Pressure Swing Adsorption: Design and Optimization for Pre-Combustion Carbon Capture

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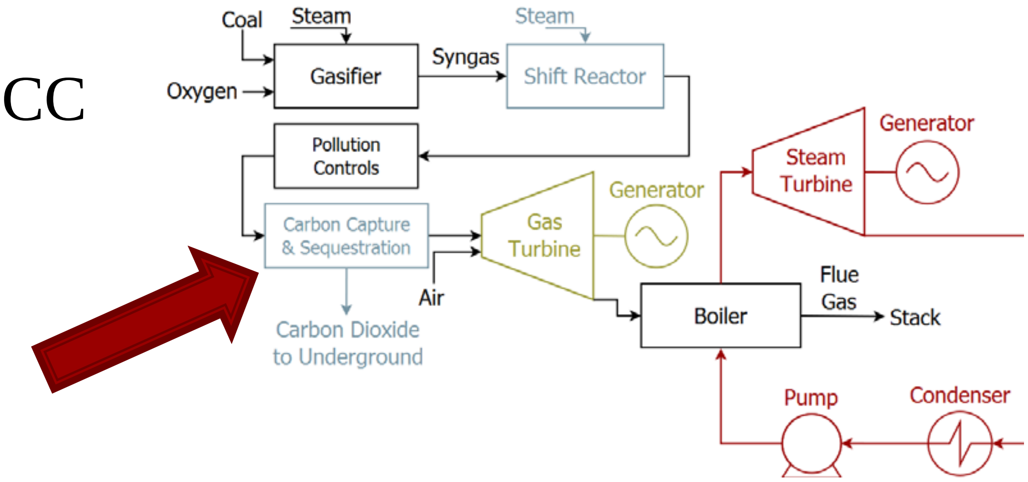
Research Objectives

- Demonstrate methods for optimal Pressure Swing Adsorption (PSA) process synthesis
- Design **cost effective** PSA cycle for H_2-CO_2 separation in IGCC power plant

Simplified IGCC Flowsheet

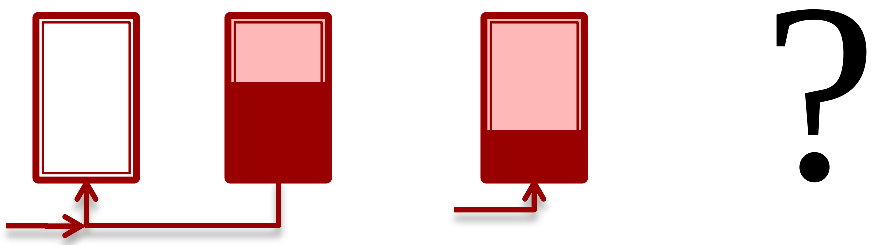


Pressure Swing Adsorption (PSA)

- Gas separation utilizing differences in adsorption phenomena
- Adsorption at high pressure, desorption at low pressure

- Numerous industrial examples
  - H_2 purification in refineries
  - O_2 concentration for medical use

