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Norwegian University of Science and Technology

Optimal Operation of Integrated Chemical Processes

With Application to Ammonia Synthesis

Julian Straus

NTNU

17. August 2018

Presentation Outline

1. Introduction
   - Ammonia Process (Chapter 2)
   - Optimal Operation (Chapter 3)
2. Optimal Operation for Subprocesses
   - Economic Nonlinear Model Predictive Control (Chapter 5)
   - Self-optimizing Control with Extremum-Seeking Control (Chapter 6+7)
   - Feedback Real-time Optimization (Chapter 8)
3. Optimal Operation through Introduction of Surrogate Models
   - Main Procedure (Chapter 10)
   - Variable Reduction using PLS Regression (Chapter 11+12)
   - Application of Self-optimizing Variables (Chapter 13)
   - Sampling for Surrogate Model Generation (Chapter 14)
4. Conclusion

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Ammonia Process

- Haber Bosch process: Fixation of atmospheric nitrogen

$$3H_2 + N_2 \rightleftarrows 2NH_3$$

- Developed in 1910s
- Strong competition and high energy demand
→ Integration of the process
→ Difficult optimization of the process
- Split into 2 sections
  1. Synthesis gas production
  2. Ammonia synthesis

Optimal Operation

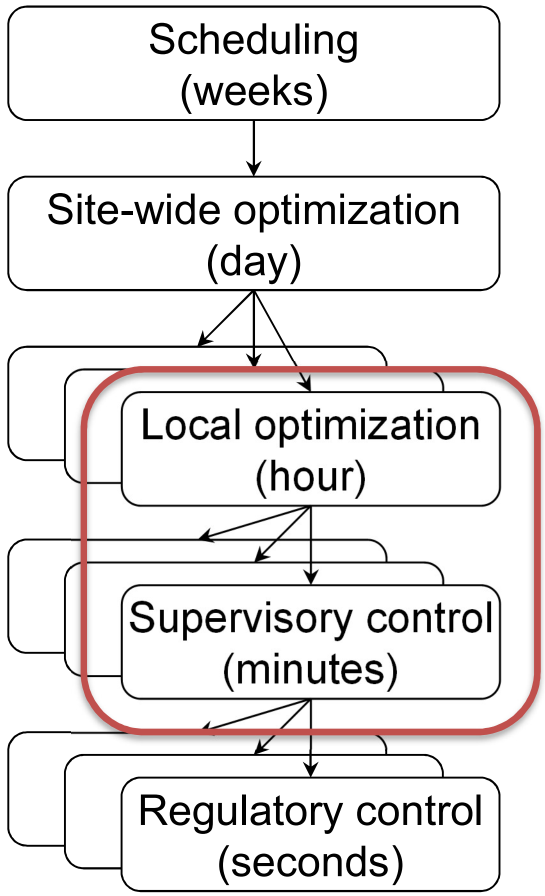
- Aim: minimizing production cost through process control
- Control structure frequently hierarchical
- Optimal operation results in an optimization problem

$$\min_{\mathbf{x}(\cdot),\mathbf{u}(\cdot)} \int_0^\infty J_{dyn}(\mathbf{x}(t),\mathbf{d}(t),\mathbf{u}(t))dt$$

$$s.t.\ \dot{\mathbf{x}} = \mathbf{f}\left(\mathbf{x}(t),\mathbf{d}(t),\mathbf{u}(t)\right), \qquad \forall t \in [0,\infty)$$

$$\mathbf{0} \geq \mathbf{h}\left(\mathbf{x}(t),\mathbf{d}(t),\mathbf{u}(t)\right), \qquad \forall t \in [0,\infty)$$

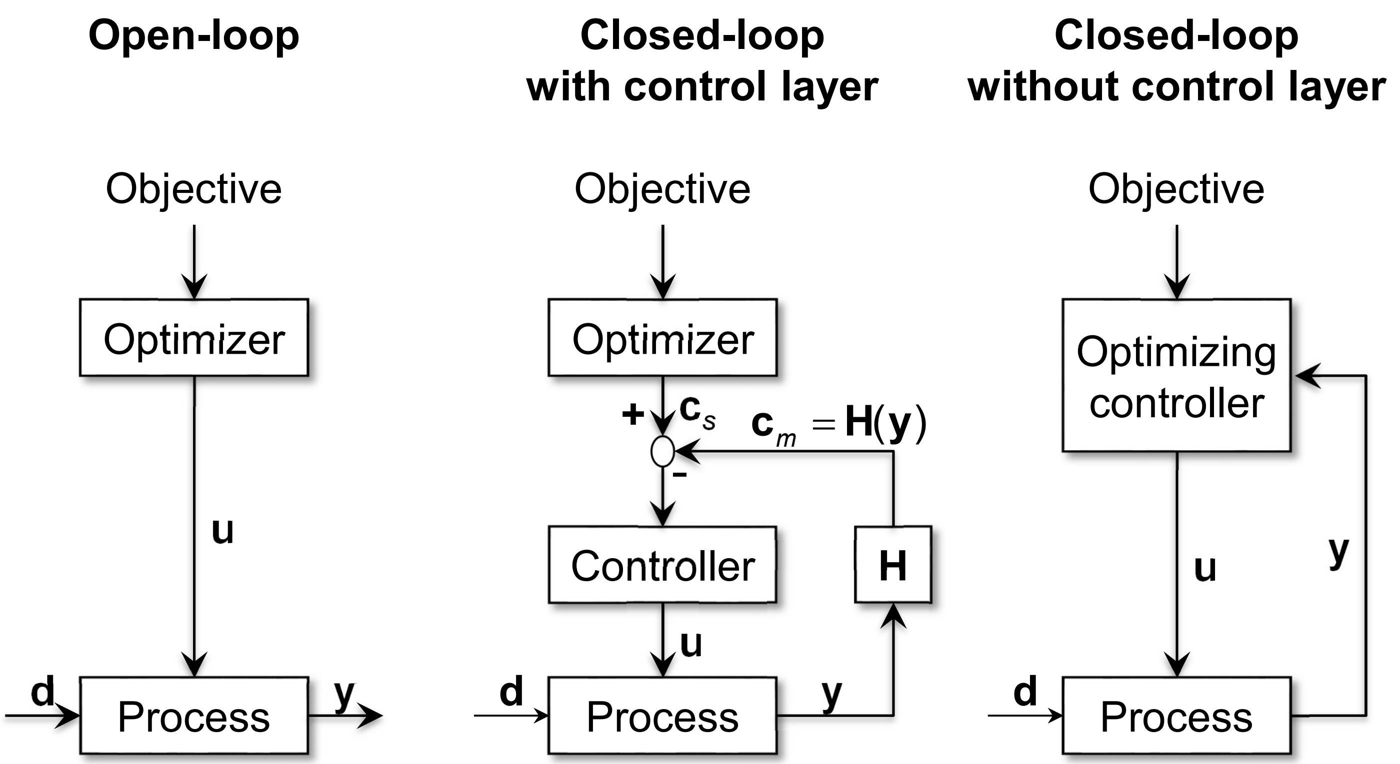
$$\mathbf{x}(0) = \mathbf{x}_0$$



S. Skogestad, Plantwide control: the search for the self-optimizing control structure, *J. Proc Control.* 10 (2000) 487-506

Optimal Operation

- Implementation of optimal operation



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1. Introduction

Optimal Operation - Integrated Processes

- Optimizer for dynamic or steady-state optimization problem required

  → Model of the process

- Problems of integrated process:
  - Nested Recycle Loops
  - Convergence of the Flowsheet
  - Simulation noise
- Two different approaches
  1. Optimal operation of subprocesses
  2. Simplified model for optimization of the overall process

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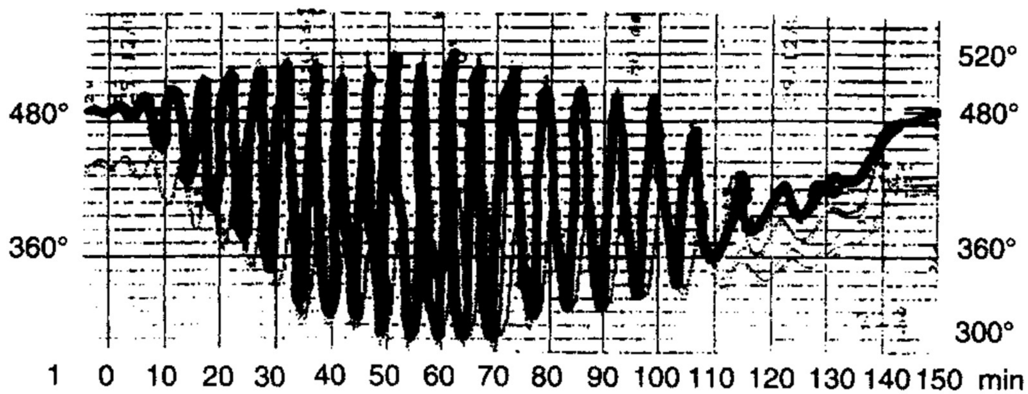
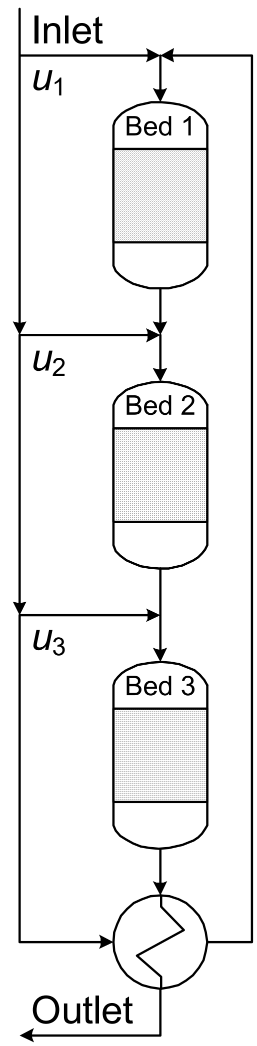
Case Study – Ammonia Reactor

