Process Optimization with Complementarity Constraints in Chemical Engineering

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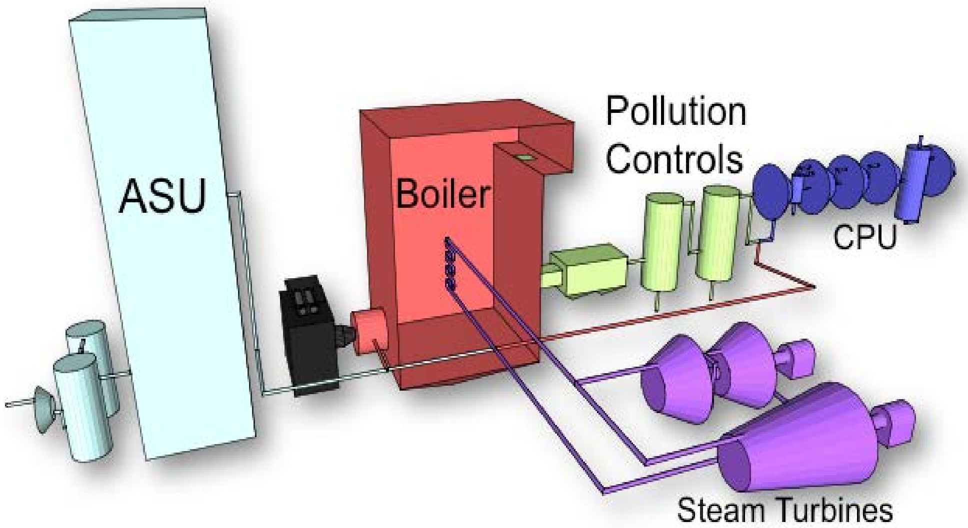
International Symposium on Mathematical Programming
July, 2015

Overview

- Introduction
  - Process optimization
  - Formulation and solution strategies
- Bilevel Optimization → MPCC
  - Phase equilibrium
  - Heat integration
- Process Optimization Case Studies
  - MHEX with phase changes (CO_2 recovery)
  - Distillation Synthesis (Air Separation)
- Conclusions

Future Generation Power Plants: Oxycombustion with CO2 Capture



Schwarze Pumpe, 30MW Pilot (2008)
Feed: Lignite; Bituminous Coal
Brandenburg, Germany

Process Optimization Models:

- **ASU – distillation (MPCC)**
- Boiler – PDAE/CFD Models
- Steam Cycle – EO models
- **CPU – MPCC models**

Bi-level Process Optimization Problems: an Alternative to (some) MINLPs

$$\underset{x,y}{Min}\ f(x, y)$$

$$\text{s.t. } g(x, y) \le 0,\ h(x,y) = 0$$

$$\underset{y}{Min}\ \bar{f}(x, y)$$

$$\text{s.t. } \bar{g}(x, y) \le 0,\ \bar{h}(x,y) = 0$$

Formulation Guidelines

- Attempt to define regular, convex inner minimization problem (optimistic bilevel problems, Dempe, 2002)
- Require connected feasible regions for inner problem variables (no exclusive ORs!)

Solving Bi-level Optimization Problems as MPCCs (Ralph, Wright, 2004)

$$\underset{x,y}{Min}\ f(x,y)$$

$$\text{s.t. } g(x,y) \le 0,\ h(x,y) = 0$$

$$\nabla_y \bar{f}(x,y) + \nabla_y \bar{g}(x,y)u + \nabla_y \bar{h}(x,y)v = 0$$

$$\bar{g}(x,y) \le 0,\ \bar{h}(x,y) = 0$$

$$0 \le u \perp \bar{g}(x,y) \le 0$$

$\underline{MINLP}$: Add binary variables

$$0 \le u_i \le M\beta^i$$

$$0 \ge \bar{g}_i(x,y) \ge M(\beta^i - 1)$$

Solving Bi-level Optimization Problems as MPCCs (Ralph, Wright, 2004)

$$\min_{x,y} \; f(x, y)$$

$$\text{s.t. } g(x, y) \le 0, \; h(x,y) = 0$$

$$\nabla_y \bar{f}(x, y) + \nabla_y \bar{g}(x, y)u + \nabla_y \bar{h}(x, y)v = 0$$

$$\bar{g}(x, y) \le 0, \; \bar{h}(x,y) = 0$$

$$0 \le u \perp \bar{g}(x, y) \le 0$$

Regularize: Relax inequalities

$$u_i \bar{g}_i(x, y) \le -\varepsilon \to 0$$

Solving Bi-level Optimization Problems as MPCCs (Ralph, Wright, 2004)

$$\underset{x,y}{Min}\ f(x,\ y)$$

$$\text{s.t. } g(x,\ y) \le 0,\ h(x,y) = 0$$

$$\nabla_y \bar{f}(x,\ y) + \nabla_y \bar{g}(x,\ y)u + \nabla_y \bar{h}(x,\ y)v = 0$$

$$\bar{g}(x,\ y) \le 0,\ \bar{h}(x,y) = 0$$

$$0 \le u \perp \bar{g}(x,\ y) \le 0$$

NCP Form : Equation with smoothed max

$$u_i - \max(0, u_i + \bar{g}_i(x,\ y)) = 0$$

$$\rightarrow u_i - \overline{\max}(u_i + \bar{g}_i(x,\ y)) = 0$$

Solving Bi-level Optimization Problems as MPCCs (Ralph, Wright, 2004)

$$\min_{x,y} \; f(x, y)$$

$$\text{s.t. } g(x, y) \le 0, \; h(x,y) = 0$$

$$\nabla_y \bar{f}(x, y) + \nabla_y \bar{g}(x, y) u + \nabla_y \bar{h}(x, y) v = 0$$

$$\bar{g}(x, y) \le 0, \; \bar{h}(x,y) = 0$$

$$0 \le u \perp \bar{g}(x, y) \le 0$$

$\underline{\ell_1 \text{ penalty}}$: Subtract $u^T \bar{g}\,(x, y)$

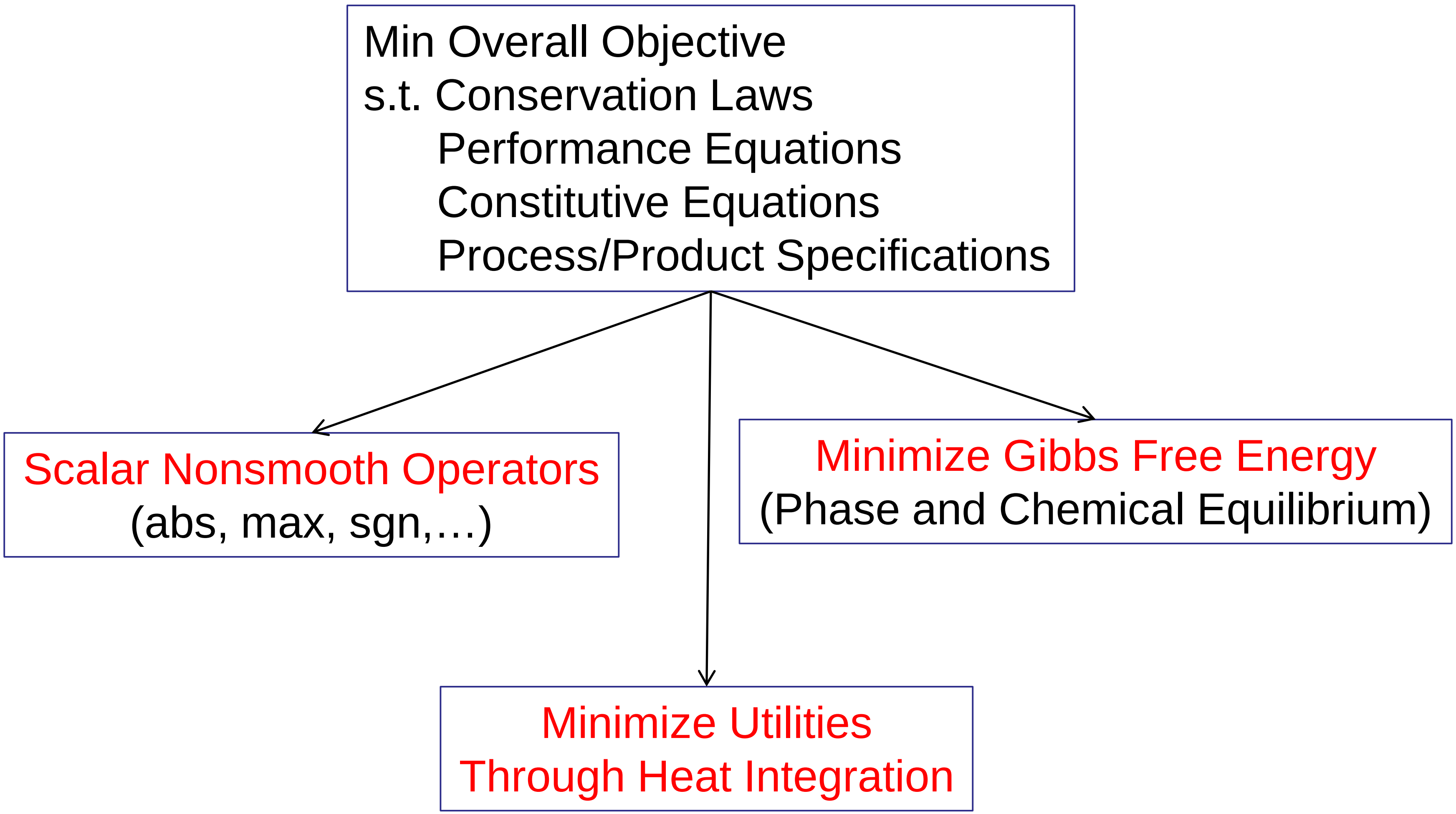
$$Min\, f(x,y) \rightarrow Min\, f(x,y) - \rho \sum_i u_i\, \bar{g}_i(x, y)$$

$$\text{s.t. } u_i \ge 0, \; \bar{g}_i(x, y) \le 0$$

Bi-level Process Optimization Models

Min Overall Objective
s.t. Conservation Laws
Performance Equations
Constitutive Equations
Scalar Nonsmooth Operators
Phase & Chem. Equilibrium
Heat Integration
Process/Product Specifications

Bi-level Process Optimization Models



Complementarity Formulations: Common Nonsmooth Functions in Process Models

- Abs(*)

$$\begin{array}{l} f(x) = s^+ - s^- \\ 0 \le s^+ \perp s^- \ge 0 \end{array} \Rightarrow |f(x)| = s^+ + s^-$$