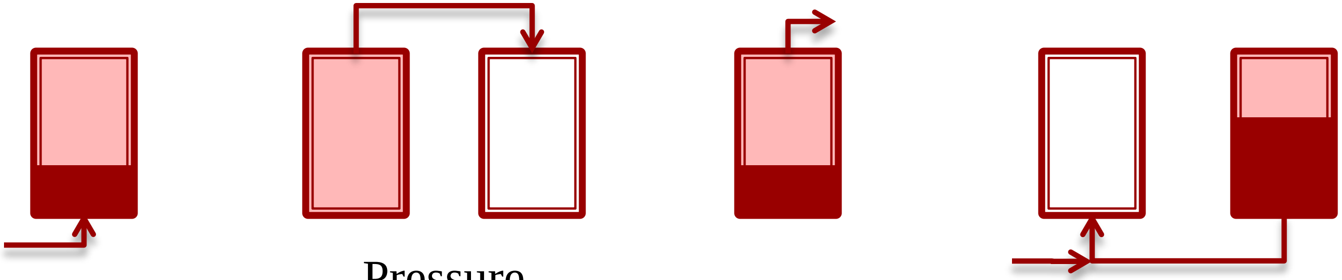
Optimal Cycle Synthesis



Discrete variables make this **too computationally expensive** to solve

Step 1　　Step 2　　Step 3　　Step 4　　…　　Step n

"Parts Box" of Steps



Adsorption

Pressure Equalization

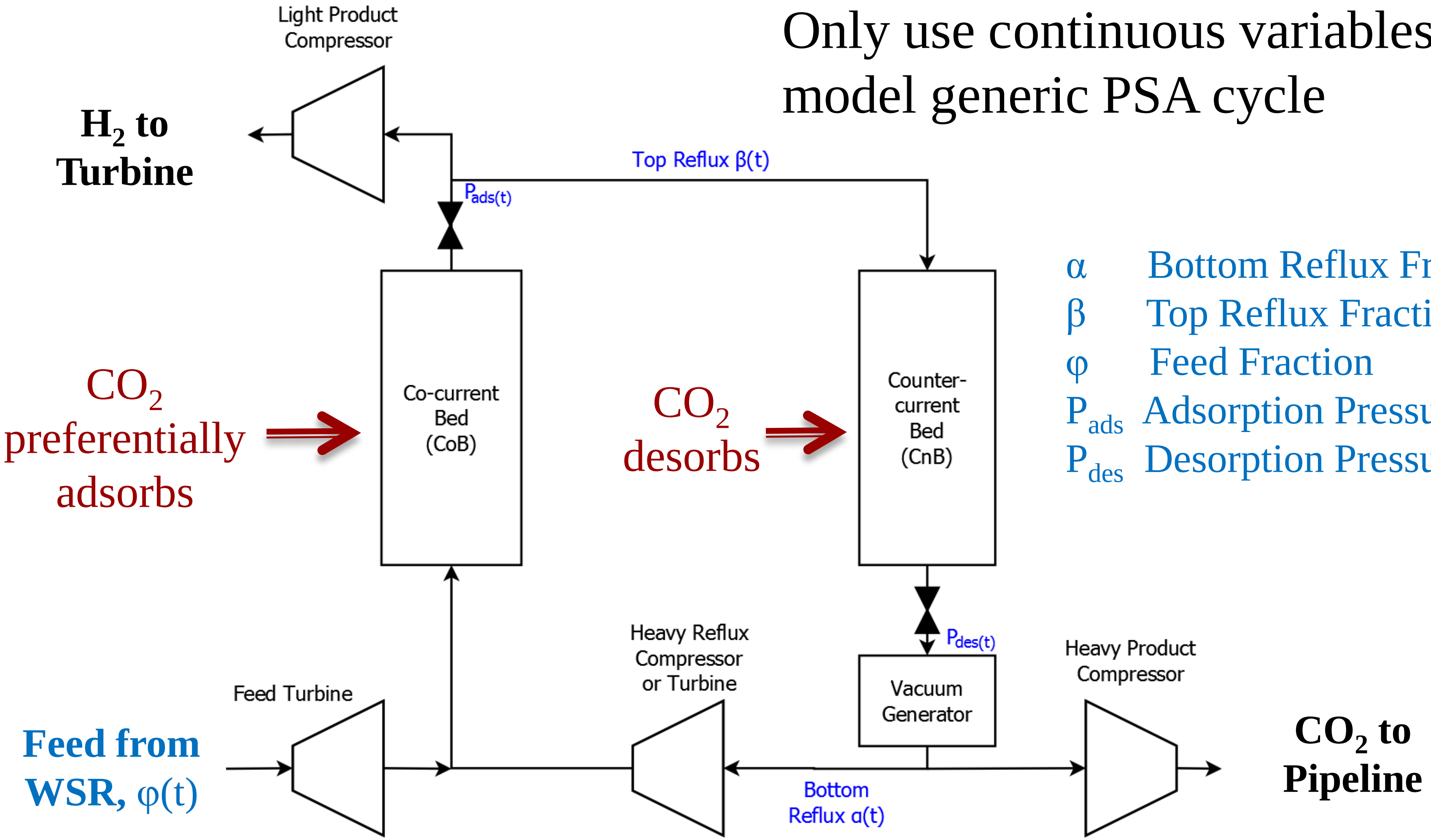
Desorption

Heavy Product Purge

And many more…

PSA “Superstructure”

Only use continuous variables to model generic PSA cycle



H_2 to Turbine

CO_2 preferentially adsorbs

CO_2 desorbs

Feed from WSR, φ(t)

CO_2 to Pipeline

α	Bottom Reflux Fraction
β	Top Reflux Fraction
φ	Feed Fraction
P_{ads}	Adsorption Pressure
P_{des}	Desorption Pressure

PSA Model: Transport Equations

Momentum (Ergun Equation)

$$-\frac{\partial P}{\partial x} = \frac{150\mu(1-\epsilon_b)^2}{d_p^2\epsilon_b^3}v + \frac{1.75}{d_p}\left(\frac{1-\epsilon_b}{\epsilon_b^3}\right)\left(\sum_i M_w^i C_i\right)v|v|$$

$$v_j(t,x) \leftarrow \begin{cases} \max(0, v_j(t,x)) & \text{if} \quad j=1 \quad \text{(co-cur. bed)} \\ \min(0, v_j(t,x)) & \text{if} \quad j=2 \quad \text{(counter-cur. bed)} \end{cases}$$

Energy

$$0 = \left(\epsilon_t \sum_i C_i(C_{pg}^i - R) + \rho_s C_{ps}\right)\frac{\partial T}{\partial t} - \rho_s \sum_i \Delta H_i^{ads}\frac{\partial q_i}{\partial t} + \frac{\partial(vh)}{\partial x} + UA(T-T_w)$$

Material

$$\epsilon_b \frac{\partial C_i}{\partial t} + (1-\epsilon_b)\rho_s\frac{\partial q_i}{\partial t} + \frac{\partial(vC_i)}{\partial x} = \cancel{D_L \frac{\partial^2 C_i}{\partial x^2}} \qquad i = 1 \ldots N_c$$

PSA Model: Adsorption

Linear Driving Force

$$\frac{\partial q_i}{\partial t} = k_i(q_i^\star - q_i) \qquad i = 1 \ldots N_c$$

