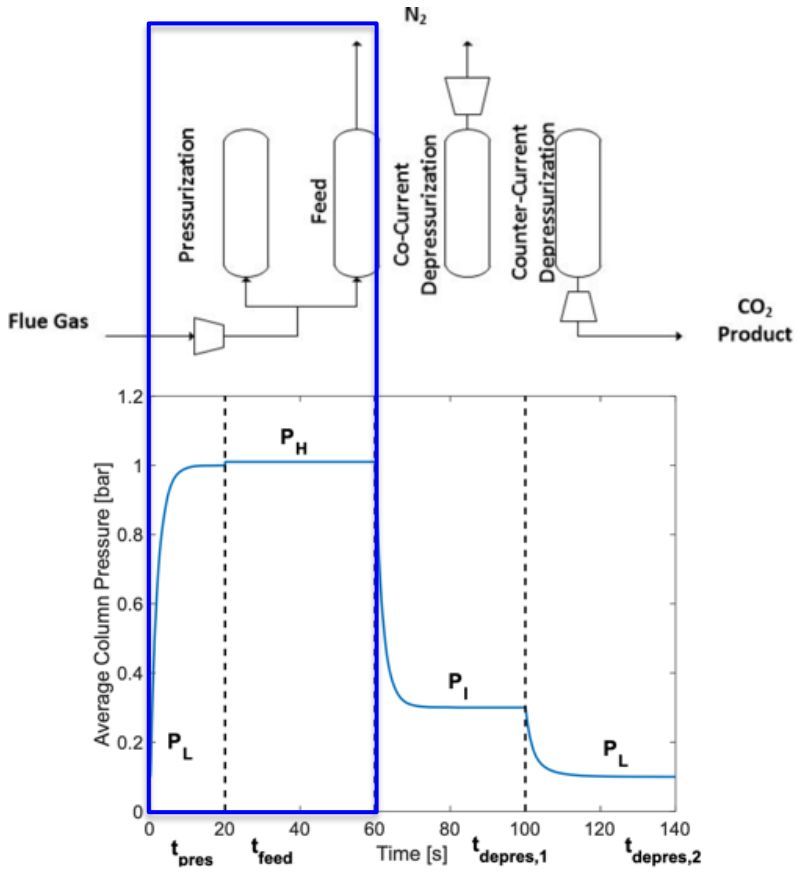
- Highest fidelity model

```
m.fs.PB = PackedBed1D(
    finite_elements=nxfe,
    step=step_name,
    has_heat_generation=True,
    has_heat_loss=True,
    pressure_drop_type="ergun_correlation",
    mass_transfer_type="linear_driving_force",
    isotherm_type="dsl",
    gas_phase_config={"property_package": m.fs.gas_properties},
    solid_phase_config={"property_package": m.fs.solid_properties},
)
```