- Min(*,*) & Max(*,*) (includes Pos(*), Neg(*))

$$\begin{array}{l} y = \min(f(x), y^{UB}) \\ f(x) - y = s \\ y \le y^{UB} \perp s \ge 0 \end{array} \qquad \begin{array}{l} y = \max(f(x), y^{LB}) \\ f(x) - y = s \\ y \ge y^{LB} \perp s \ge 0 \end{array} \qquad \begin{array}{l} Pos(f(x)) = \max(f(x), 0) \\ Neg(f(x)) = \min(f(x), 0) \end{array}$$

- Signum(*)

$$signum(x) = \begin{cases} -1 & x < 0 \\ 1 & x > 0 \end{cases} \Rightarrow \begin{array}{ll} \min & -u * x \\ s.t. & -1 \le u \le 1 \end{array} \Rightarrow signum(x) = u$$

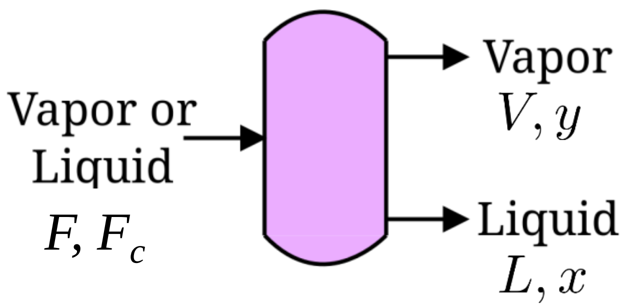
- If * Then * Else * (includes Piecewise Functions)

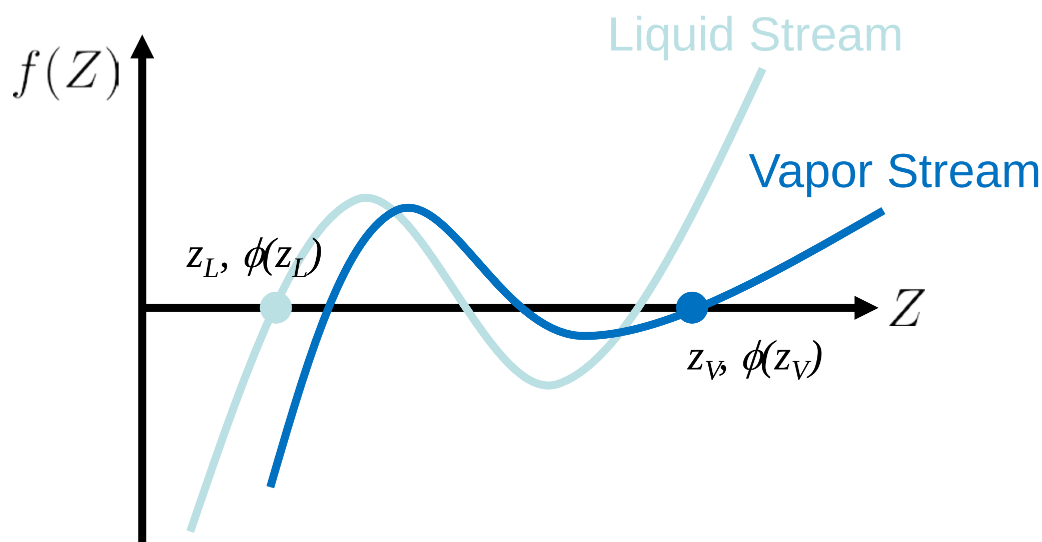
$$\begin{array}{ll} \min & u(x - x_{switch}) \\ s.t. & 0 \le u \le 1 \end{array}$$
$$y = (u) f_1(x) + (1-u) f_2(x)$$

$$\begin{array}{c} (x - x_{switch}) - \lambda_0 + \lambda_1 = 0 \\ 0 \le \lambda_0 \perp u \ge 0 \\ 0 \le \lambda_1 \perp (1-u) \ge 0 \end{array}$$

CEOS Phase Equilibrium through Complementarity (Kamath, Grossmann, B., 2011)

$$Z^3 - (1 + B - uB)Z^2 + (A + wB^2 - uB - uB^2)Z - AB - wB^2 - wB^3 = 0$$





$$F = L + V$$

$$F_c = Lx_c + Vy_c, \quad \forall c \in \{Comps\}$$

$$FH^F + Q = LH^L + VH^V$$

$$y_c = K_c(T, P, x, y)x_c$$

$$0 \leq x_c, y_c \leq 1$$

$$0 \leq L, V \leq F$$

$$K_c = \phi_c^L / \phi_c^V, \quad \forall c \in \{Comps\}$$

$$f(z_L) = 0 \qquad f(z_V) = 0$$

$$f'(z_L) \geq 0 \qquad f'(z_V) \geq 0$$

$$f''(z_L) \leq 0 \qquad f''(z_V) \geq 0$$

Mass Balance + Necessary KKT conditions

CEOS Phase Equilibrium through Complementarity (Kamath, Grossmann, B., 2011)

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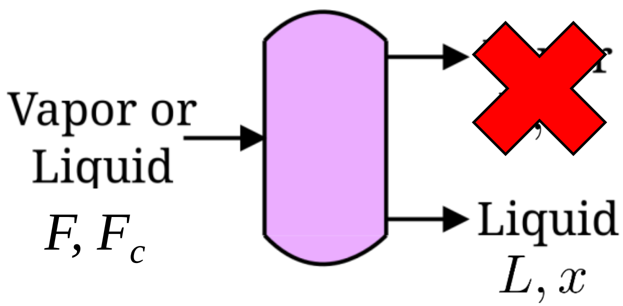
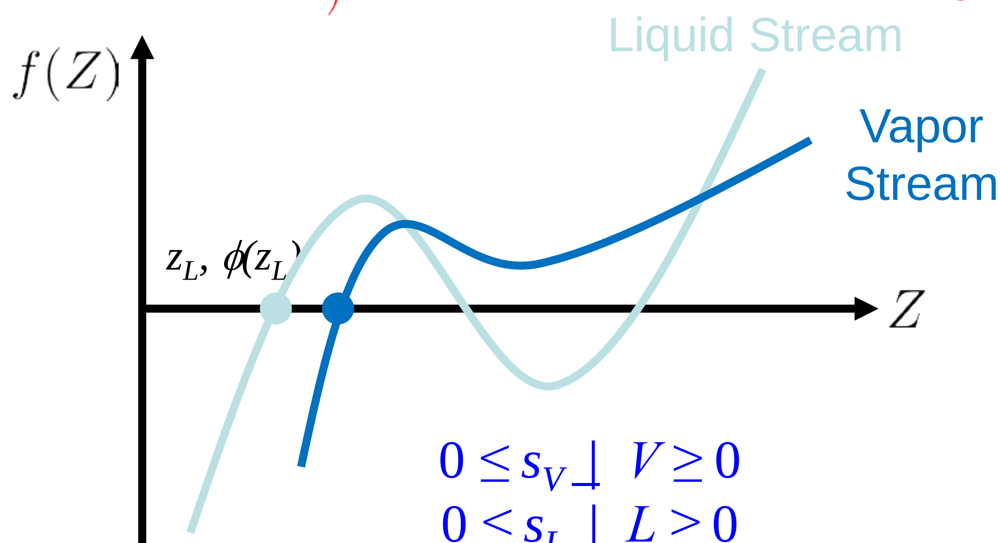




$$0 \le s_V \perp V \ge 0$$
$$0 \le s_L \perp L \ge 0$$
$$-s_L \le \beta - 1 \ge s_V$$