Dual-Site Langmuir Isotherm

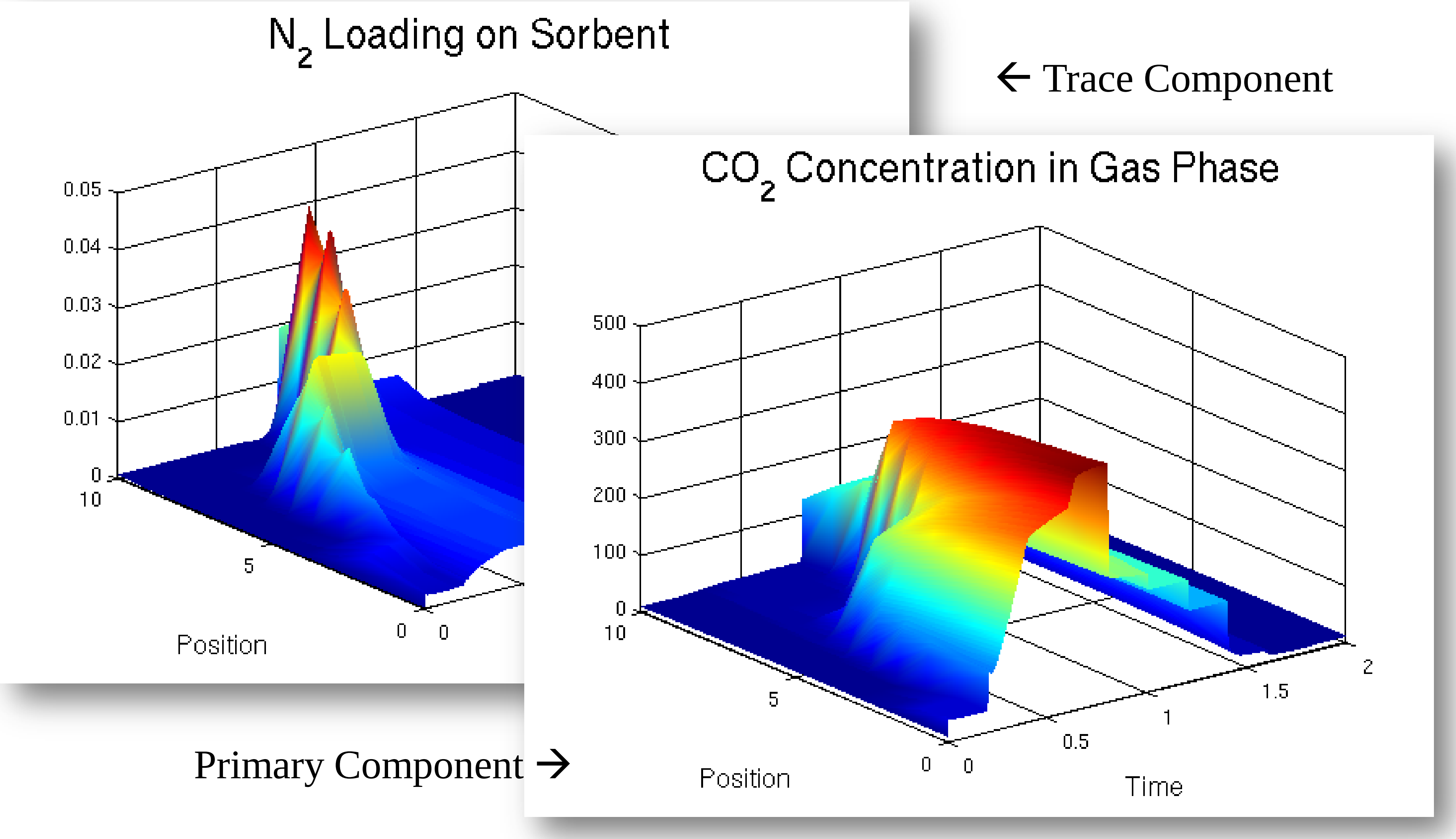
$$q_i^\star = \frac{q_1^s b_{1i} P_i}{1 + \sum_j b_{1j} P_j} + \frac{q_2^s b_{2i} P_i}{1 + \sum_j b_{2j} P_j} \qquad i = 1 \ldots N_c$$

where $q_{mi}^s = k_{mi}^1 + k_{mi}^2 T \qquad b_{mi} = k_{mi}^3 exp(k_{mi}^4 / T) \qquad m = 1, 2$