- Three bed ammonia reactor
- Three manipulated variables **u**
- Heat integration for reduced cost through reactor outlet heat exchanger
- Cost function: rate of extent of reaction ξ

$$\dot{\xi} = \dot{m}_{\text{Inlet}} \left(w_{\text{NH}_3,\text{Outlet}} - w_{\text{NH}_3,\text{Inlet}} \right) \quad \text{in [kg NH}_3\text{/s]}$$

- Exhibits limit cycle and reactor extinction





J. Morud, S. Skogestad, Analysis of Instability in an Industrial Ammonia Reactor, *AIChE J.* 44 (1998) 888-895

Economic Nonlinear Model Predictive Control

- Solves dynamic optimization problem for a time horizon t_{max}

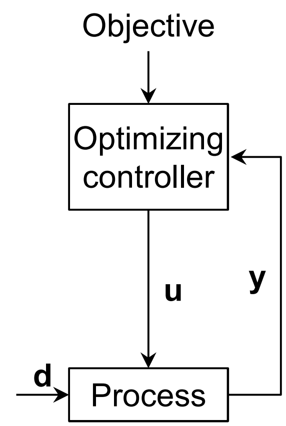
$$\min_{\mathbf{x}(\cdot),\mathbf{u}(\cdot)} \int_0^{t_{max}} J_{dyn}(\mathbf{x}(t),\mathbf{d}(t),\mathbf{u}(t))dt$$

$$s.t. \quad \dot{\mathbf{x}} = \mathbf{f}\left(\mathbf{x}(t),\mathbf{d}(t),\mathbf{u}(t)\right), \qquad \forall t \in [0,t_{max}]$$

$$\mathbf{0} \geq \mathbf{h}\left(\mathbf{x}(t),\mathbf{d}(t),\mathbf{u}(t)\right), \qquad \forall t \in [0,t_{max}]$$

$$\mathbf{x}(0) = \mathbf{x}_0$$

Closed-loop without control layer



- Implements first calculated input of the trajectory
- Problems:
  - Required time for solving the optimization problem
  - Feasibility of the solution to the optimization problem and stability

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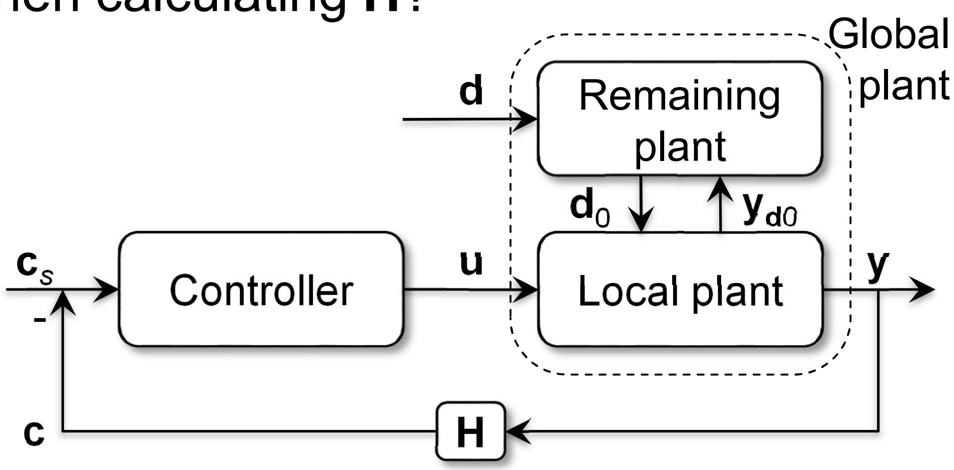
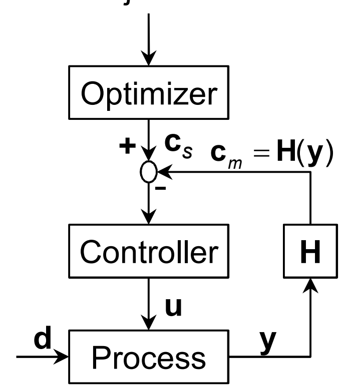
2. Optimal Operation of Subprocesses

Self-optimizing Control (SOC)

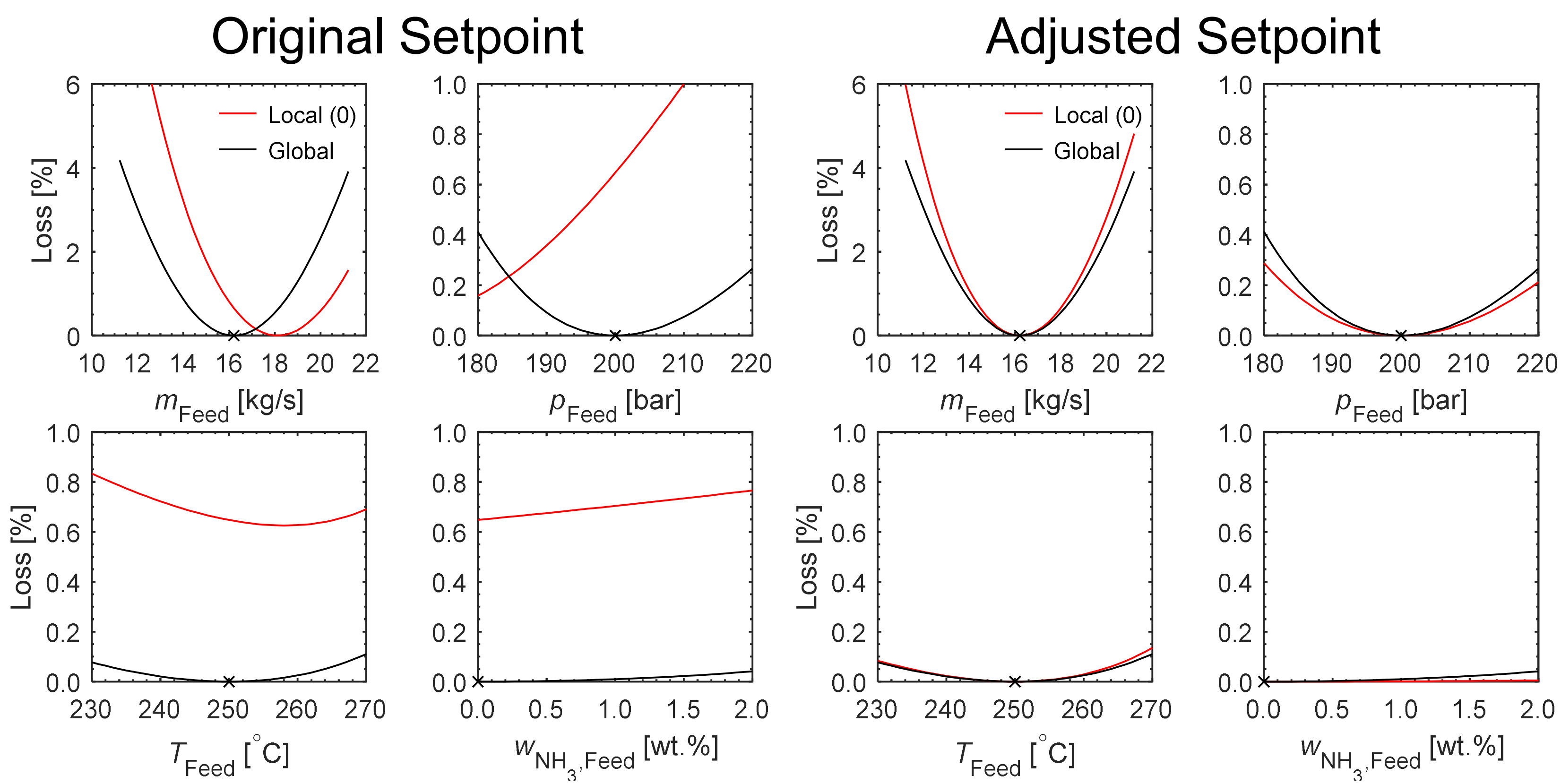
- Constant setpoint policy
  → Selection of controlled variables $\mathbf{c} = \mathbf{H}\mathbf{y}$
- Based on steady-state optimization considering the disturbances and local linearization
- What happens if remaining plant is neglected when calculating **H**?