$$F = L + V$$

$$F_c = Lx_c + Vy_c, \quad \forall c \in \{Comps\}$$

$$FH^F + Q = LH^L + VH^V$$

$$y_c = \beta \; K_c(T, P, x, y)x_c$$

$$0 \le x_c, y_c \le 1$$

$$0 \le L, V \le F$$

$$K_c = \phi_c^L / \phi_c^V, \quad \forall c \in \{Comps\}$$

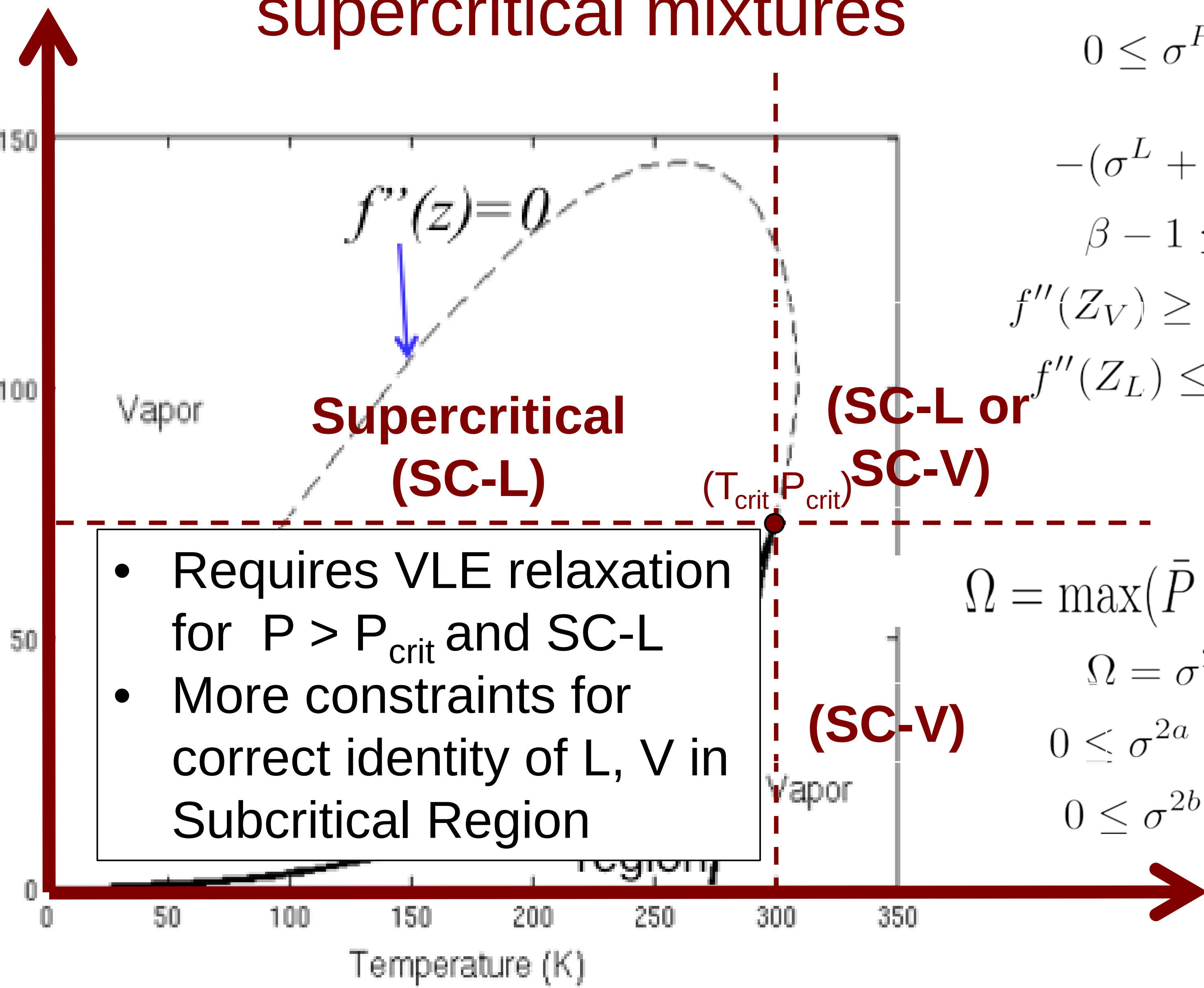
$$f(z_L) = 0 \qquad f(z_V) = 0$$

$$f'(z_L) \ge 0 \qquad f'(z_V) \ge 0$$

$$f''(z_L) \le 0 \qquad f''(z_V) \ge 0$$

$$f''(z_L) \le Ms_L \qquad f''(z_V) \ge -Ms_V$$

f''(z) constraint misclassifies supercritical mixtures



- Requires VLE relaxation for $P > P_{crit}$ and SC-L
- More constraints for correct identity of L, V in Subcritical Region

$$P + \sigma^P \geq \bar{P}$$

$$0 \leq \sigma^P \perp \xi \geq 0$$

$$-(\sigma^L + \xi) \leq \beta - 1$$

$$\beta - 1 \leq \sigma^V + \xi$$

$$f''(Z_V) \geq -M(\sigma^V + \xi)$$

$$f''(Z_L) \leq M(\sigma^L + \xi)$$

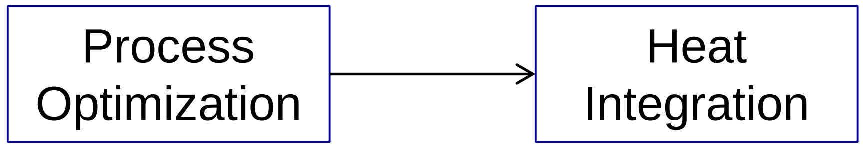
$$\Omega = \max(\bar{P} - P, T - \bar{T})$$

$$\Omega = \sigma^{2a} - \sigma^{2b}$$

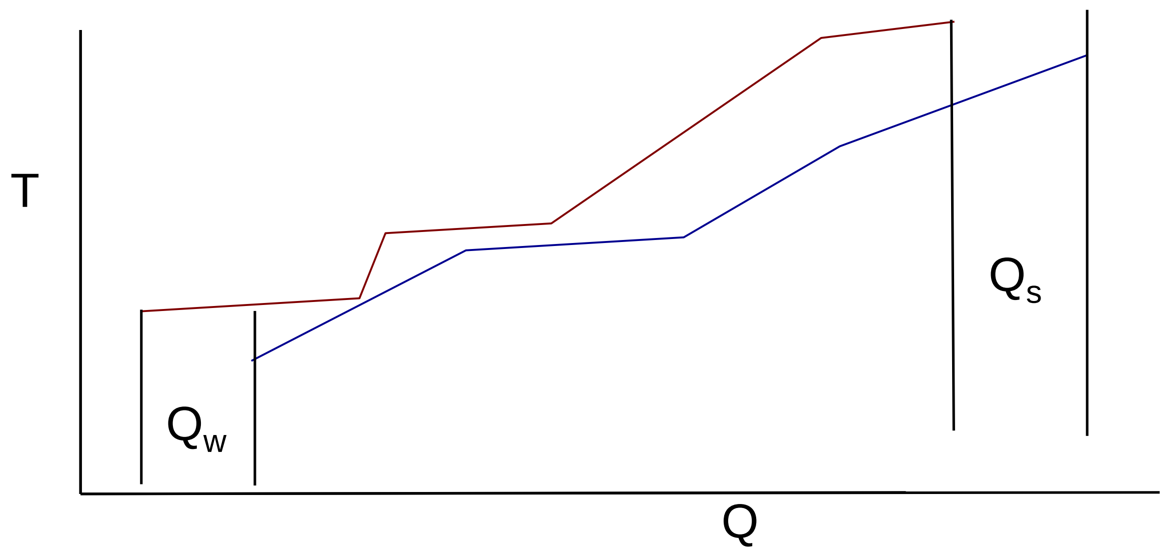
$$0 \leq \sigma^{2a} \perp \sigma^{2b} \geq 0$$

$$0 \leq \sigma^{2b} \perp V \geq 0$$

Bilevel Optimization: Simultaneous Process Optimization & Heat Integration
(Duran, Grossmann, 1986)



- Process optimization and heat integration tightly coupled
- Allows production, power, capital to be properly considered
- Data for pinch curves adapted by optimization



Bilevel Optimization: Simultaneous Process Optimization & Heat Integration
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Process Optimization ⟷ Heat Integration

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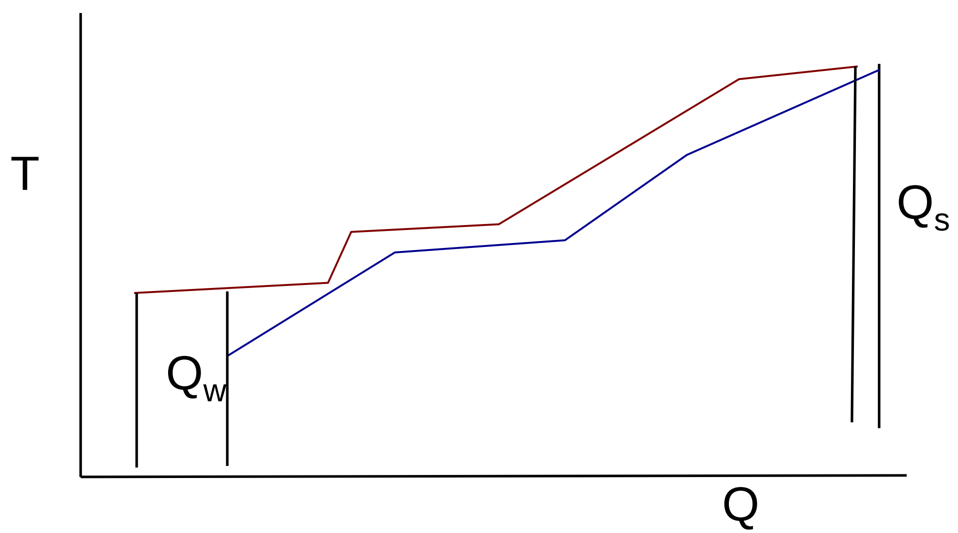


Simultaneous Process Optimization & Heat Integration

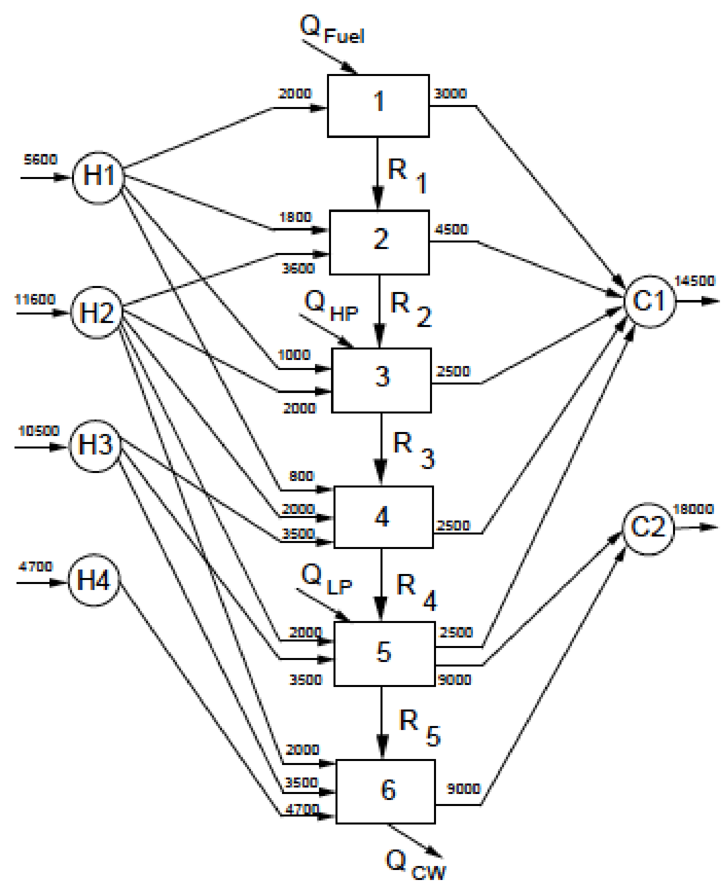
$$\min \; f(x) = \Phi(x) + c_s Q_s + c_w Q_w$$
$$s.t. \quad h(x) = 0$$
$$g(x) \le 0$$

Flowsheet objective, process model and constraints

LP Transshipment Model

- Stream temperatures as pinch candidates
- Energy balance over each temperature interval
- Form energy cascade with nonnegative heat flows

→ Models pinch curves



Bilevel Reformulation: Simultaneous Process Optimization & Heat Integration

$$\begin{aligned} \min \; & f(x) = \Phi(x) + c_s Q_s + c_w Q_w \\ s.t. \; & h(x) = 0 \\ & g(x) \le 0 \end{aligned}$$

Flowsheet objective, process model and constraints

$$Q_s \ge \sum_{j=1}^{n_c} f_j c_{p,j} \quad [\max\{0, t_j^{out} - (T^p - \Delta T_{\min})\} - \max\{0, t_j^{in} - (T^p - \Delta T_{\min})\}]$$
$$- \sum_{i=1}^{n_H} F_i C_{p.i} \; [\max\{0, T_i^{in} - T^p\} - \max\{0, T_i^{out} - T^p\}], \; p \in P$$
$$Q_w = Q_s + \sum_{i=1}^{n_H} F_i C_{p,i} (T_i^{in} - T_i^{out}) - \sum_{j=1}^{n_C} f_j c_{p,j} \quad (t_j^{out} - t_j^{in})$$