Thermodynamics: Ideal Gas Law

Take away: **complex non-linear PDAE model**

Sample Simulation Results



N_2 Loading on Sorbent

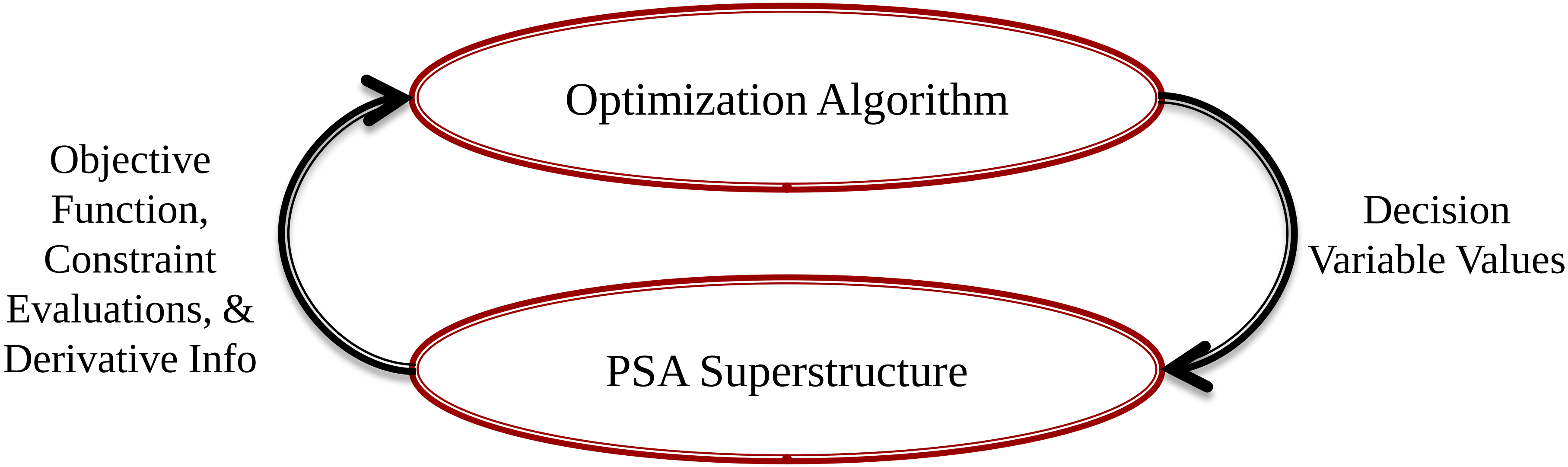
← Trace Component

CO_2 Concentration in Gas Phase

Primary Component →

Optimization Methodology

Minimize specific energy (kWh/tonne CO_2 captured)



Optimization Algorithm

Objective Function, Constraint Evaluations, & Derivative Info

Decision Variable Values

PSA Superstructure

- PSA Bed Model
- Connectivity Equations
- Compressor and Turbine Model
- Valve Equations
- **Cyclic Steady-State Constraint**

3 approaches to accommodate cyclic-steady state constraint

1. Periodic Boundary Conditions

$$z_0 \overbrace{\left[\ \underline{u_1}\quad \underline{u_2}\quad \underline{u_3}\quad \underline{u_4}\quad \underline{u_5}\ \right]}^{\text{5 Slot PSA Cycle}} z(t_f)$$

Constraint linking initial and final bed state variables

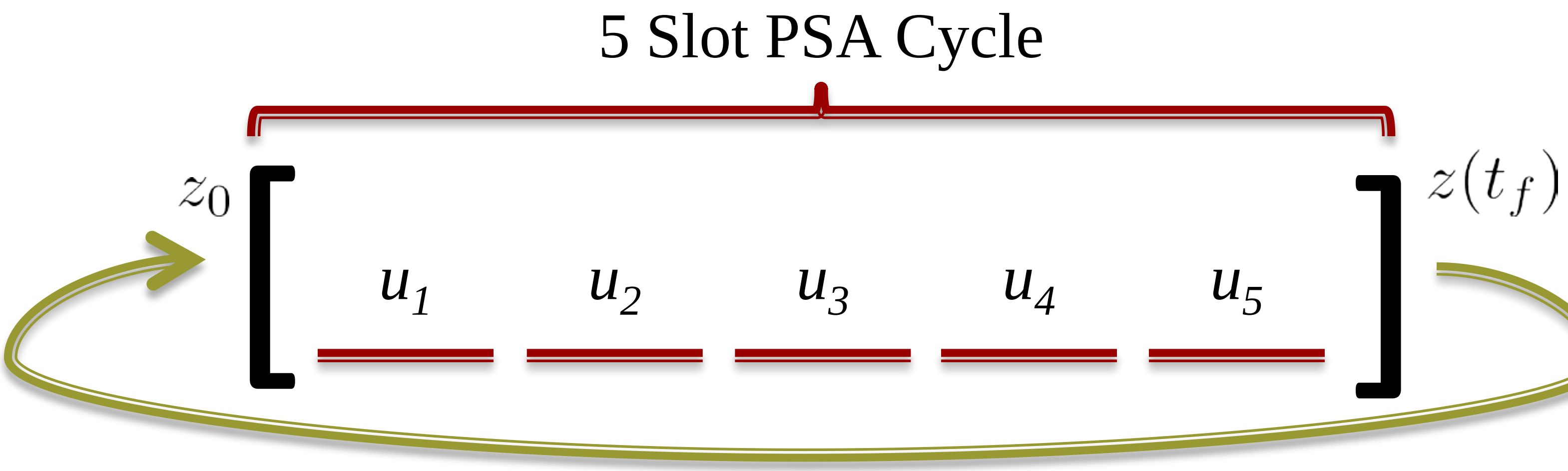
$$z_0 - z(t_f) = 0$$

+ exact and smooth → derivative based optimization algorithms

- large problem (z_0 and u_i optimization variables)

- expensive derivatives (from direct sensitivity equations)