IDAES-PSE Standard Process Model

- Case study: first two steps in the FVPSA cycle

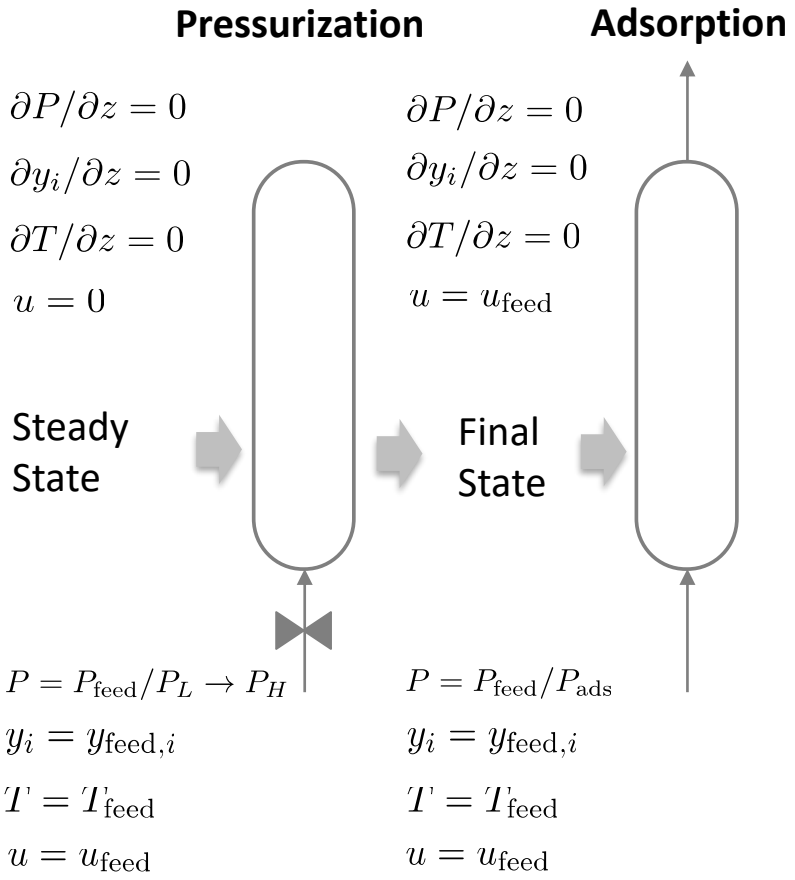


Leperi, K. T., Chung, Y. G., You, F., & Snurr, R. Q. (2019). Development of a general evaluation metric for rapid screening of adsorbent materials for postcombustion CO2 capture. *ACS Sustainable Chemistry & Engineering*, *7*(13), 11529-11539.

- Discretization:
  - Pyomo.DAE capability that is built in IDEAS-PSE
  - **Length domain**: Single-point collocation method, CSTR approximation to handle "steep front"
  - **Time domain**: Backward finite differences

IDAES-PSE Standard Process Model

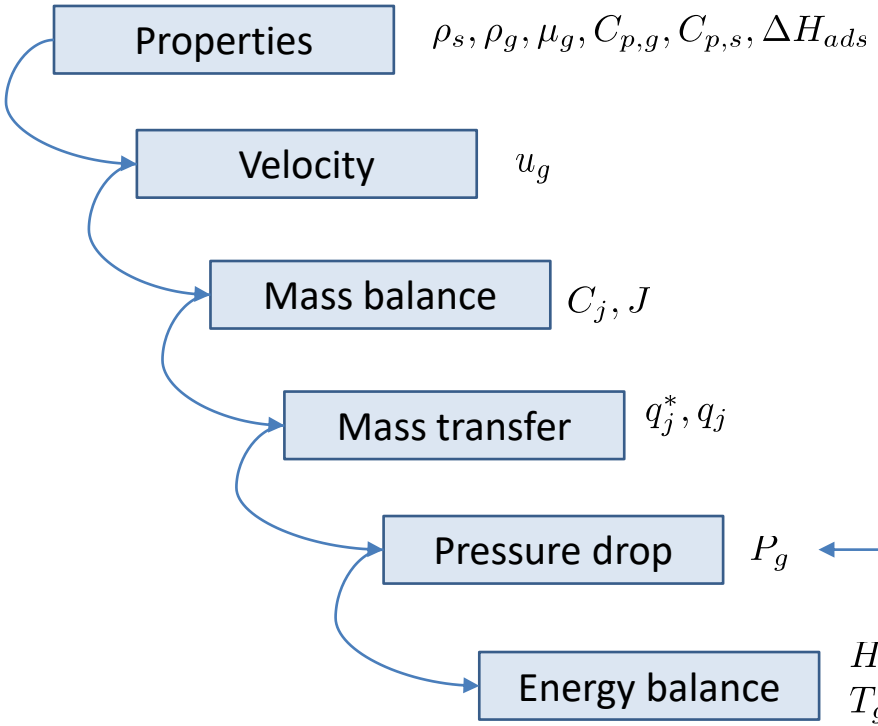
- Case study: first two steps in the FVPSA cycle



- Boundary conditions:
  - **Pressurization**: feed at inlet, outlet valve closed
  - **Adsorption**: feed at inlet, steady production at outlet
- Initial conditions:
  - **Pressurization**: steady state (no accumulations)
  - **Adsorption**: pressurization final state (solve t_0 block)