Replace with smoothed max(ξ, 0) functions
Further improved at points where $\xi = 0$.
(Unroll summations)

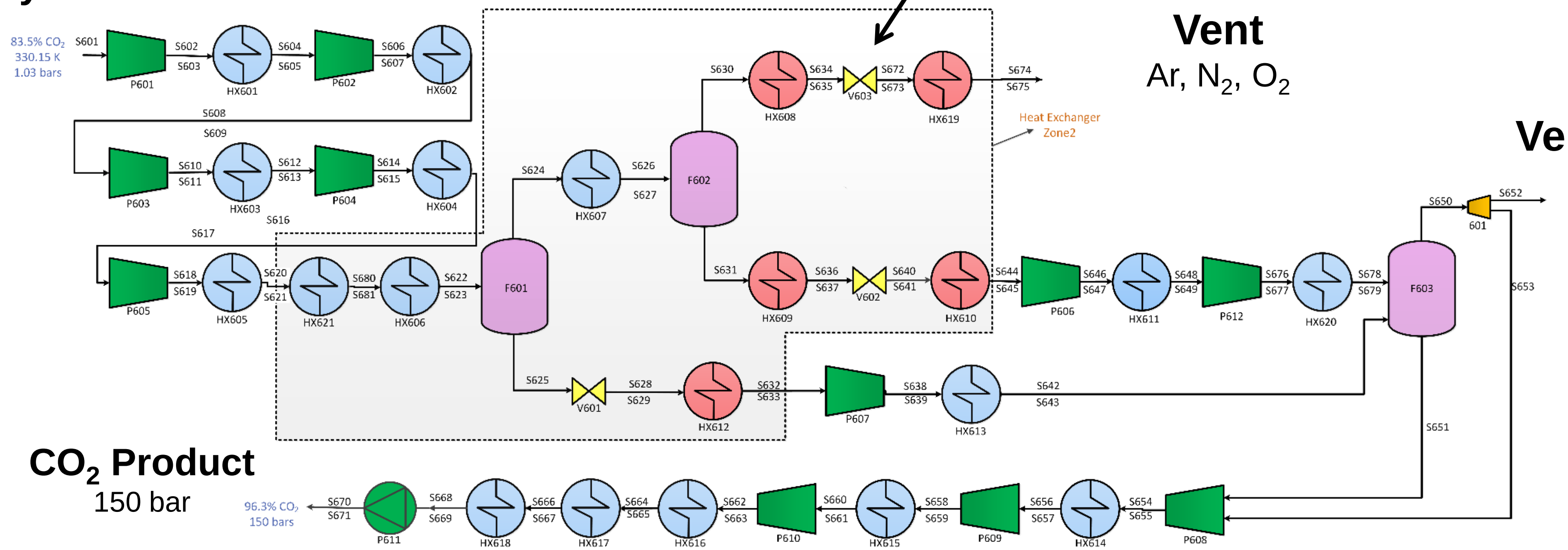
CO_2 Processing Unit (CPU)

(adapted from Fu and Gundersen, 2012)

Zone 2
Multistream Heat Exchanger(s)

Zone 1
Chilled Sea Water

Dry Flue Gas



Zone 1
Chilled Sea Water

Compression/Expansion Work
Cooling/Heating for Heat Exchange
Phase Separation Units

CPU Optimization Problem

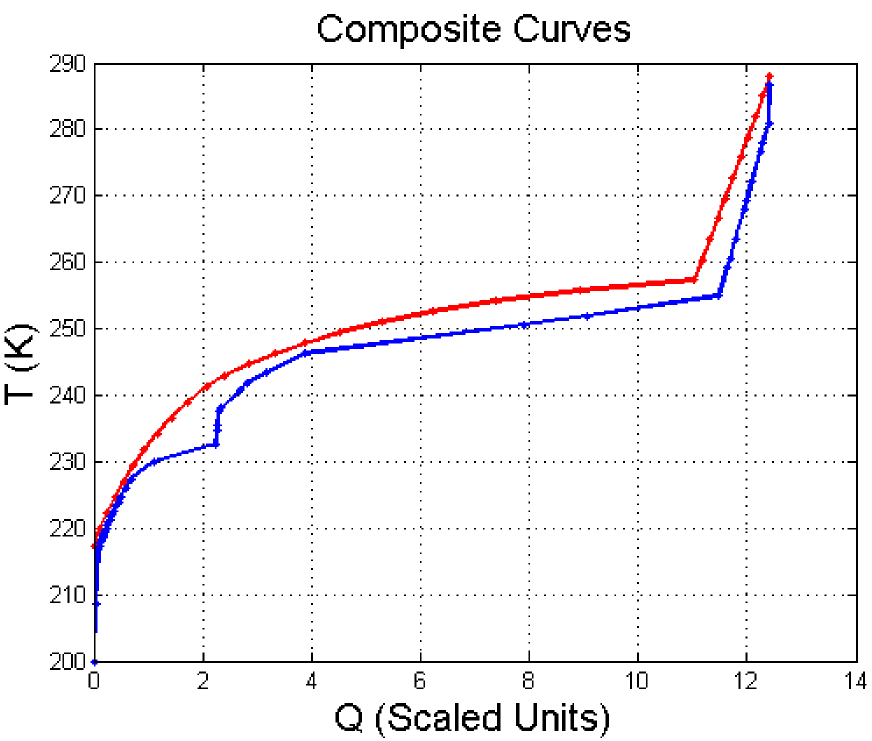
Minimize Shaft Work + 0.01 (Q^s_1 + Q^w_1) + 5 (Q^s_2 + Q^w_2)
+ Complementarity Penalties

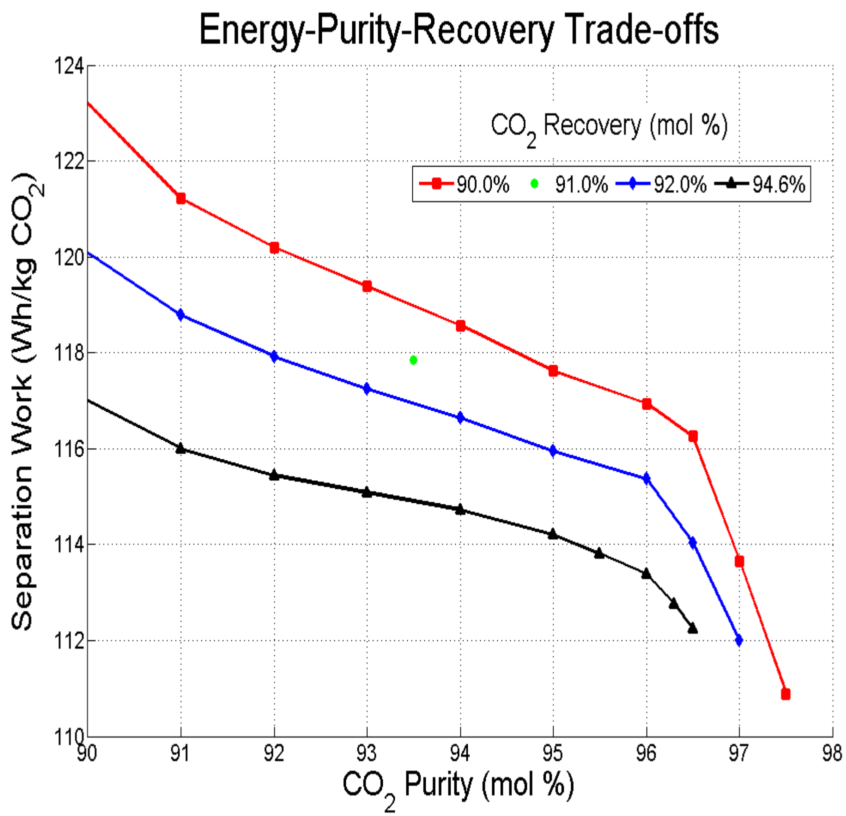
s.t. Flowsheet Connectivity
CEOS (Peng-Robinson) Thermodynamics
Heat Integration Model with Multiple Zones
Avoid Dry Ice Constraints
Zone 1 Utility Min. Temperature
Pump and Compressor Model
Other Unit Operation Models
CO_2 Recovery ≥ 96.3 mole %
CO_2 Purity ≥ 94.6 mole %

Final NLP size: 11,285 constraints, 11,808 variables
Entire 6 step NLP-based sequence: 308 CPUs

CPU Optimization Results

(Dowling et al., 2015)

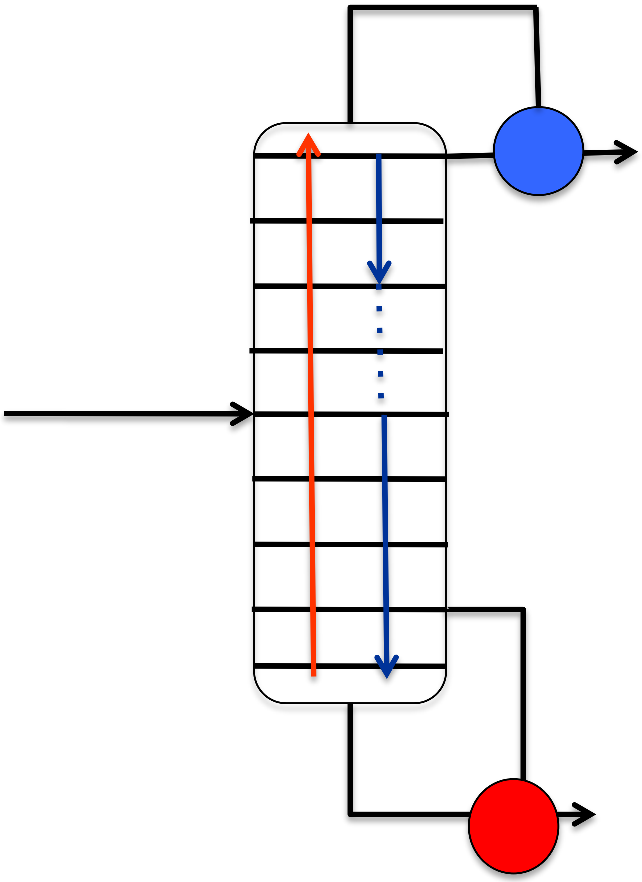




- Optimal heat integration through D-G Formulation
- Superior Heat/Power integration compared to literature
- Global solutions promoted through Multi-start NLP

Distillation: Complementarity Formulation

(Raghunathan, B, 2002)



- Consists of Mass, Equilibrium, Summation and Heat (MESH) equations
- Continuous Variable Optimization
  - number of stages
  - feed location
  - reflux ratio