2. Direct Substitution



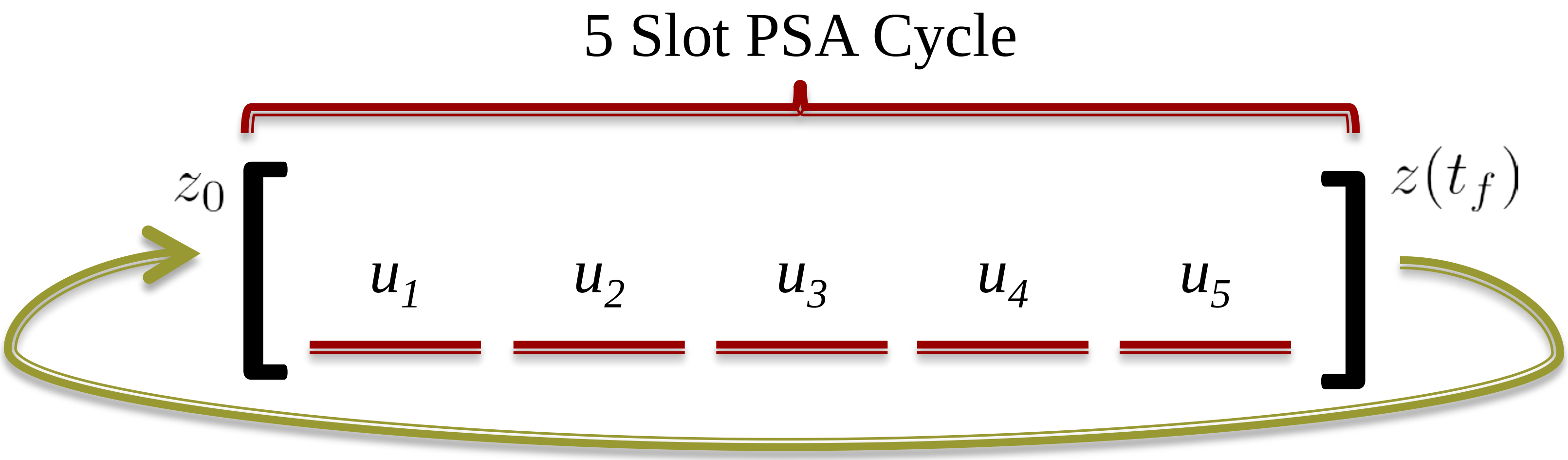
5 Slot PSA Cycle

z_0 $\left[\; \underline{u_1} \;\; \underline{u_2} \;\; \underline{u_3} \;\; \underline{u_4} \;\; \underline{u_5} \;\right]$ $z(t_f)$

Repeat direct substitution until $|z_0 - z(t_f)| < \epsilon$

+ “natural”… mimics process start-up
+ simple implementation
+ medium size problem (z_0 *not* optimization variables)
- not smooth → derivative free optimization

3. Fixed Horizon



5 Slot PSA Cycle

Repeat direct substitution a fixed number of times (M)

+ exact and smooth → derivative based optimization algorithms
+ medium size problem (z_0 *not* optimization variables)
- expensive objective function and constraint evaluations
- expensive derivatives (from adjoint sensitivity equations)

Implementation Details

- IPOPT for derivative based formulations (1, 3)
  - First derivatives from sensitivity equations
  - Second derivatives approximated with LBFGS
- BOBYQA for direct substitution (2)
  - DFO code based on quadratic approximation to objective function
  - Accommodates variable bounds

Case Study 1

Approach	Obj. Func $kWh/tonne\ CO_2$	CPU Time/Iter $h{:}mm{:}ss$	Iter
Periodic Bnd. Cnd. (1)	89.63	0:08:49	187
Derivative Free (2)	146.42	0:04:46	566
Fixed Horizon (3)	98.46	0:20:41	397

- Common far starting point
- DFO approach terminates at a much poorer solution
  - Local minima?
- Some challenges with gradient-based convergence
  - Terminate due to resource limits or integrator failure
  - Noisy first derivatives, approximate second derivatives

Case Study 2

Part A: Two Components (CO_2, H_2)

Approach	Obj. Func $kWh/tonne\ CO_2$	CPU Time/Iter *h:mm:ss*	Iter
Periodic Bnd. Cnd. (1)	83.51	0:10:37	82
Derivative Free (2)	114.21	0:03:28	1215
Fixed Horizon (3)	86.46	0:21:09	56