IDAES-PSE Standard Process Model

- Case study: first two steps in the FVPSA cycle



Approximation -> Ergun

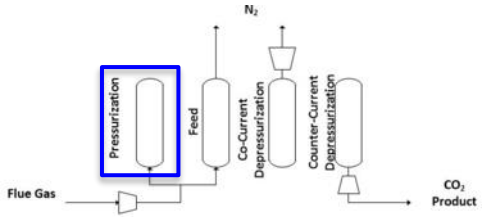
gas energy accumulation: off -> on

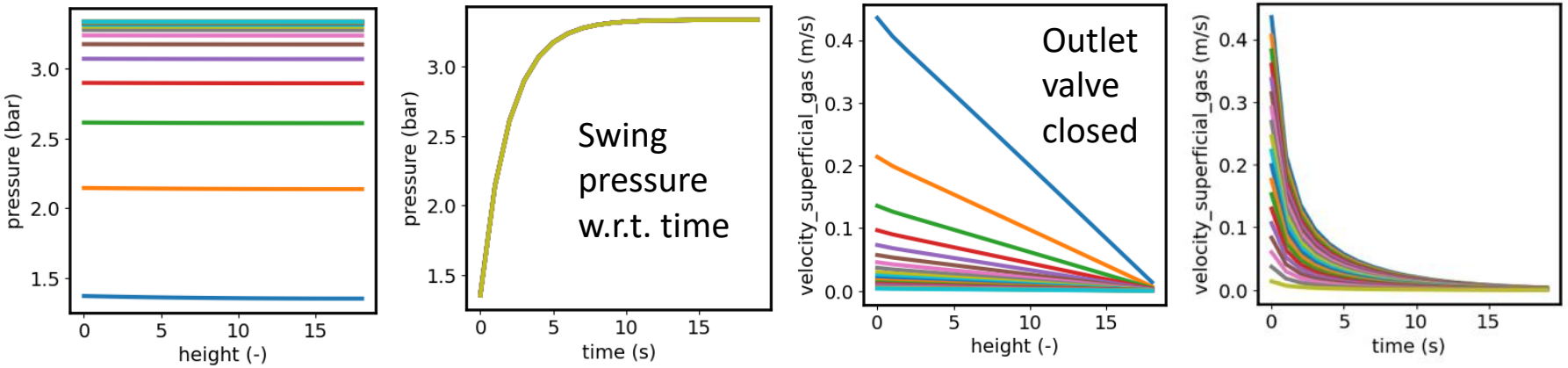
Cascade initialization routine

- Scaling: IDAES-PSE template
- Initialization:
  - Cascade initialization using IPOPT solving discretized NLP
- Simulation:
  - Backward Euler via IDEAS-PSE PETSc interface