Closed-loop with control layer



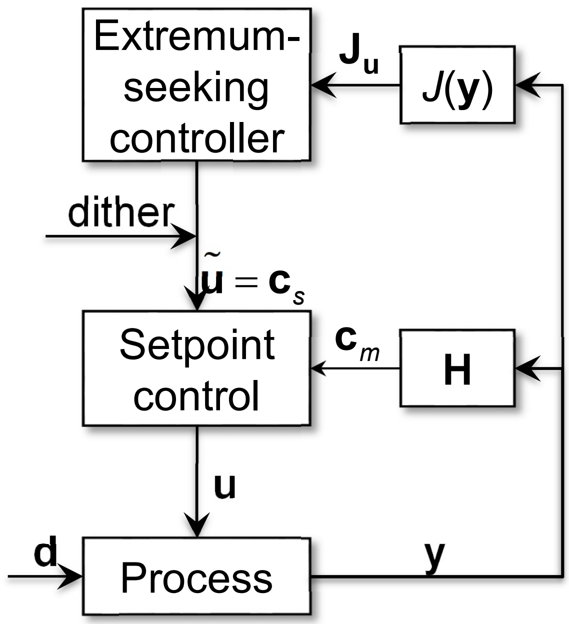
Self-optimizing Control (SOC)



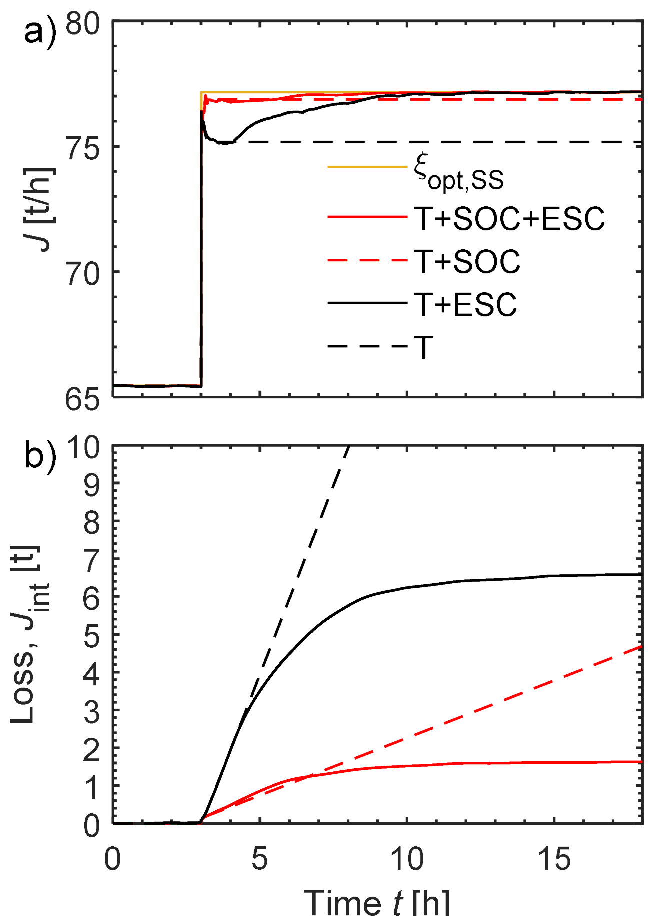
→ Requires adjustment of setpoint to controllers

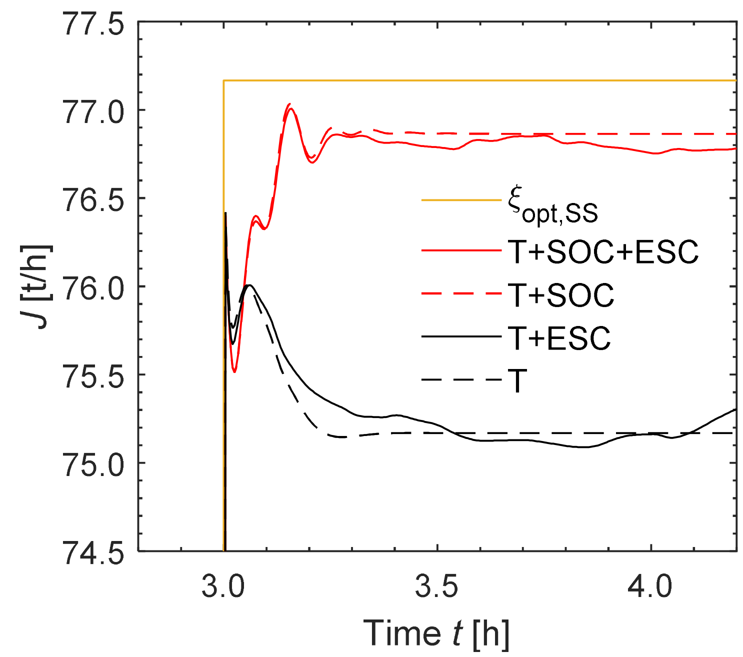
Self-optimizing Control (SOC) + Extremum-seeking Control (ESC)

- Extremum-seeking control as optimizing layer for setpoint adjustment
- Self-optimizing control for fast close-to-optimal disturbance rejection



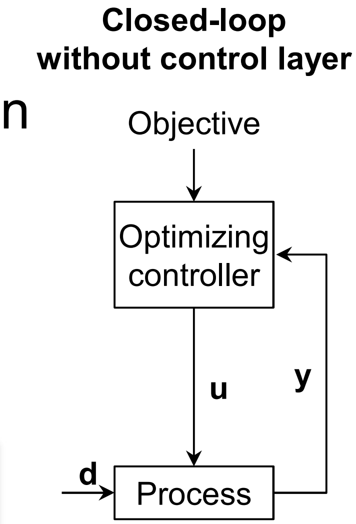
Self-optimizing Control (SOC) + Extremum-seeking Control (ESC)

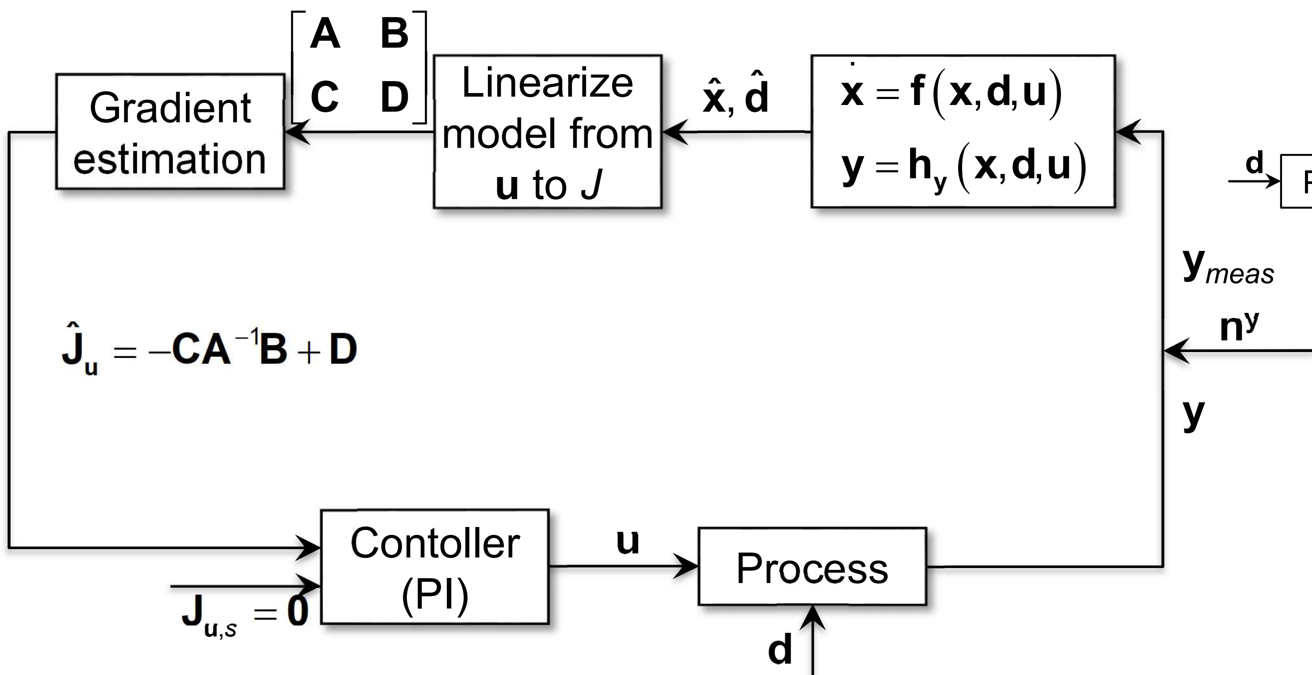




Feedback Real-time Optimization

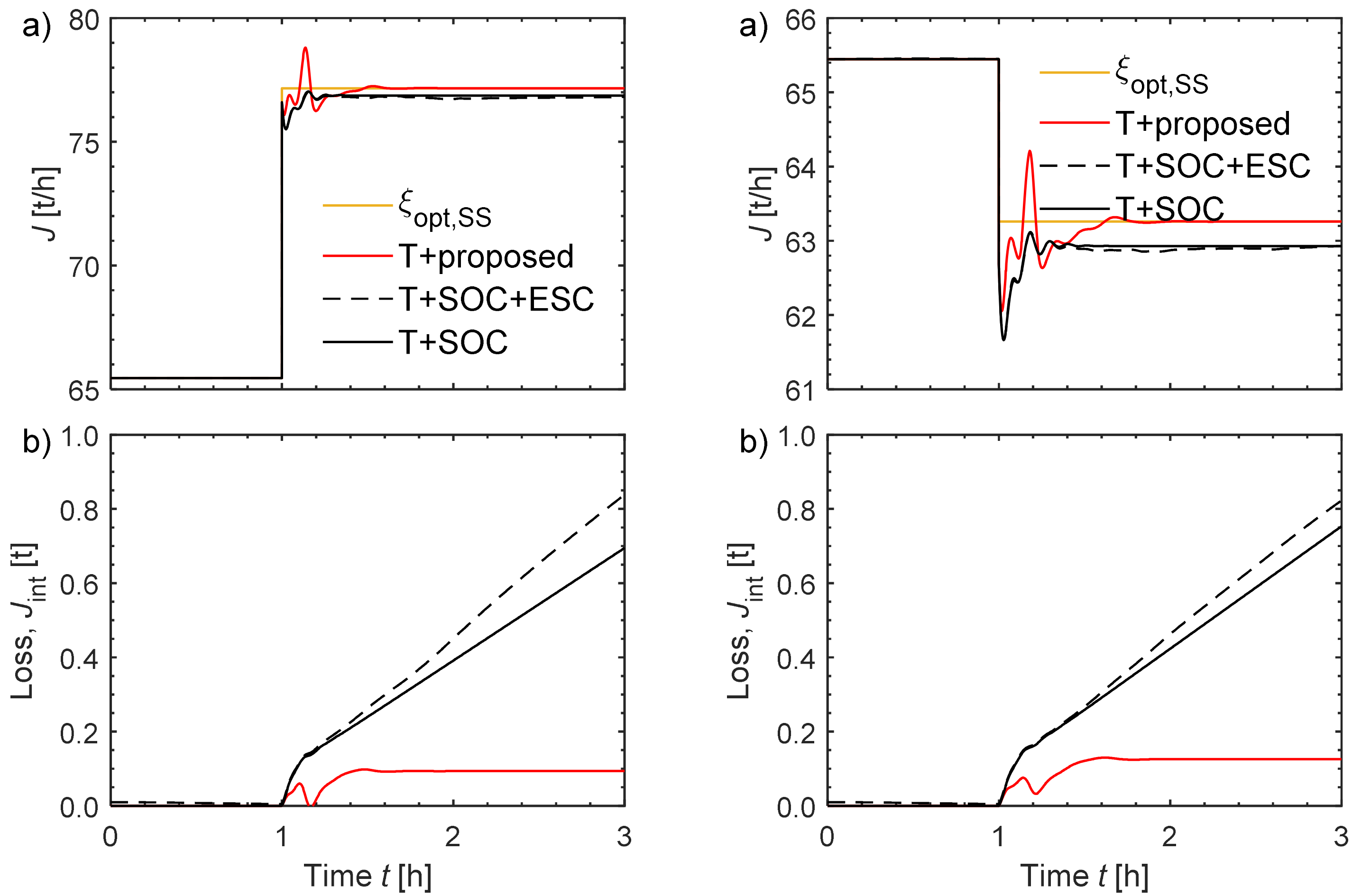
- ESC rather slow
- E-NMPC complicated through online optimization
- Translation of optimization problem into a feedback problem:





Feedback Real-time Optimization

Flowrate Disturbance (measured) | **Activity disturbance (estimated)**



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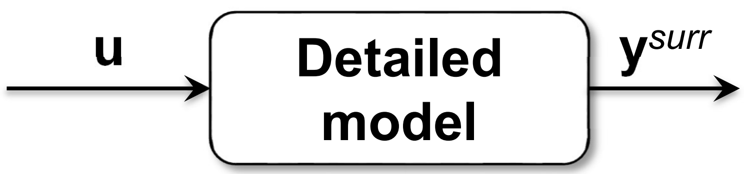
Surrogate Model

- Detailed models often computational expensive to solve
- Introduction of surrogate models reduces computation load
- Surrogate model:
  Simple input (**u**)-output (**y**^{surr}) representation (**y**^{surr} $=$ **g**'(**u**)) of a detailed model



- Input: Connection and decision variables
- Output: Connection and economic variables

Main Procedure

Original (steady-state) optimization problem:

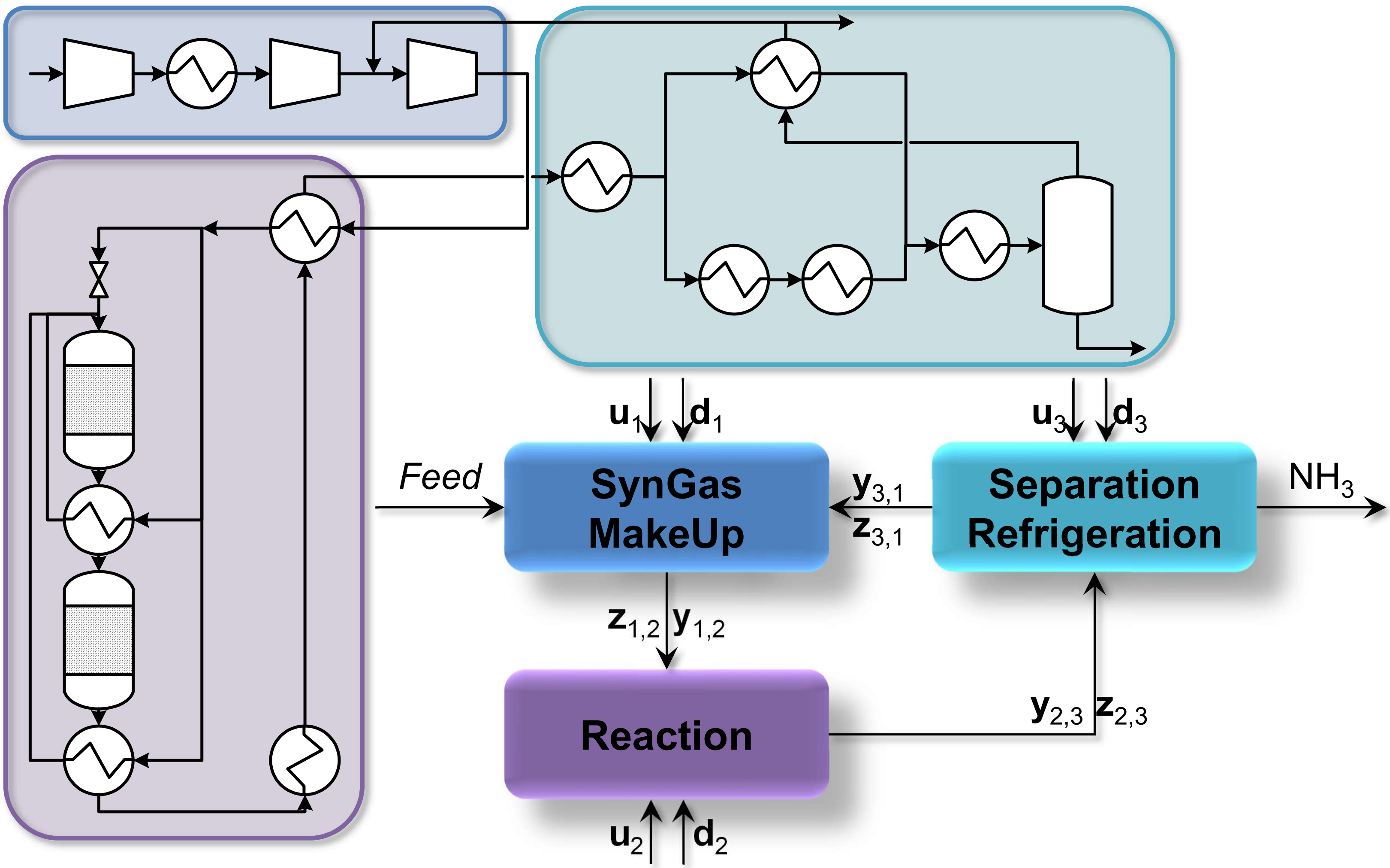
$$\min_{\mathbf{x},\mathbf{u}} \; J(\mathbf{x},\mathbf{d},\mathbf{u})$$
$$s.t. \; \mathbf{0} = \mathbf{g}(\mathbf{x},\mathbf{d},\mathbf{u})$$
$$\mathbf{0} \geq \mathbf{h}(\mathbf{x},\mathbf{d},\mathbf{u})$$

1. Split original model $\mathbf{g}$ into n submodels $\mathbf{g}_i$
2. Calculate surrogate models $\mathbf{g}_{i,k}$' for submodels $\mathbf{g}_i$
3. Combine surrogate models in big model through connection constraints $\mathbf{0} = \mathbf{z}_{i,k} - \mathbf{y}_{i,k}$
4. Optimize new problem

$$\min_{\mathbf{x},\mathbf{u}} \; J(\mathbf{x},\mathbf{u},\mathbf{d})$$
$$s.t. \; \mathbf{0} = \mathbf{y}_{i,k} - \mathbf{g}_{i,k}'\left(\mathbf{d}_i,\mathbf{z}_{k,i},\mathbf{u}_i\right) \quad i \in 1\ldots n, \forall k \neq i$$
$$\mathbf{0} = \mathbf{z}_{i,k} - \mathbf{y}_{i,k} \quad i \in 1\ldots n, \forall k \neq i$$
$$\mathbf{0} \geq \mathbf{h}_i\left(\mathbf{x}_i,\mathbf{d}_i,\mathbf{u}_i\right) \quad i \in 1\ldots n$$

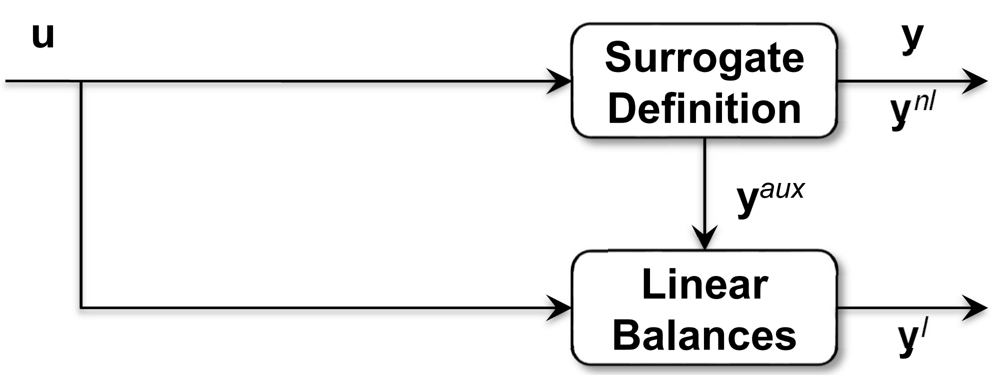
Main Procedure - Ammonia Synthesis

$$0 = \mathbf{z}_{1,2} - \mathbf{y}_{1,2}$$
$$0 = \mathbf{z}_{3,1} - \mathbf{y}_{3,1}$$
$$0 = \mathbf{z}_{2,3} - \mathbf{y}_{2,3}$$