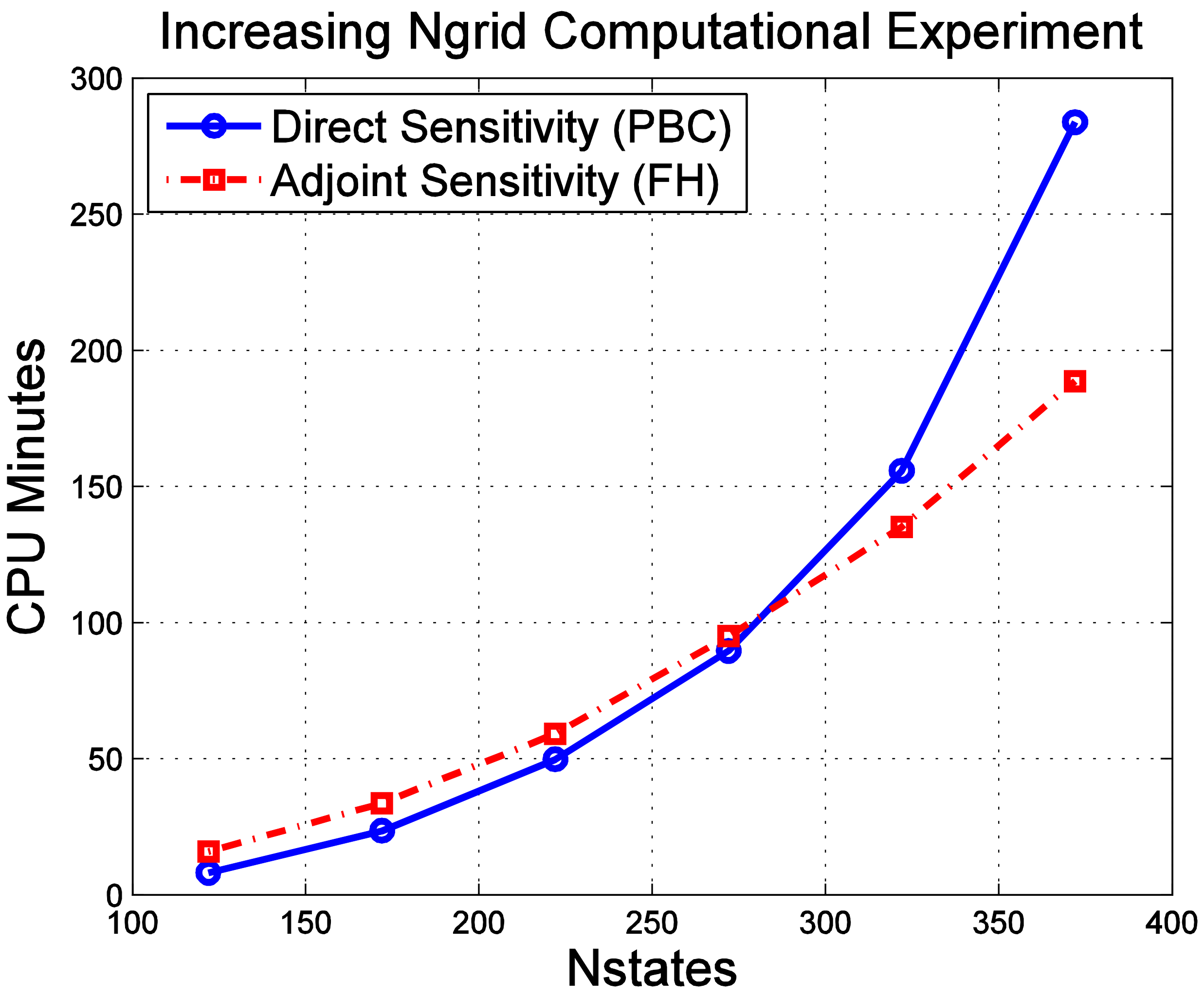
Part B: Five Components (CO_2, H_2, CH_4, N_2, CO)

Approach	Obj. Func $kWh/tonne\ CO_2$	CPU Time/Iter *h:mm:ss*	Iter
Periodic Bnd. Cnd. (1)	89.37	1:00:15	470
Derivative Free (2)	109.04	0:11:29	2500+
Fixed Horizon (3)	86.81	1:27:52	260

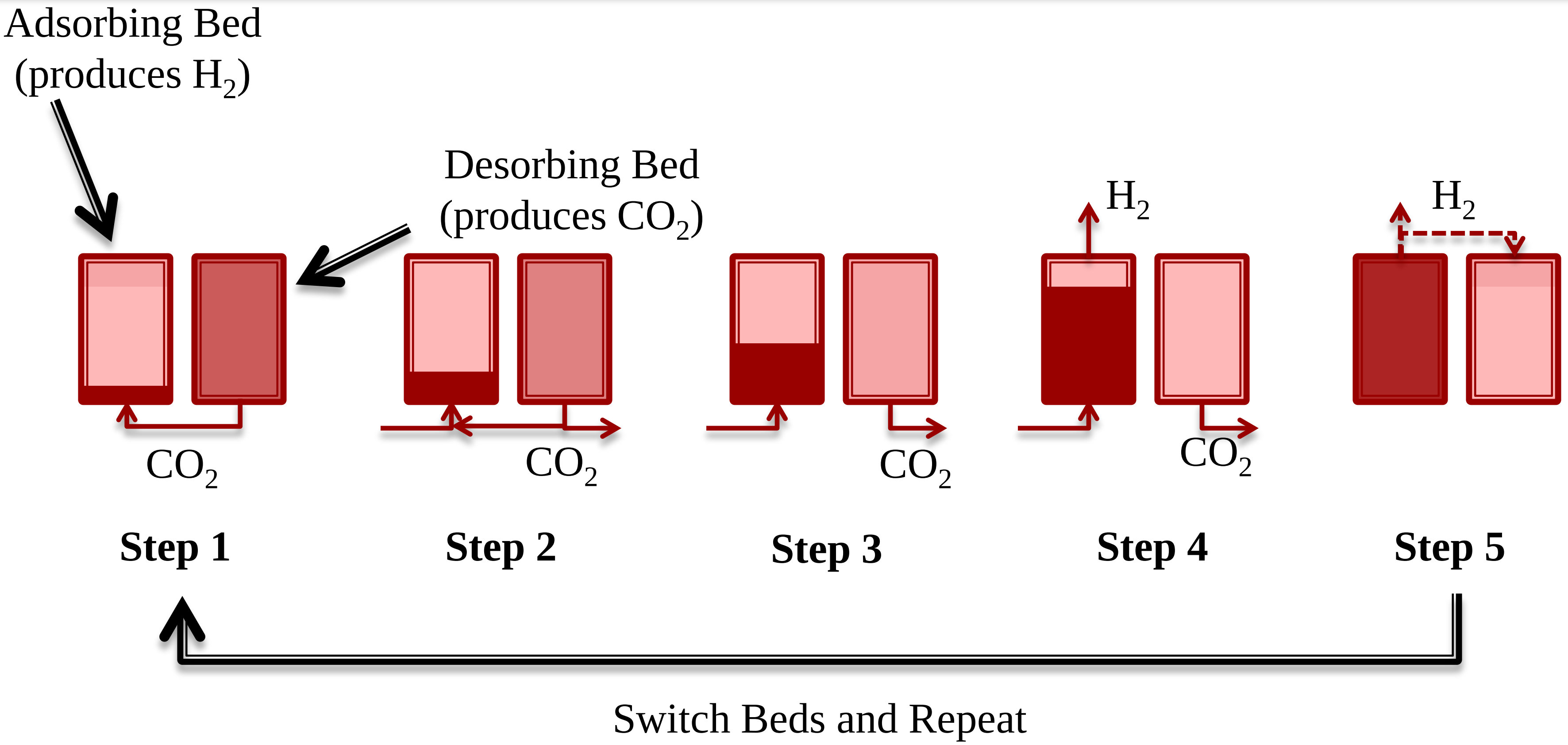
- Common near starting point
- DFO approach terminates at an infeasible solution