- When phases disappear, MESH fails.
- Reformulate phase minimization,
  - embed complementarity
  - Model dry stages, Vaporless stages

- Initialization with Shortcut models based on Kremser Equations (Kamath, Grossmann, B., 2010)

Distillation Optimization (MESH Model)

Minimize Reboiler Duty

s.t. Top/ Bottom Product Specifications

$$(L_i + DL + rd)x_{ij} + DVy_{ij} = V_{i-1}y_{i-1,j} \quad i \in CON$$

$$L_i x_{ij} + V_i y_{ij} = L_{i+1}x_{i+1,j} + V_{i-1}y_{i-1,j} + \sum_k f_{ik} Fd_k xf_{dj} + g_i \cdot rd \cdot x_{ij} \quad i \in COL$$

$$Bx_{ij} + V_i y_{ij} = L_{i+1}x_j + \sum_k f_{ik} Fd_k xf_{dj} \quad i \in REB$$

$$y_{ij} = \beta_i K_{ij} x_{ij}$$

$$-s_i^V \le \beta_i - 1 \le s_i^L$$

$$0 \le L_i \perp s_i^L \ge 0$$

$$0 \le V_i \perp s_i^V \ge 0$$

$$(L_i + DL + rd)hl_i + DV \cdot hv_i = V_{i-1}hv_{i-1} + Q_c \quad i \in CON$$

$$L_i hl_i + V_i hv_i = L_{i+1}hl_{i+1} + V_{i-1}hv_{i-1} + \sum_k f_{ik} Fd_k shf_{dj} + g_i \cdot rd \cdot hl_i \quad i \in COL$$

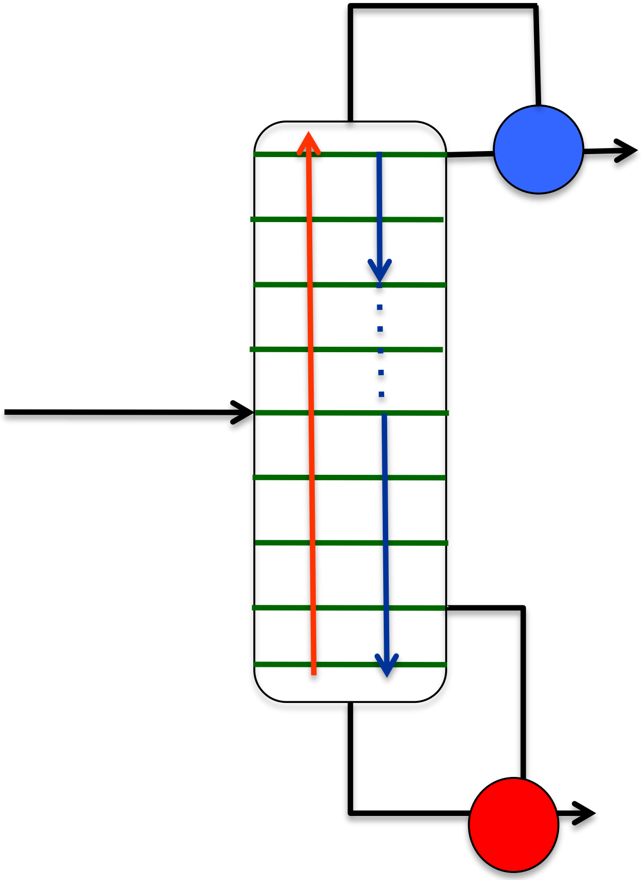
$$B \cdot hl_i + V_i hv_i = L_{i+1}hl_{i+1} + \sum_k f_{ik} Fd_k shf_{dj} + Q_H \quad i \in REB$$

$$\sum_J x_{ij} - \sum_J y_{ij} = 1$$

$$R_{total} = R \cdot D$$

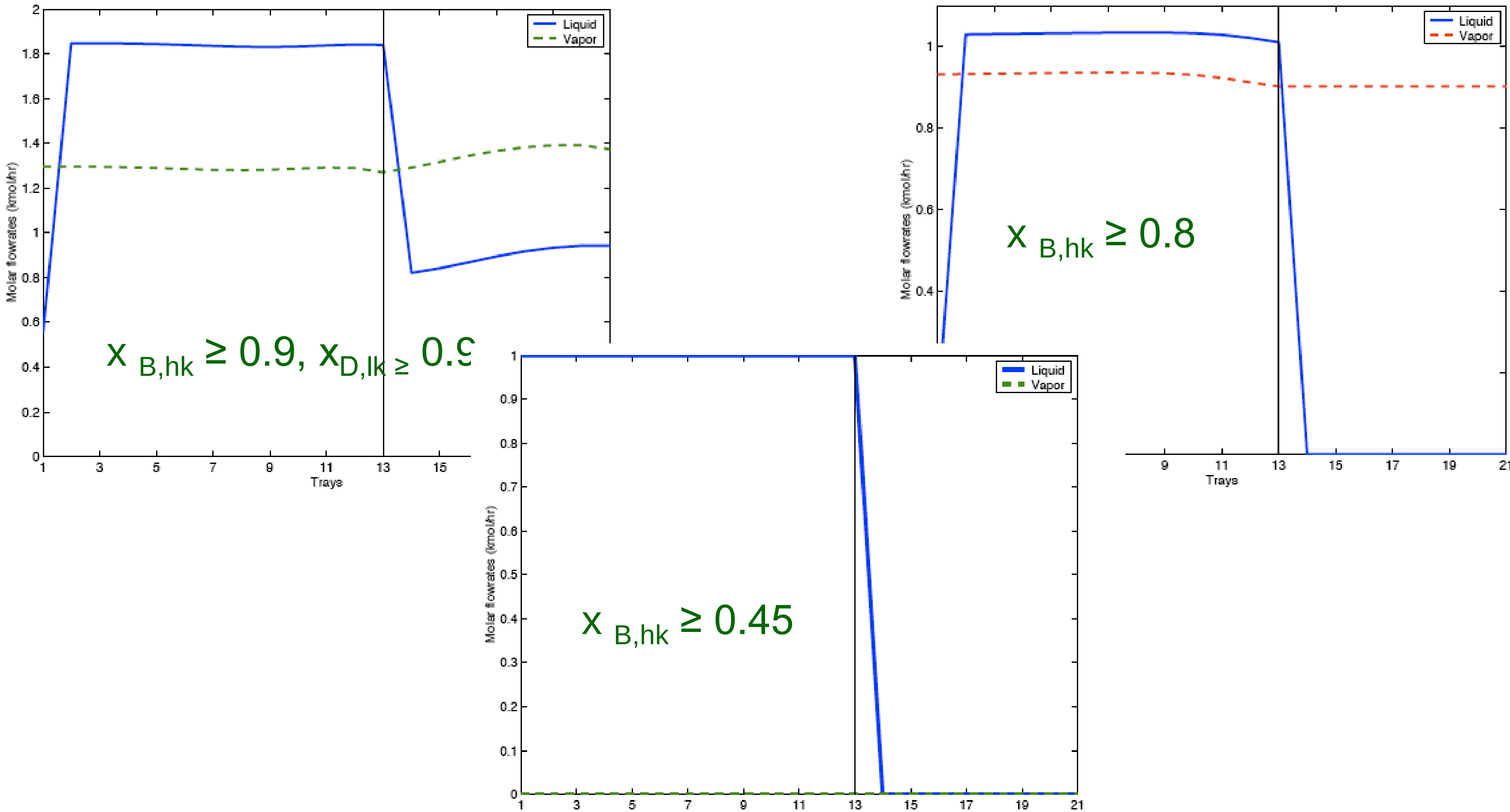
$$L_{N_{\max}} = (1 - rdf)R_{total}$$

$$rd = rdf \cdot R_{total}$$



Five component Separation
Fixed: 20 Stages, Feed = 12
Reboil and Reflux as decisions

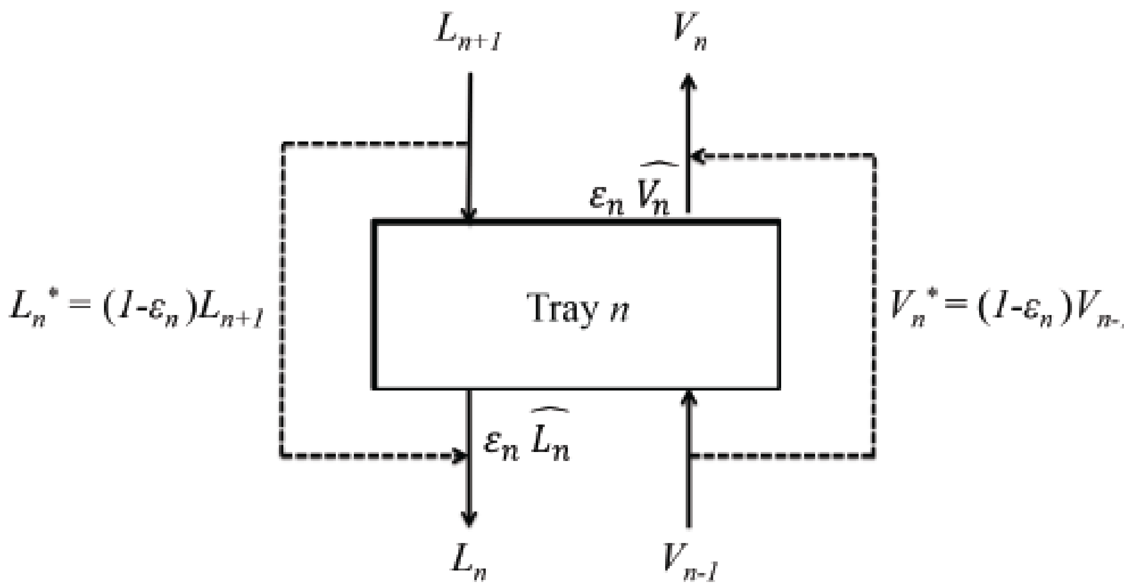
Distillation Results – Min Heat Duty



Case	Product purity constraints	Optimal reboiler duty (MW)
1	$x_{\text{top},n\text{-hexane}} \geq 0.9$, $x_{\text{bottom},n\text{-nonane}} \geq 0.9$	28.14
2	$x_{\text{bottom},n\text{-nonane}} \geq 0.8$	19.337
3	$x_{\text{bottom},n\text{-nonane}} \geq 0.45$	0.0