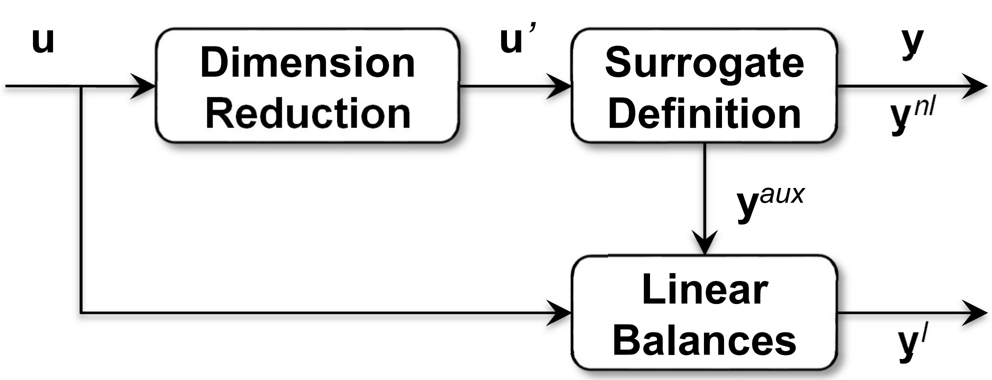
Variable Reduction using PLSR

- Connection variables can result in high number of independent variables n_u
- Sampling and surrogate model fitting computation expensive with high n_u
- Reduction necessary:
  1. Introduction of linear mass balances



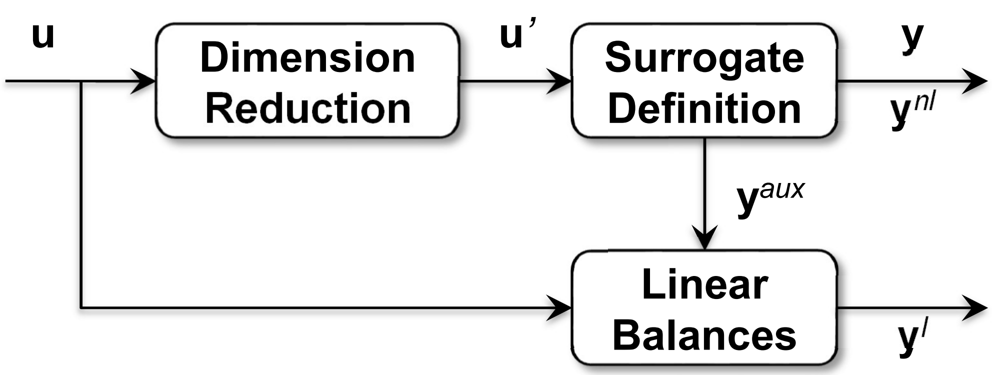
Variable Reduction using PLSR

- Connection variables can result in high number of independent variables n_u
- Sampling and surrogate model fitting computation expensive with high n_u
- Reduction necessary:
  1. Introduction of linear mass balances
  2. Reduction of n_u through PLSR: $\mathbf{u}' = \mathbf{W}^\mathsf{T}\mathbf{u}$



Variable Reduction using PLSR

- Connection variables can result in high number of independent variables n_u
- Sampling and surrogate model fitting computation expensive with high n_u
- Reduction necessary:
  1. Introduction of linear mass balances
  2. Reduction of n_u through PLSR: $\mathbf{u}' = \mathbf{W}^\mathsf{T}\mathbf{u}$
  3. Fitting of surrogate model using new latent and dependent variables



$$\mathbf{y}^{surr} = \begin{bmatrix} \mathbf{y}^{nl} \\ \mathbf{y}^{aux} \end{bmatrix}$$

$$\mathbf{y} = \begin{bmatrix} \mathbf{y}^{nl} \\ \mathbf{y}^{l} \end{bmatrix}$$

$$n_{u'} < n_u$$

Variable Reduction using PLSR

- Reaction section, ammonia synthesis loop
  - 7 feed variables
    $$\mathbf{z} = \left[p_{in} \quad T_{in} \quad \dot{n}_{H_2,in} \quad \dot{n}_{N_2,in} \quad \dot{n}_{NH_3,in} \quad \dot{n}_{Ar,in} \quad \dot{n}_{CH_4,in} \right]^T$$
  - 3 manipulated variables
    $$MV = \left[n_{S1} \quad n_{S2} \quad T_{HEx} \right]^T$$
  - 3 dependent variables
    $$\mathbf{y}^{surr} = \left[p_{out} \quad T_{out} \quad \xi \right]^T$$
- Surrogate model structure

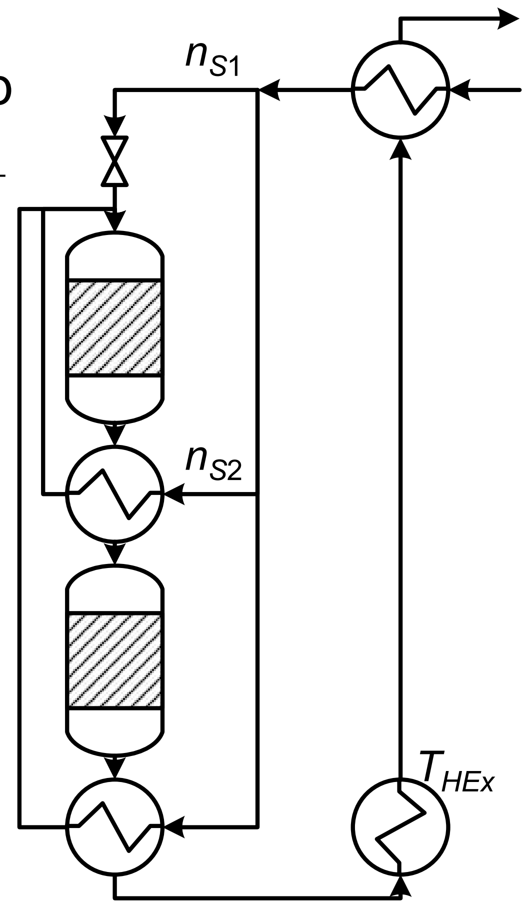
$$\mathbf{u}'_k = \mathbf{W}_k^T \mathbf{u} \qquad k = 1,2,3 \qquad (1)$$

$$p_{out} = g_1(\mathbf{u}'_1) \qquad (2)$$

$$T_{out} = g_2(\mathbf{u}'_2) \qquad (3)$$

$$\xi = g_3(\mathbf{u}'_3) \qquad (4)$$

$$\dot{n}_{i,out} = \dot{n}_{i,in} + \nu_i \xi \qquad (5)$$



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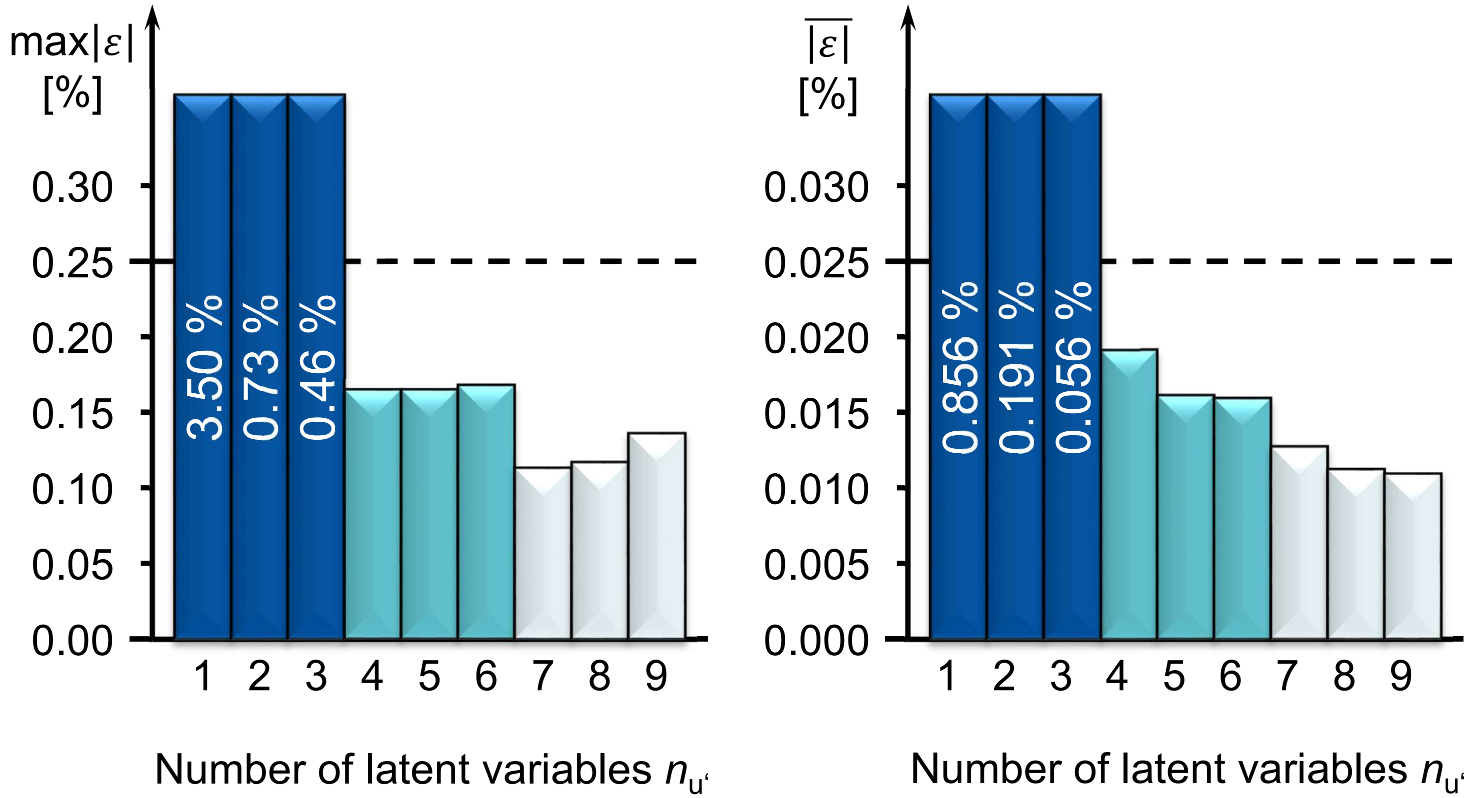
Optimal Operation through Introduction of Surrogates