Problem Complexity

Adjoint sensitivity computationally adventitious for large systems



Designed Cycle



Adsorbing Bed
(produces H_2)

Desorbing Bed
(produces CO_2)

Step 1

Step 2

Step 3

Step 4

Step 5

Switch Beds and Repeat

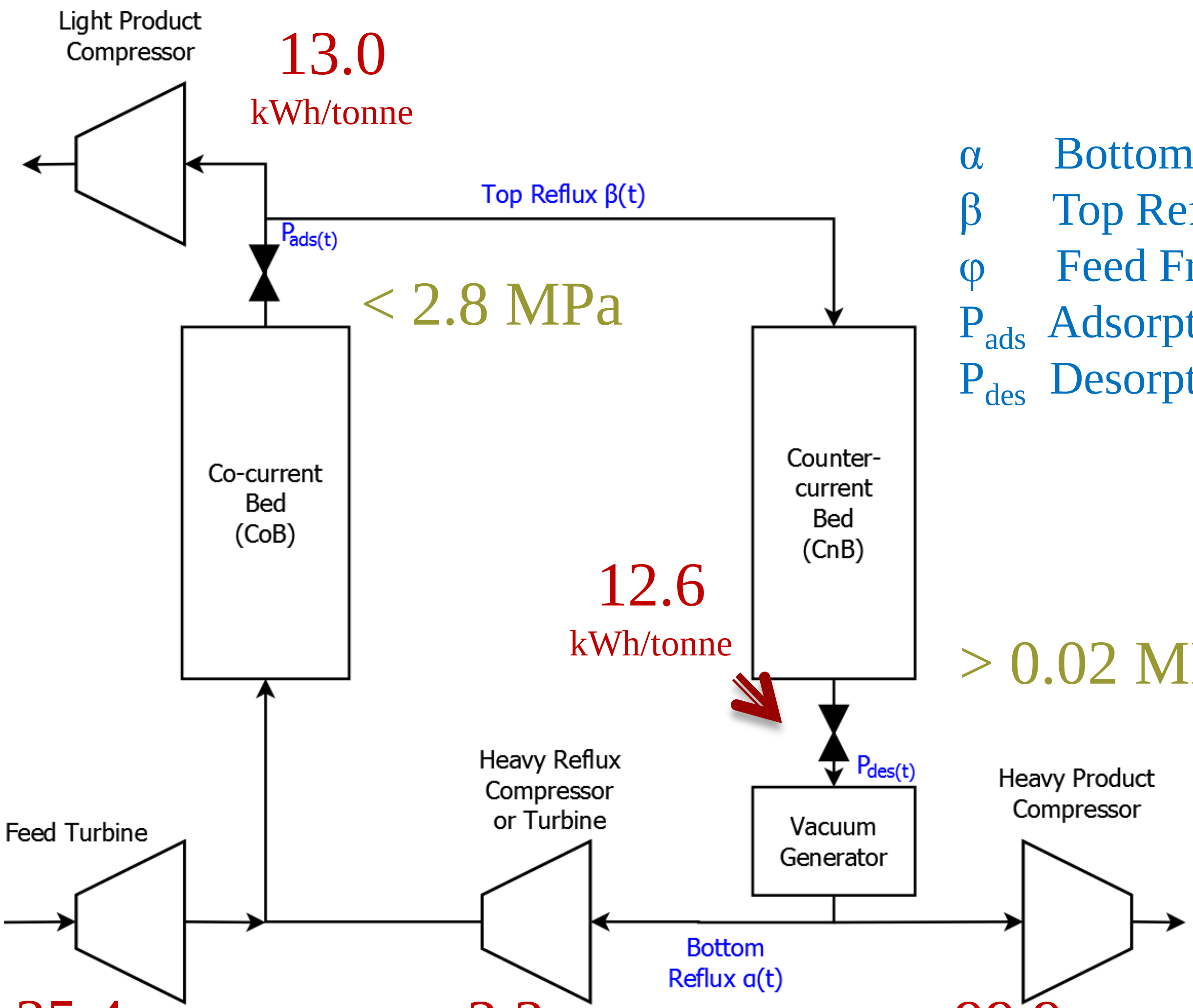
Best 5 Component
Solution

Legend: CO_2 Sorbent Loading

Low

High

86.8 kWh/tonne CO_2 captured



3.2 MPa

H_2 to Turbine

13.0 kWh/tonne

< 2.8 MPa

12.6 kWh/tonne

> 0.02 MPa

5.1 MPa

Feed from WSR, φ(t)