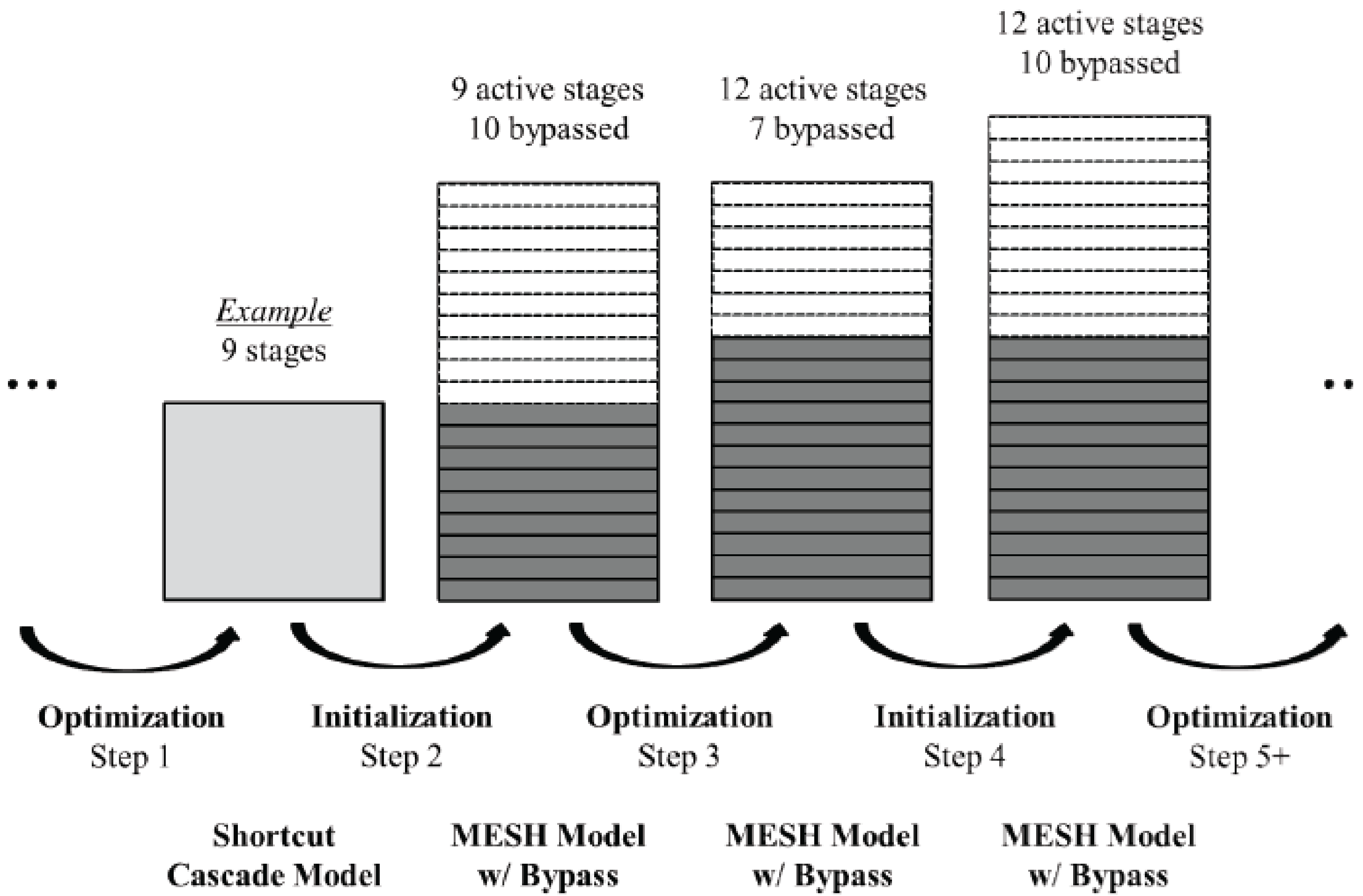
Disappearing Phases Allow Bypass Stages: MPCC Optimization Formulation

(Dowling, B., 2014)



- Dummy streams equilibrium streams based on MPCC for phase equilibrium → $\hat{L}, \hat{V}$
- Bypass ***usually*** leads to binary solution for ε.
- Mixing discouraged in optimization (energy inefficient)
- Fractional ε is physically realizable.
- #Stages = $\sum_n \varepsilon$

MPCC sequence with Distillation Models



Heat Integration and Distillation Optimization
Air Separation Units

Boiling pts (1 atm.)

- Oxygen: 90 K
- Argon: 87.5 K
- Nitrogen: 77.4 K

- Feedstock (air) is free: dominant cost is power for compression
- Multicomponent distillation with tight heat integration
- Nonideal Phase Equilibrium: Cubic Equations of State
- Phase conditions not known *a priori*

ASU Synthesis Optimization Problem

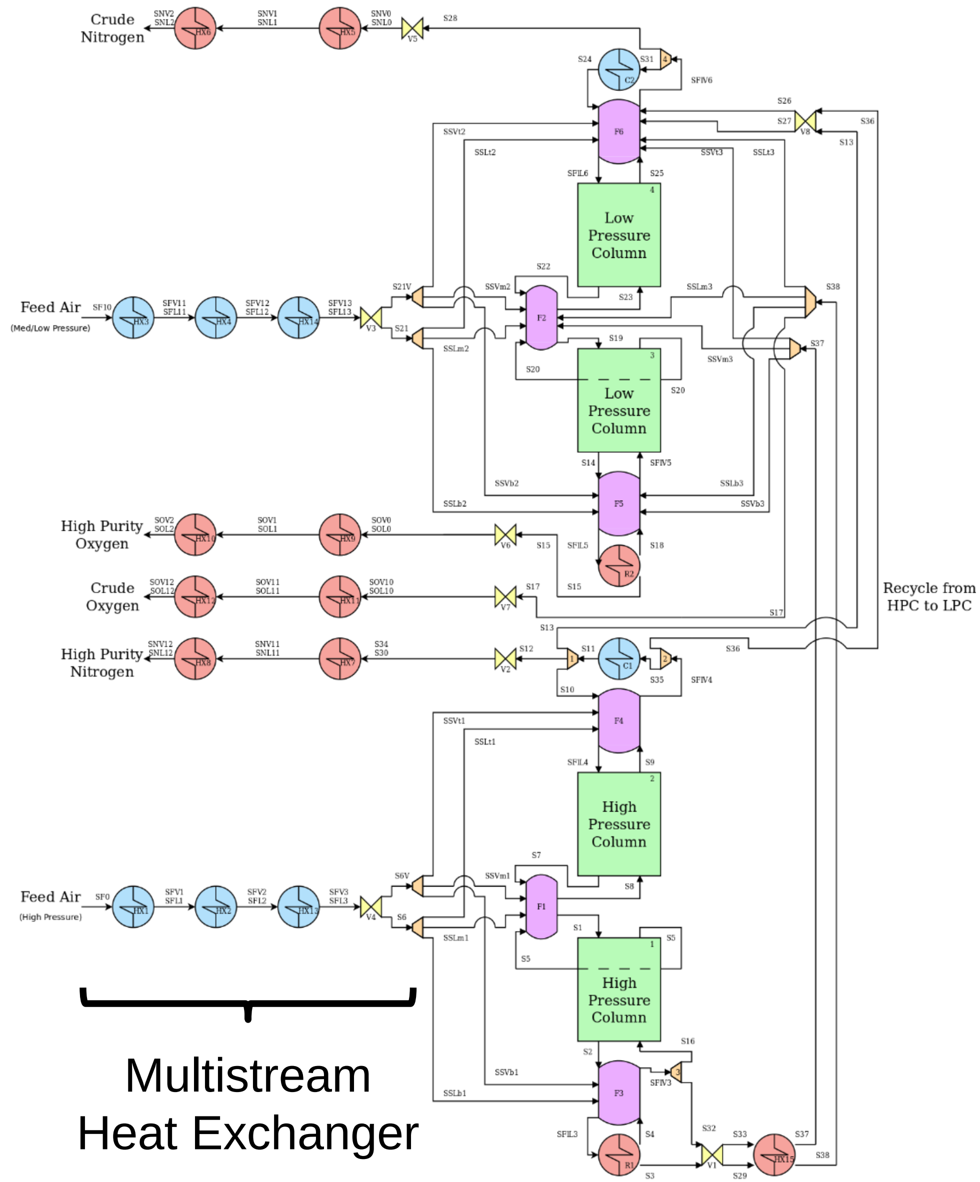
Minimize Compression Energy + Q^s + Q^w
+ Complementarity Penalties

s.t. Flowsheet Connectivity
CEOS (Peng-Robinson) Thermodynamics
Heat Integration Model
Distillation Cascade Model
Unit Operation Models
O_2 Purity ≥ 95 mole %

- Find optimal T, P, flows in superstructure
- MPCC with ~16,000 variables and constraints
- Automated 6-step NLP-based initialization, simple → complex models
- Multi-start procedure to promote global solutions
- ~16 CPU min for 6-step sequence using CONOPT3