3

Variable Reduction using PLSR - Outlet Pressure



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Optimal Operation through Introduction of Surrogates

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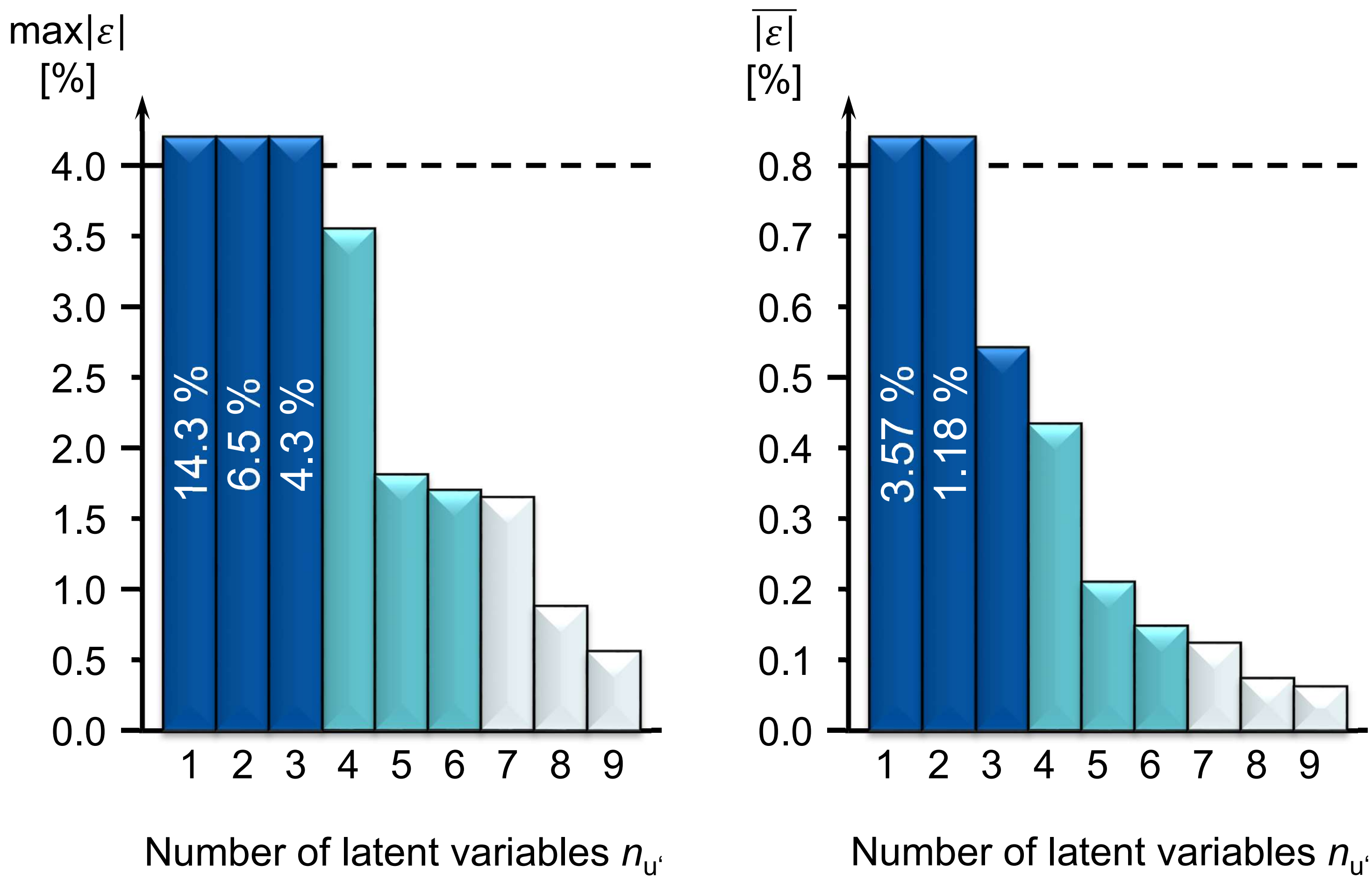
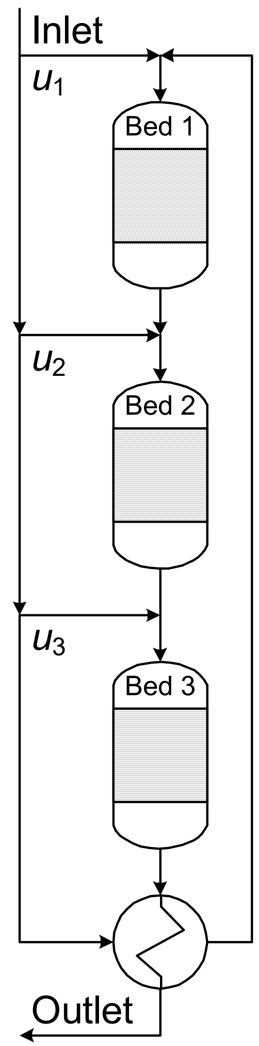
Variable Reduction using PLSR - Extent of Reaction



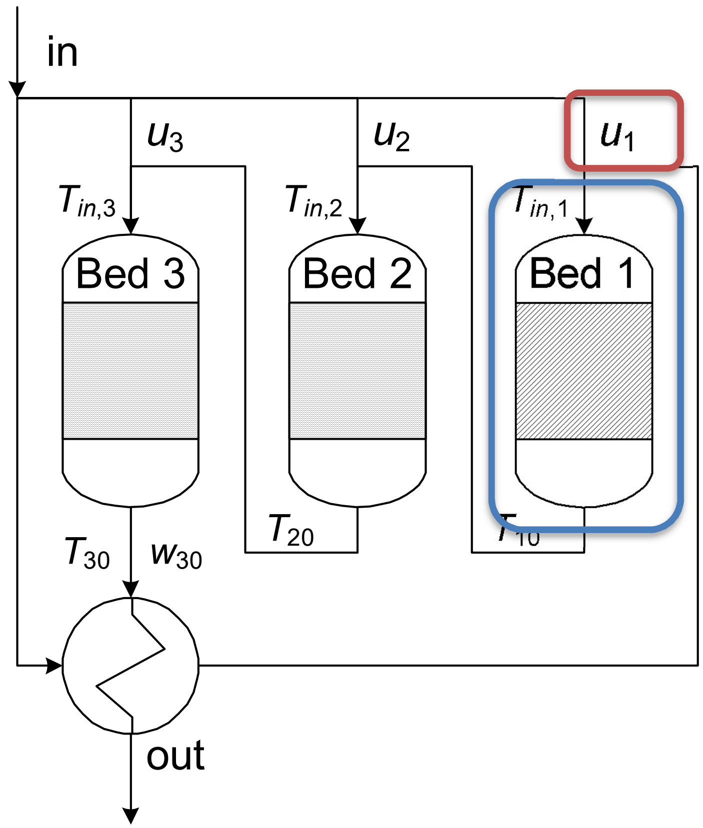
Application of Self-optimizing Variables

- Idea: Simplify response surface through change of independent variables (sample interesting regions)
- Initial independent variables:
  - Feed
    $$\mathbf{z} = \begin{bmatrix} \dot{m}_{in} & p_{in} & T_{in} & w_{NH_3,in} & R_{H_2/N_2,in} \end{bmatrix}^T$$
  - Manipulated variables
    $$\mathbf{u} = \begin{bmatrix} u_1 & u_2 & u_3 \end{bmatrix}^T$$
- Initial dependent variables
  - Output
    $$\mathbf{y}^{surr} = \begin{bmatrix} \xi \\ T_{out} \end{bmatrix}$$
- Change of variables from **u** to **c** *via* self-optimizing control principles, *i.e*. add equality constraints:
  $$\mathbf{g}_{SOC} = \mathbf{c} - \mathbf{H}\mathbf{y} = \mathbf{0}$$



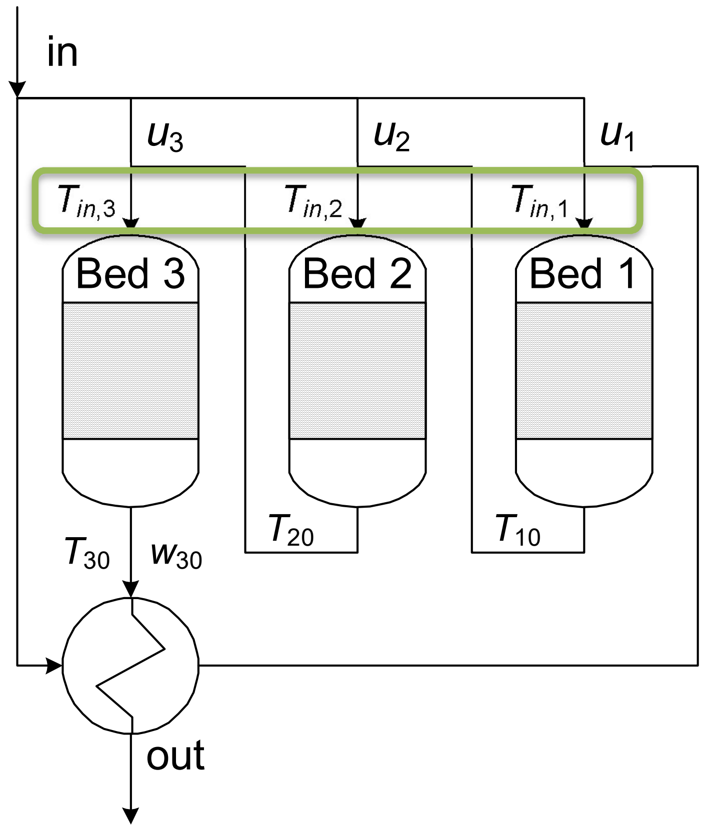
Application of Self-optimizing Variables