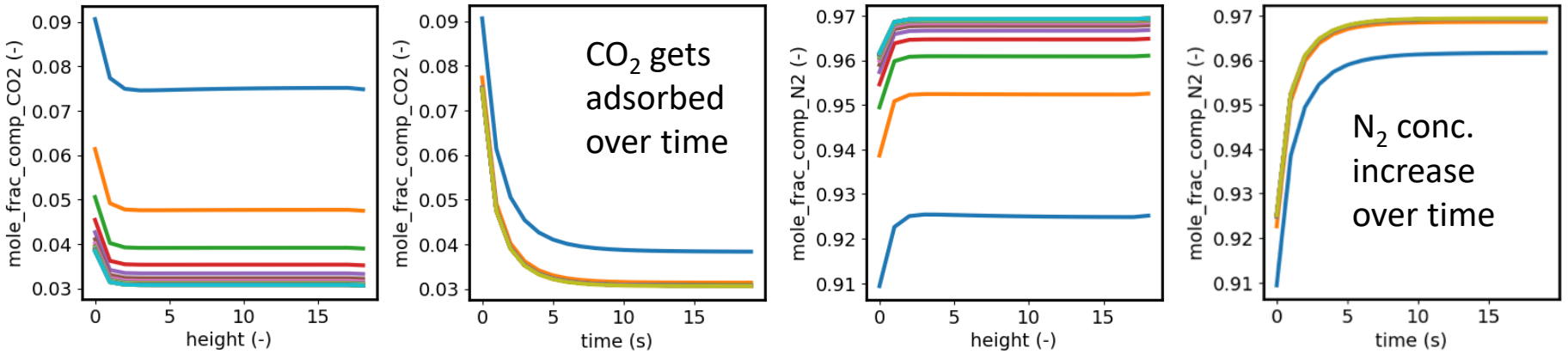
IDAES-PSE Standard Process Model

- Case study: FVPSA cycle pressurization step





Pressure profiles **Velocity profiles**

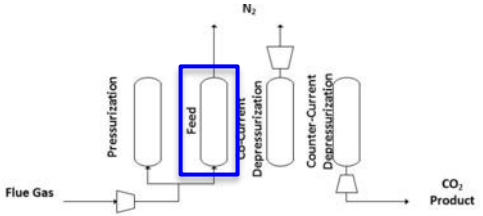


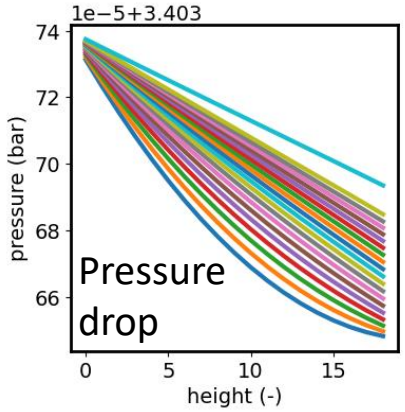
CO2 mole fraction profile **N2 mole fraction profile**

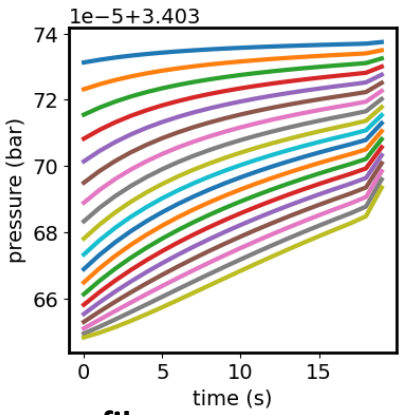
First element — Final element

IDAES-PSE Standard Process Model

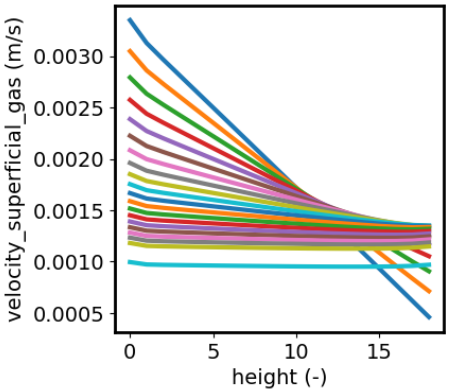
- Case study: FVPSA cycle adsorption step