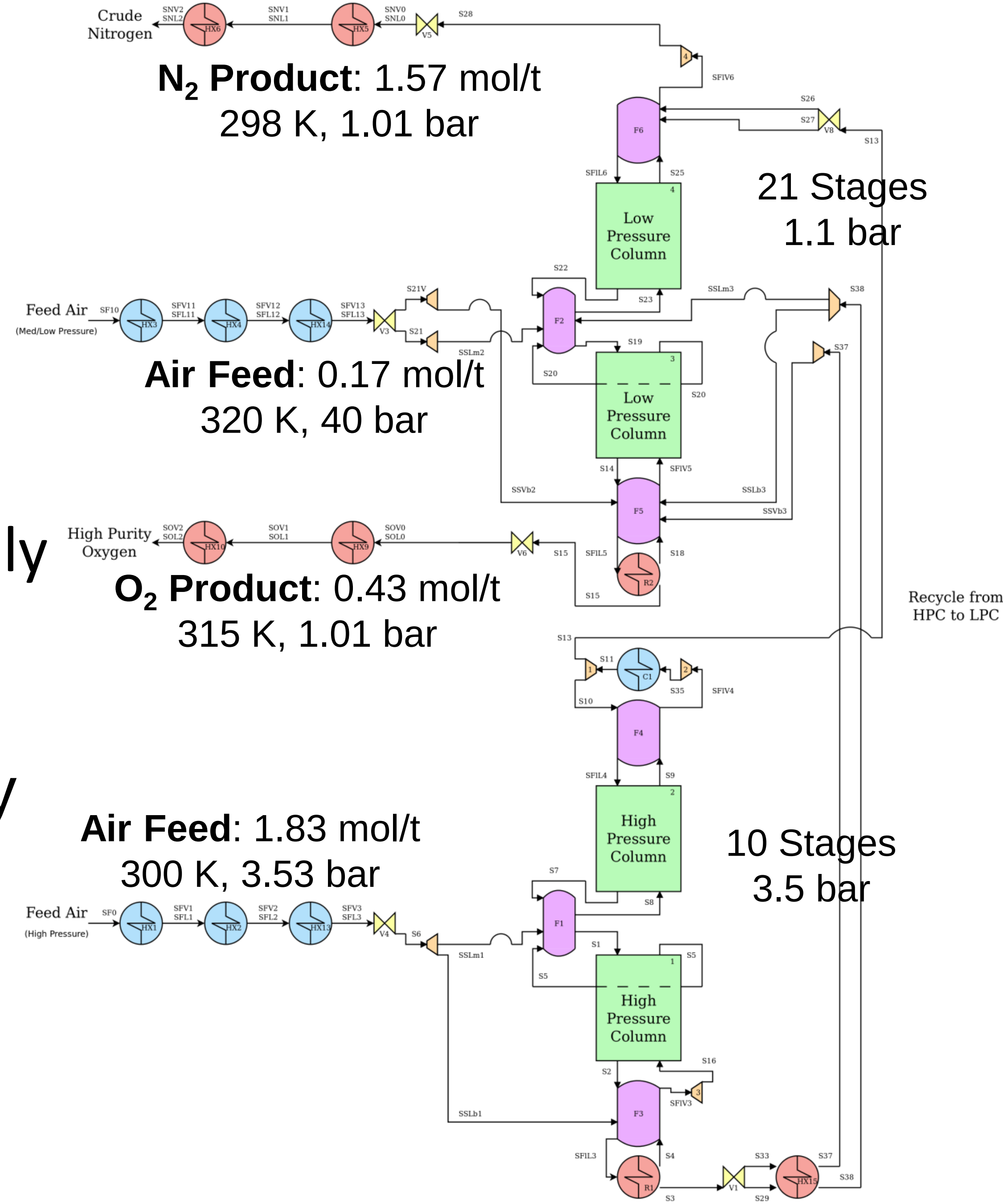
ASU Superstructure

- Flash vessels represent feed stages
- Cascade sections contain a variable number of stages
- NLP selects P, T, flowrates and best recycle configuration

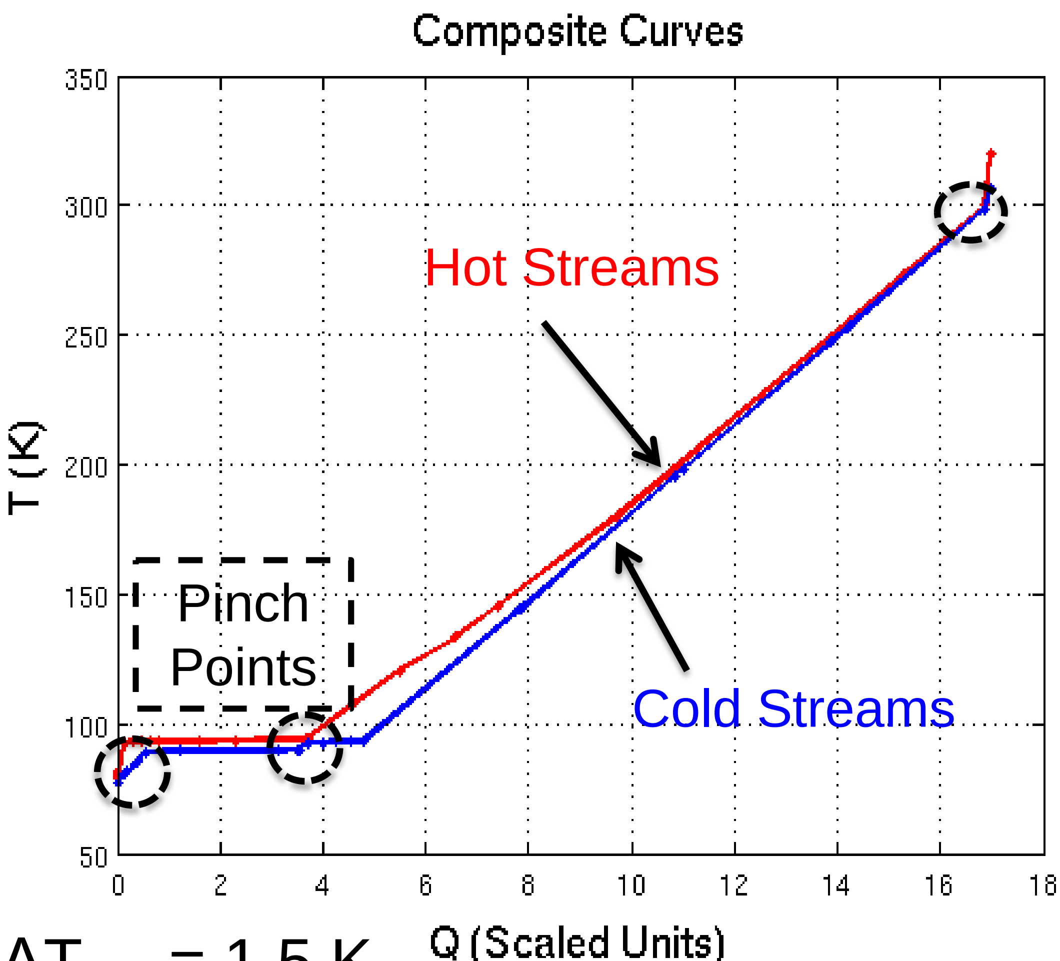


Optimized ASU Process

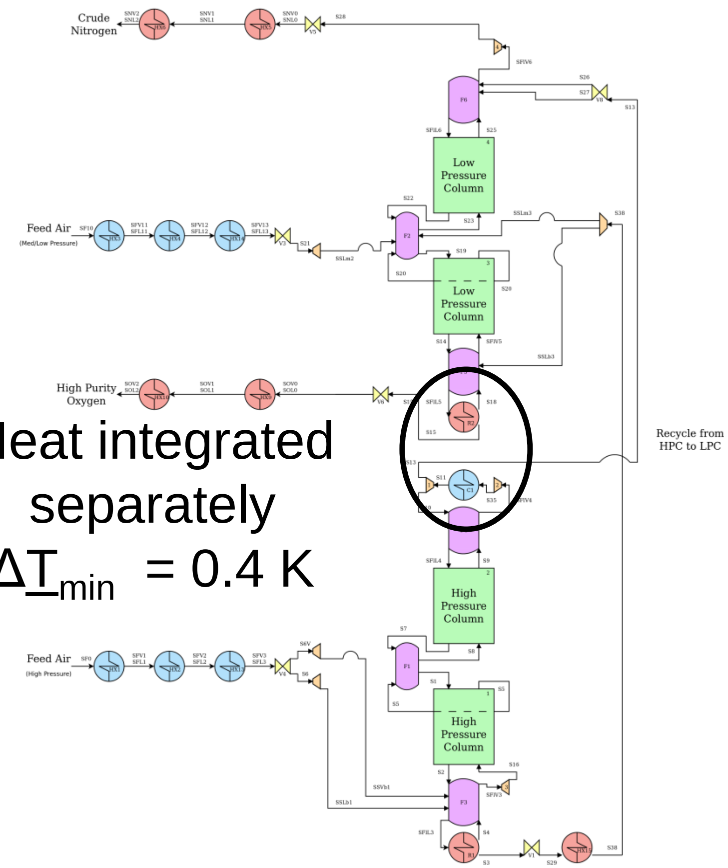
- Balanced Reboiler/Condenser
- No external utilities, only compression
- ΔT_{min} = 1.5 K: 86% compressor efficiency
- **Optimal Power: 196 kWh/tonne O_2**