-35.4 kWh/tonne

-3.3 kWh/tonne

99.9 kWh/tonne

15 MPa

CO_2 to Pipeline

α	Bottom Reflux Fraction
β	Top Reflux Fraction
φ	Feed Fraction
P_{ads}	Adsorption Pressure
P_{des}	Desorption Pressure

Technology Comparison

Economic Metric: Cost of Electricity

IGCC **without** Carbon Capture*	IGCC with **Selexol** Carbon Capture*	IGCC with **PSA** Carbon Capture
$ 76 / MWh	$ 106 / MWh	**$ 103 - 109 / MWh**

Goal: $ 83 / MWh

- Results are with **activated carbon**
- Future work: consider advanced sorbents

**Cost and Performance Baseline for Fossil Energy Plants Vol 1: Bit. Coal and Nat. Gas to Elec.*, NETL (2010)

Conclusions

- Compared three PSA optimization formulation
- Developed novel application of adjoint sensitivity equations to PSA optimization
- Demonstrated potential cost competitiveness of PSA for H_2-CO_2 separation in IGCC power plant with an *activated carbon sorbent*

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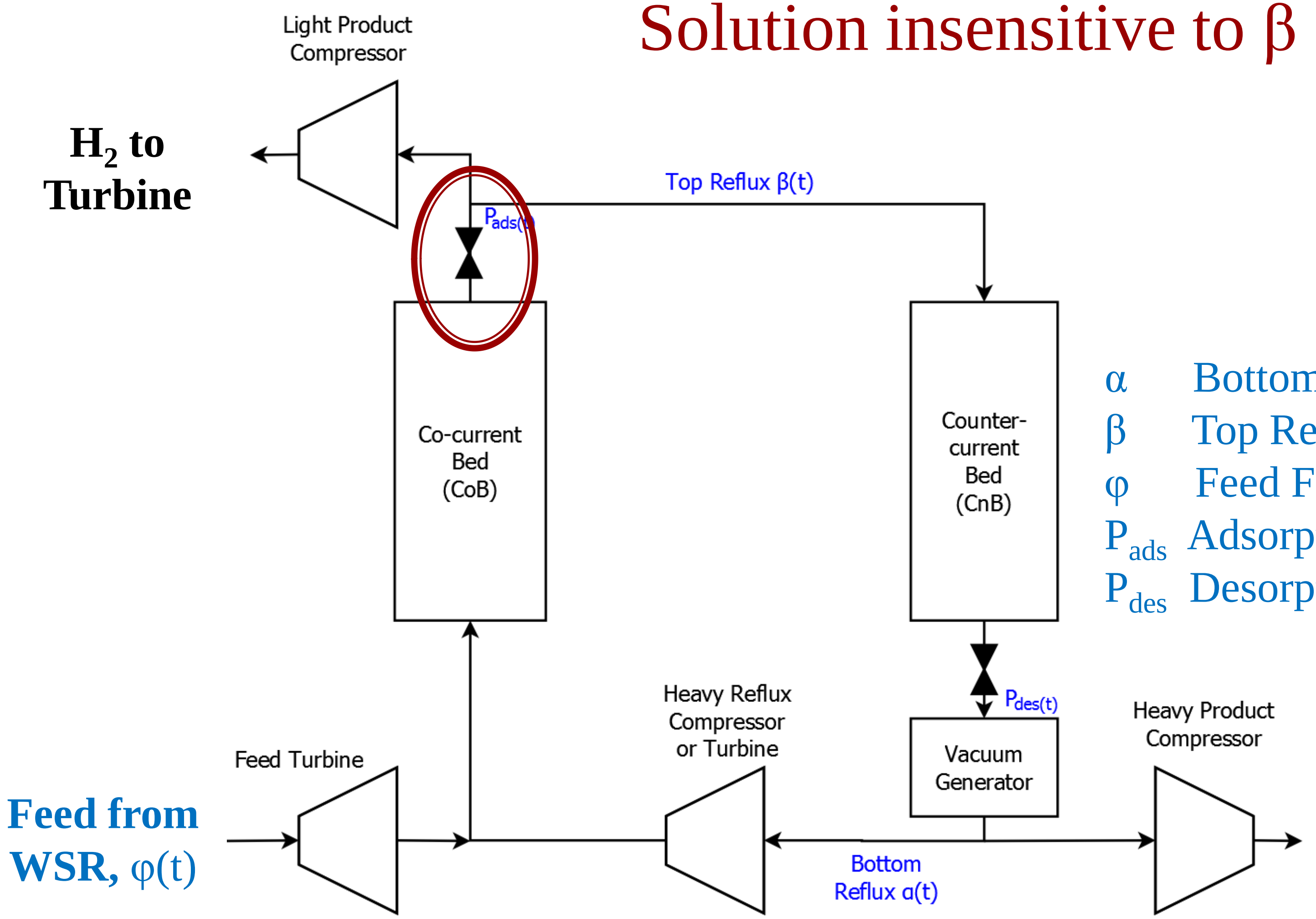
Optimization Convergence

Valve closes when $P < P_{ads}$

Solution insensitive to β and P_{ads}



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