- Aim: Maximize rate of extent of reaction
- Local SOC variables per bed



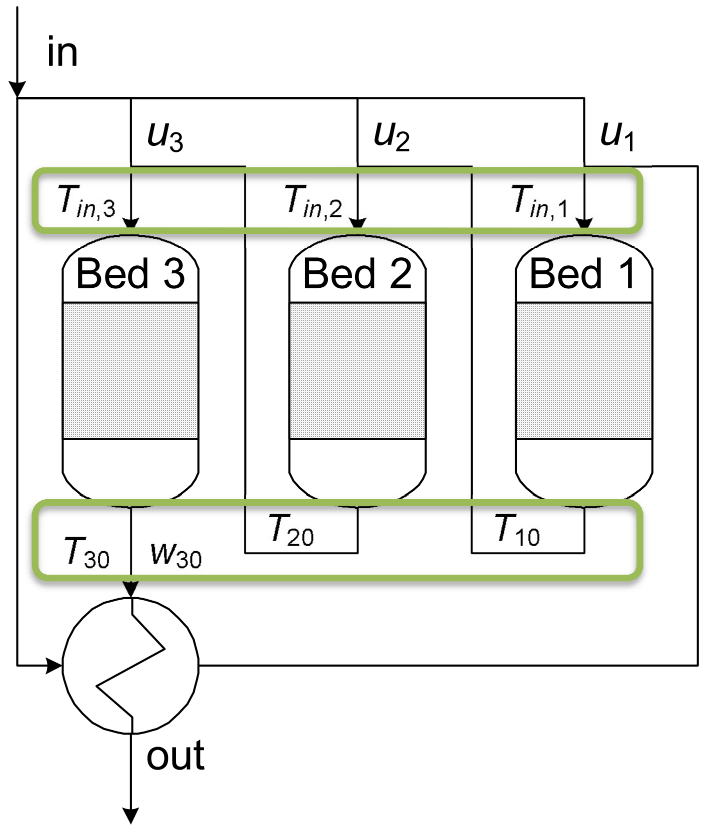
Application of Self-optimizing Variables

- Aim: Maximize rate of extent of reaction
- Local SOC variables per bed
- 4 different SOC variable combination tested
  - Inlet temperatures



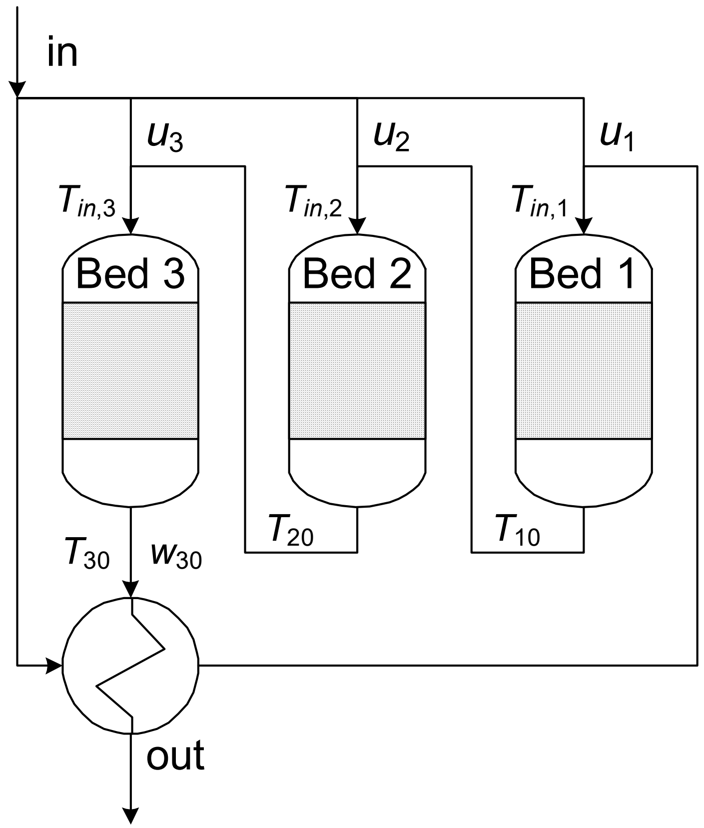
Application of Self-optimizing Variables

- Aim: Maximize rate of extent of reaction
- Local SOC variables per bed
- 4 different SOC variable combination tested
  - Inlet temperatures
  - Inlet and outlet temperatures



Application of Self-optimizing Variables

- Aim: Maximize rate of extent of reaction
- Local SOC variables per bed
- 4 different SOC variable combination tested
  - Inlet temperatures
  - Inlet and outlet temperatures
  - 1 optimal temperature per bed
  - 2 optimal temperatures per bed
- Error with respect to true optimum



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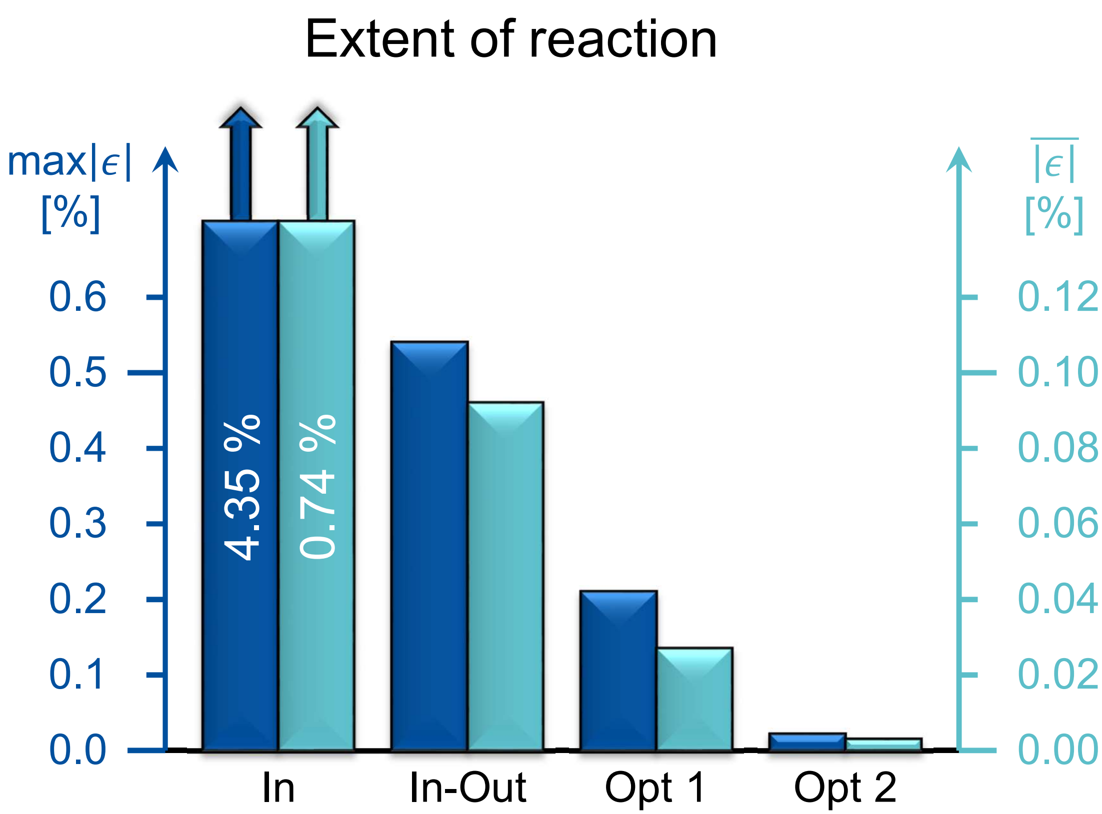
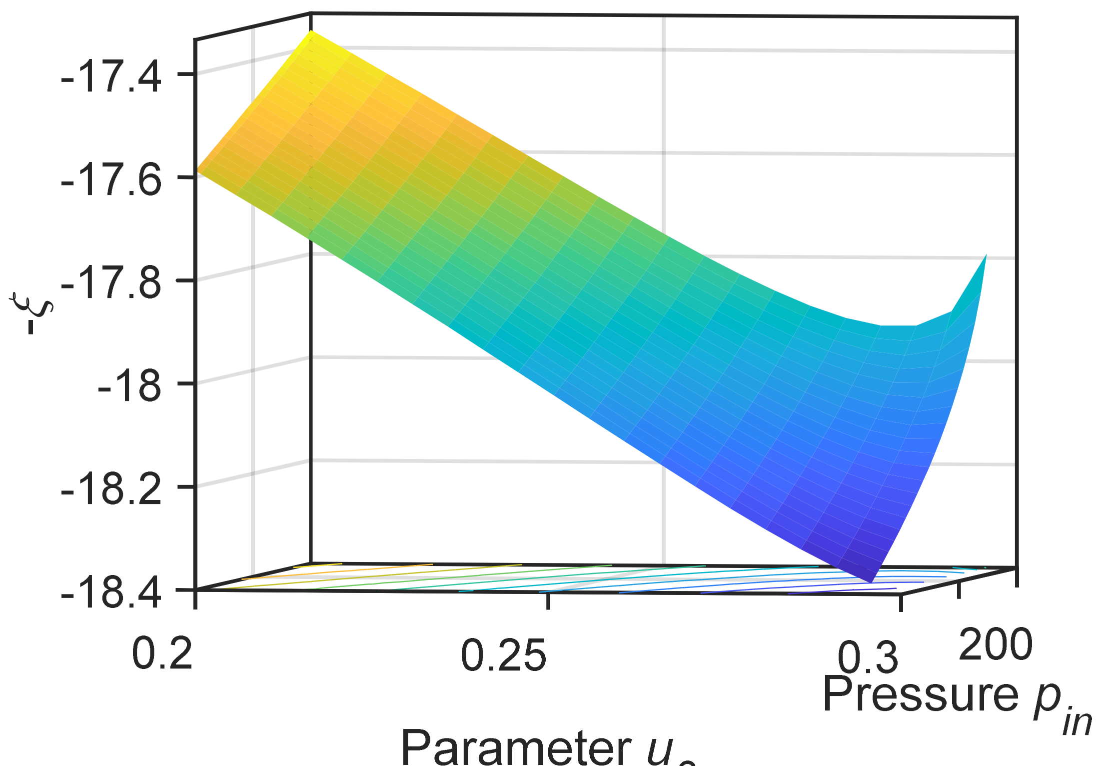