Heat Integration Results



ΔT_{min} = 1.5 K

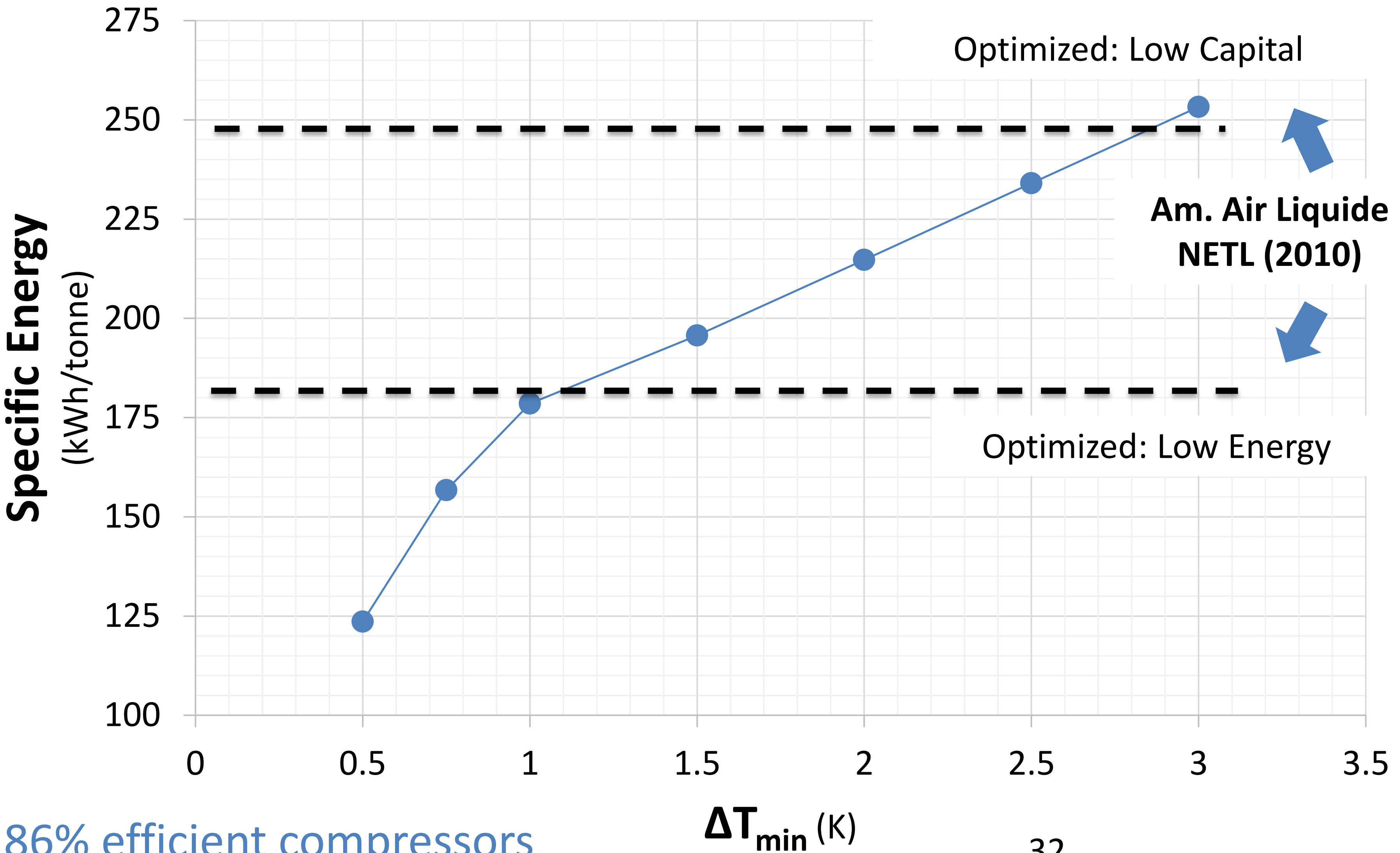


Heat integrated separately

ΔT_{min} = 0.4 K

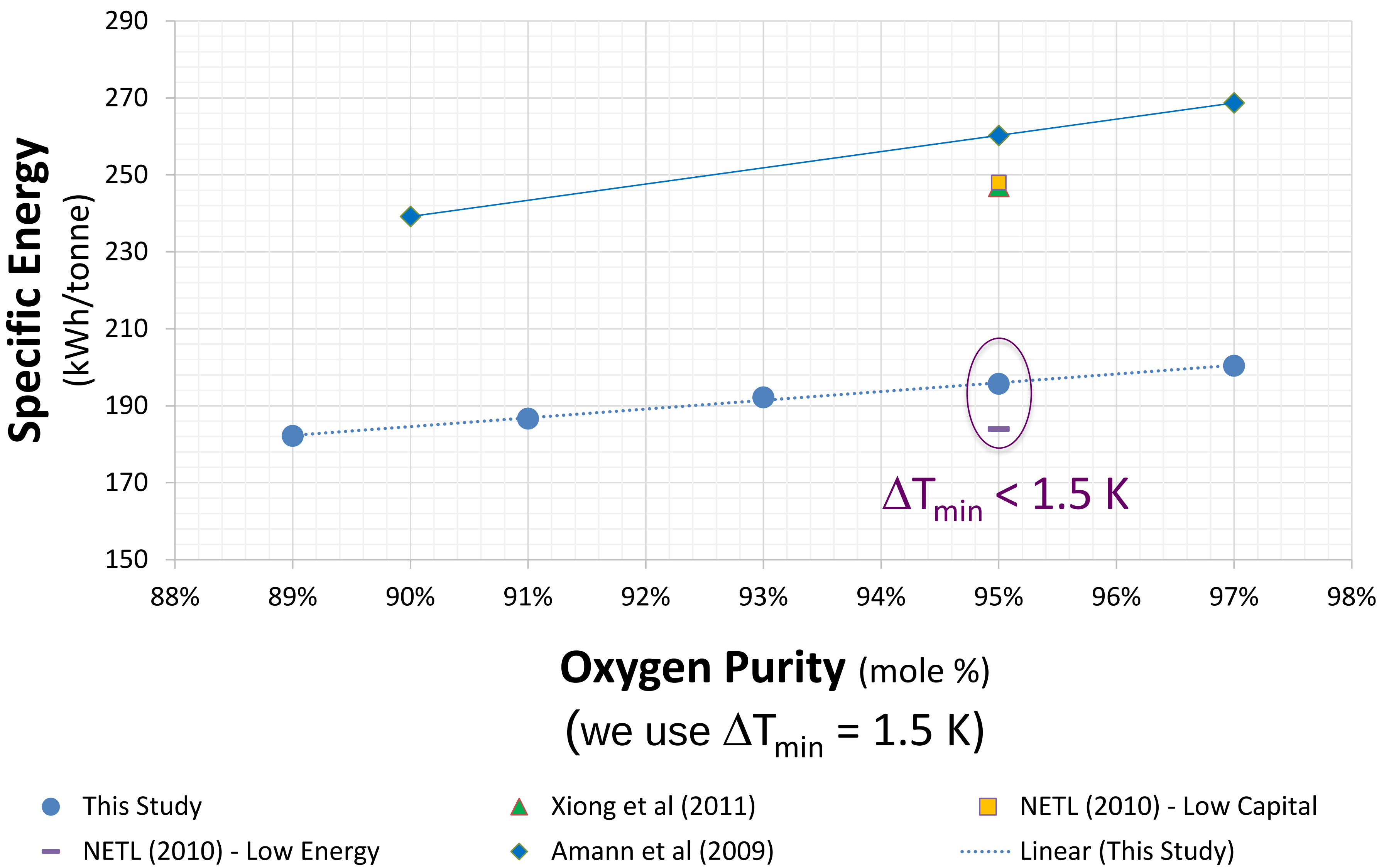
Tight heat integration with multiple pinch points

ASU Parametric Optimization wrt ΔT_{min}



86% efficient compressors

Process Optimization Based on O_2 Purity



Conclusions

- Equation Oriented Process Optimization
  - Fast Newton-based NLP solvers
  - Requires robust formulations and initializations
- Exploit bilevel problems as MPCCs
  - Simultaneous heat integration and optimization
  - Phase (and chemical) equilibrium
  - Optimal synthesis of distillation sequences
- Process optimization applications
  - CO_2 Compression Processes (HEX, compressors, phase changes)
  - Heat integrated separation (ASUs)
  - ***Integrated flowsheet optimization***

Acknowledgements

Funding:

CCSI Carbon Capture Simulation Initiative

NETL National Energy Technology Laboratory

U.S. Department of Energy

Qianwen Gao, CMU	CPU optimization
David Miller, DOE/NETL	CCSI Technical Team Lead
Jinliang Ma, NETL/URS	CCSI, 1D/3D hybrid boiler model
John Eason, CMU	ROM trust region opt. framework
Cheshta Balwani, CMU	CPU optimization, CEOS refinement
Michael Matuszewski, NETL/Pitt	ASU Data and Comparisons

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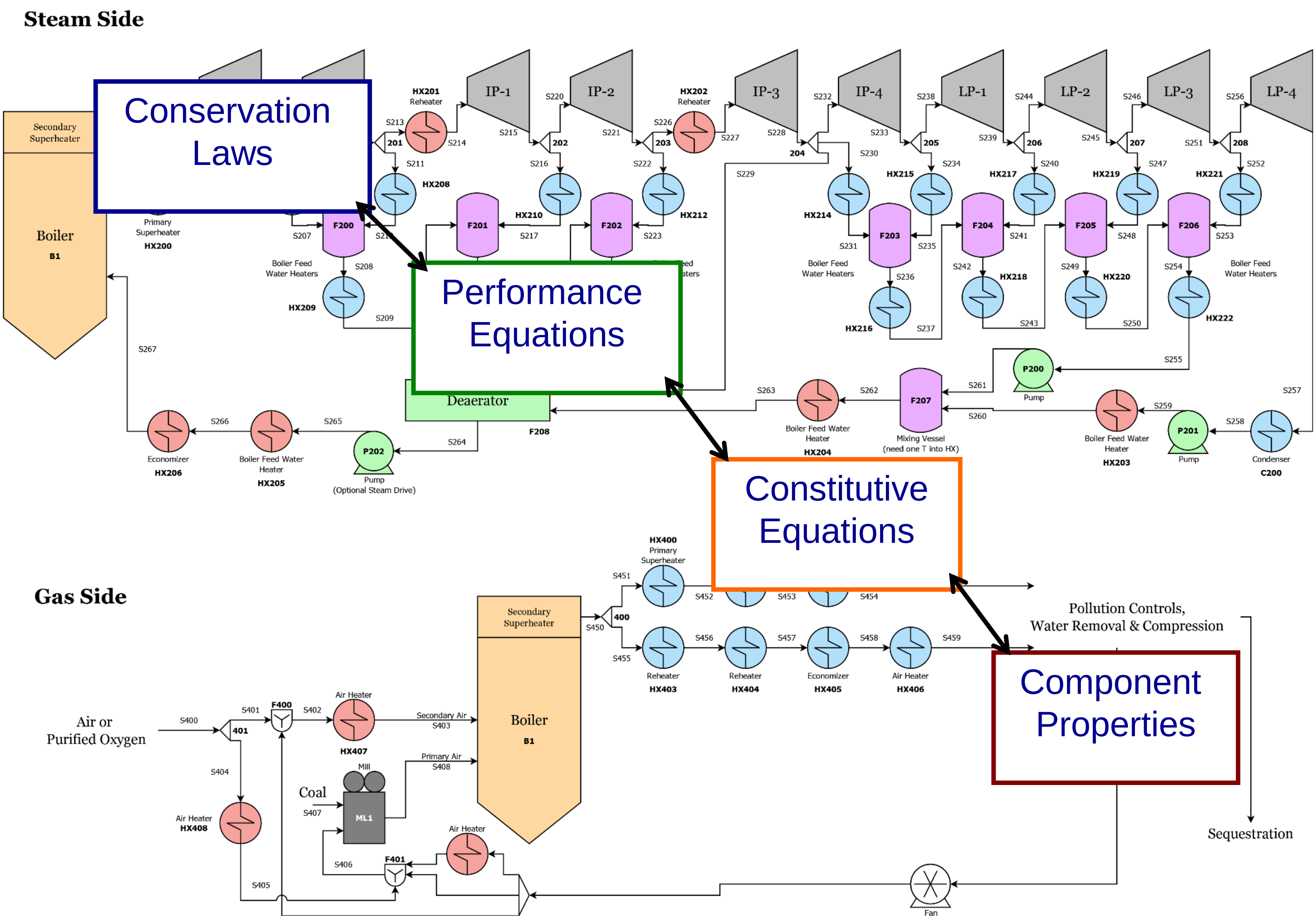
Process Optimization for Oxycombustion



Emerging Equation-Oriented Framework for Process Optimization

- Model in GAMS (or AMPL, AIMMS)
- Exact Jacobians/Hessians and sparse equation structure
- Fast Newton-based NLP solvers
- NLP sensitivity (post-optimality and interpretation, multi-level opt., …)
- EO-Modeling Enables:
  - Efficient MINLP Strategies
  - Efficient Global Optimization
  - Large-scale Optimization under Uncertainty

- But process models are not just equations!