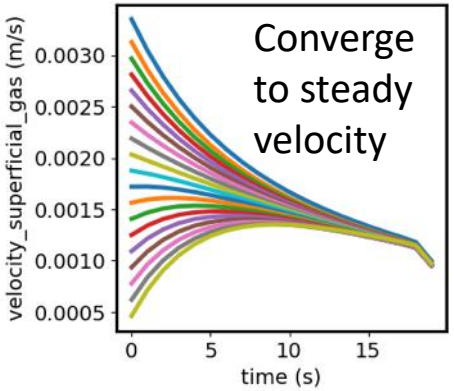




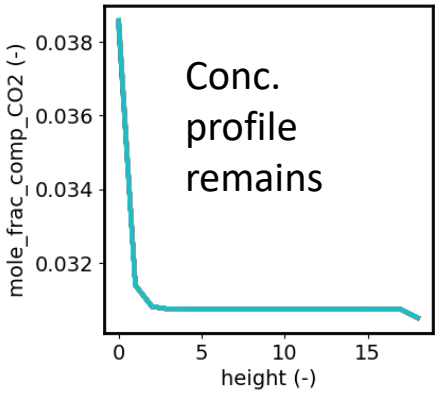


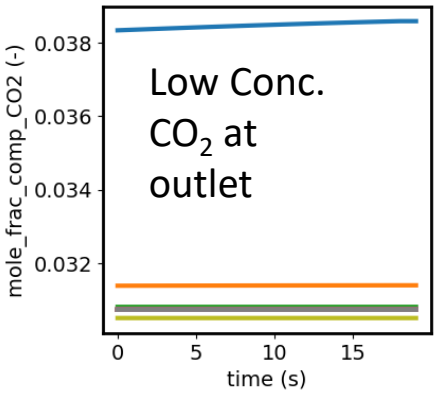
Pressure profiles



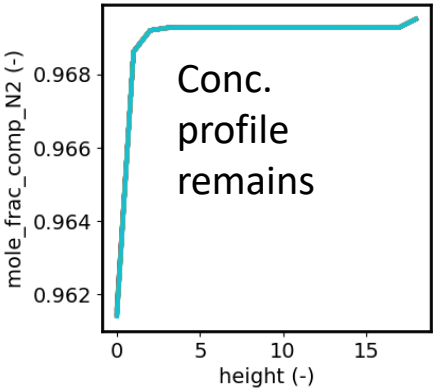


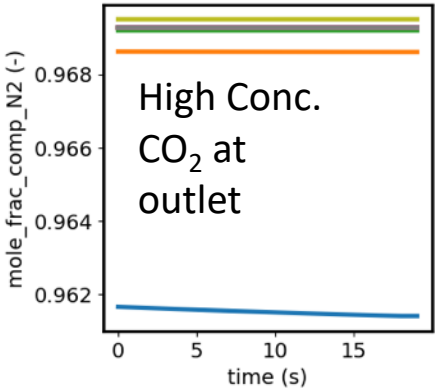
Velocity profiles





CO2 mole fraction profile





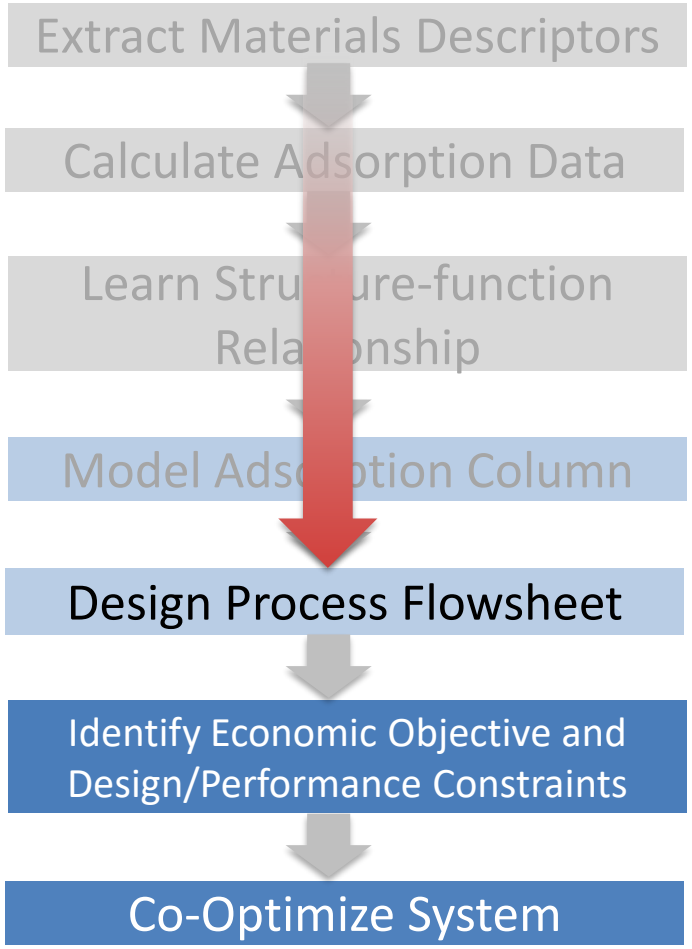
N2 mole fraction profile

First element

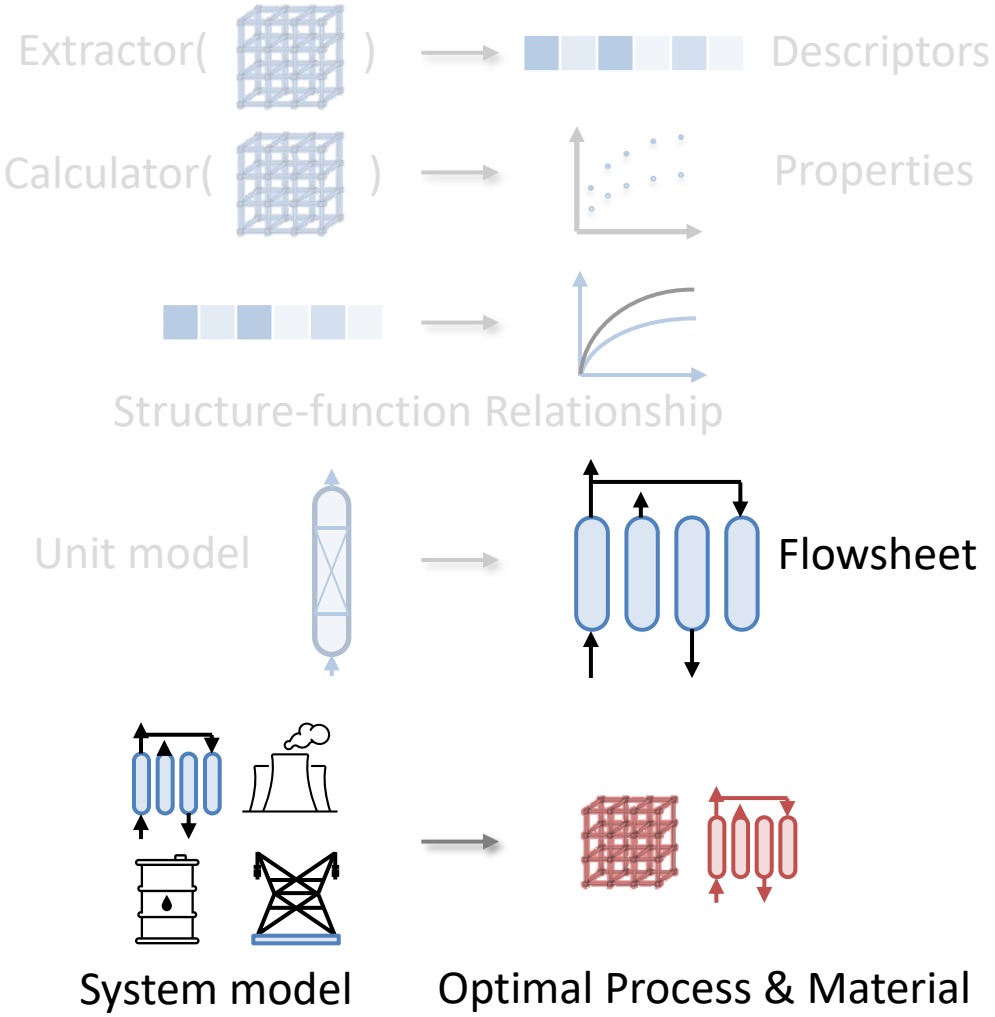
Final element

Next Step: PSA Cycle Simulation & Optimization

Proposed Roadmap



Expected Outcome



Conclusions

- Opportunities exist in incorporating **materials** design with **process** design/operation in a **co-optimization framework**
- A **PSA process with MOF adsorbents co-optimization** workflow is implemented within the IDAES-PSE integrated platform
  - **Optimizable MOF structure-function relationships** were learned via a custom-built ML-assisted surrogate learning workflow
  - **Fidelity-tunable PSA Column unit models** were developed and will be contributed to the IDEAS-PSE model library

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