Application of Self-optimizing Variables

- Limit cycle behavior and reactor extinction close to optimal point
- Complicates normal sampling



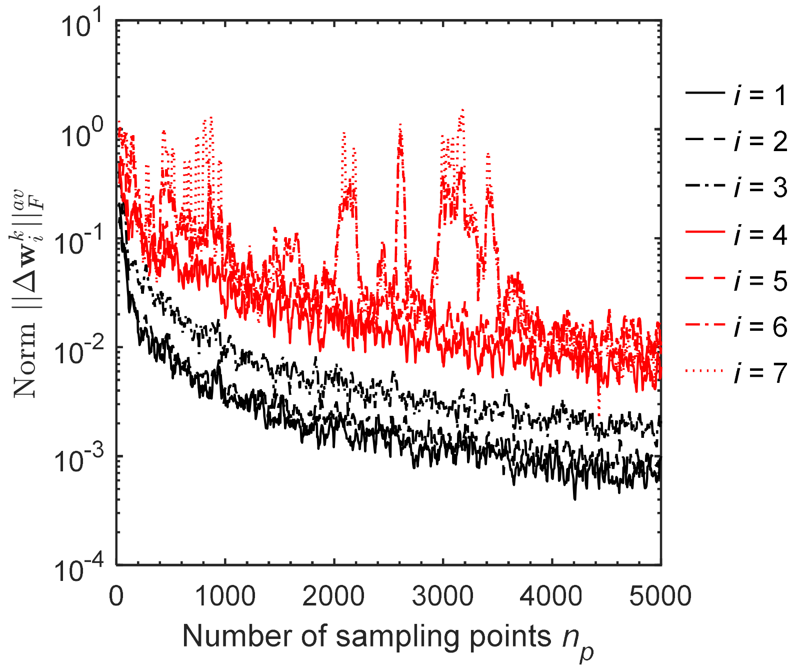
Sampling for Surrogate Model Generation

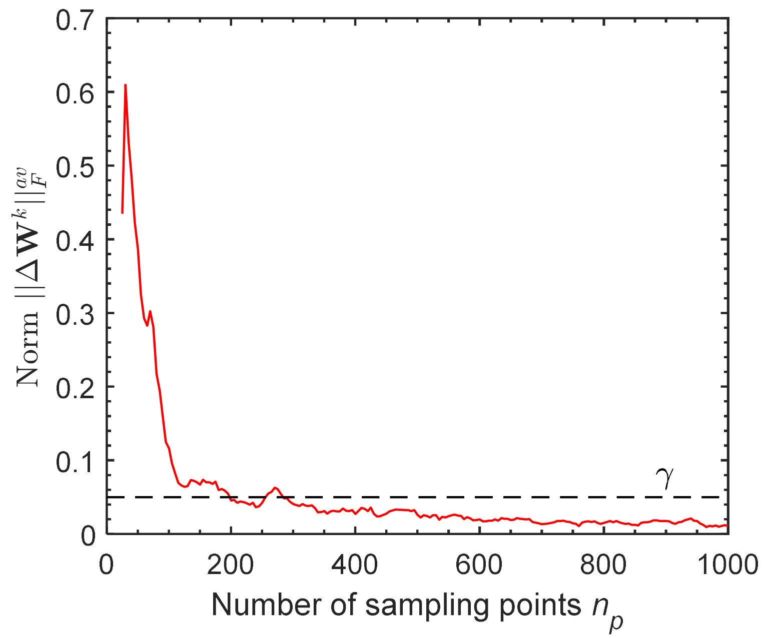
- Sampling crucial for:
  - Performance of surrogate model
  - Computational expense
- Common sampling approaches
  - Predefined
  - Adaptive
- Aim: Sampling without
  - Surrogate model fitting
  - Over-sampling

Development of a sampling method based on partial least square regression

Sampling for Surrogate Model Generation

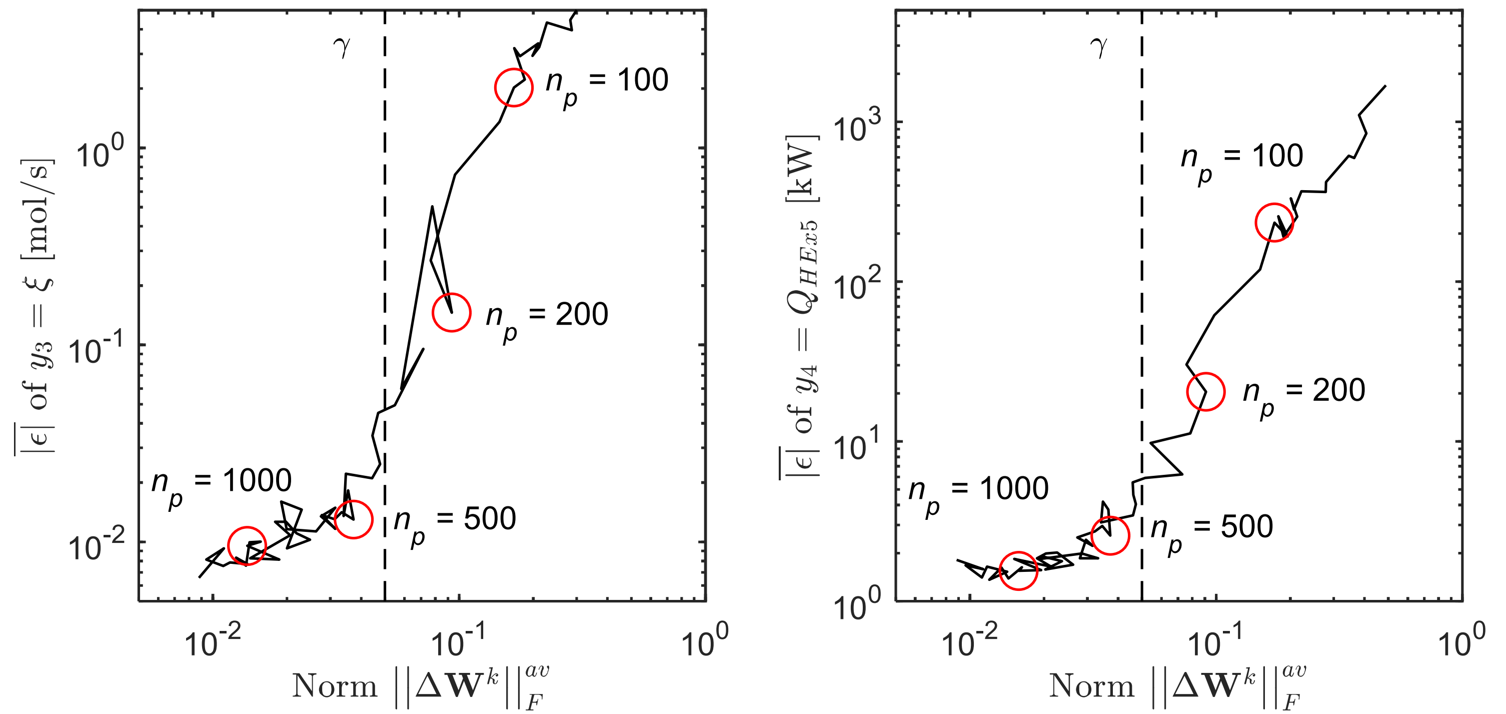
- Weights $\mathbf{W}^k$ change with growing sampling space ($\mathbf{u}' = \mathbf{W}^\top \mathbf{u}$)
- Convergence of the significant weights





Sampling for Surrogate Model Generation

- Convergence corresponds to flattening in error improvement: Reaction section ($n_p = 2000$ sampled points)



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Conclusion

- Optimal operation methods
  - Self-optimizing control in recycle systems
  - Combination of self-optimizing control and extremum-seeking control for removal of steady-state loss
  - Feedback real-time optimization for fast disturbance rejection

- Optimization of integrated process
  - Method for surrogate model-based optimization
  - Independent variable reduction through PLS regression
  - Simplification of response surface through self-optimizing variables
  - Termination criteria for sampling without the need of surrogate model fitting

Thank you for attending my defense