From sensor tips to state and parameter estimates

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COOPOL
Control and Real-time Optimisation of Intensive Polymerisation
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AVT Aachener Verfahrenstechnik | RWTH AACHEN UNIVERSITY

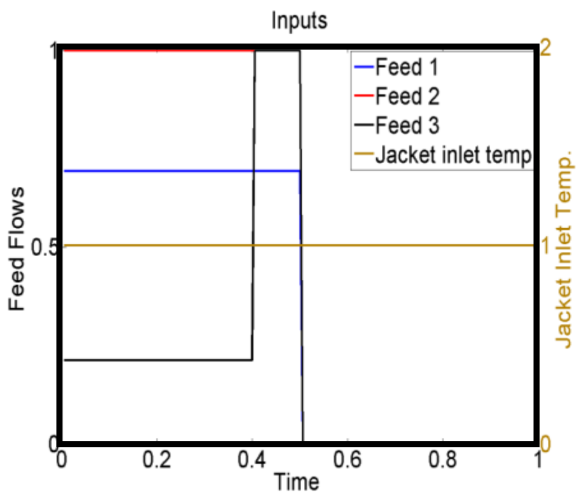
Outline

- Motivation: Recipe-based process operation and its drawbacks
- State-based operation.
- State estimation and observability
- Parameter estimation and identifiability
- Raman spectra to concentration and particle sizes.
- Summary

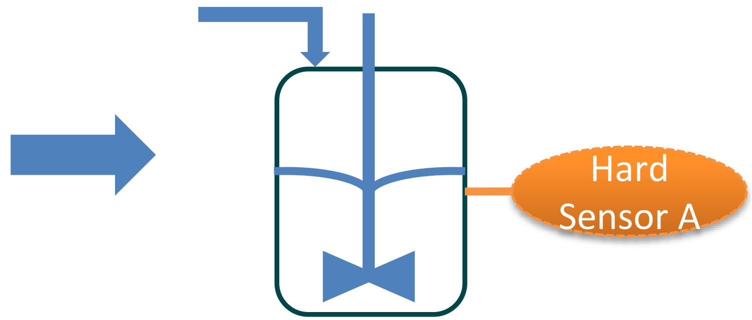
Recipe-based Operation

- Current industrial practice.

- Fixed recipe



- Operation based on heuristics and experience.



- Process monitoring using few sensors

Drawbacks of time-based operation:

- Susceptibility to process disturbances.
- Online process optimization not possible.

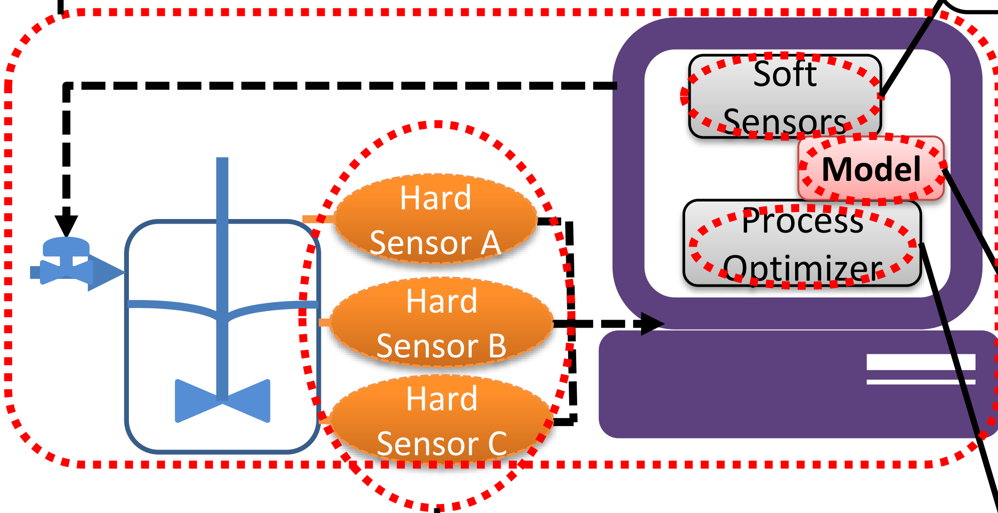
Can be solved by

Model-based control/
State-based operation

State-based Operation

Data processing framework for data transfer between different components.

Soft sensors (state estimators) to predict unmeasured states reliably.



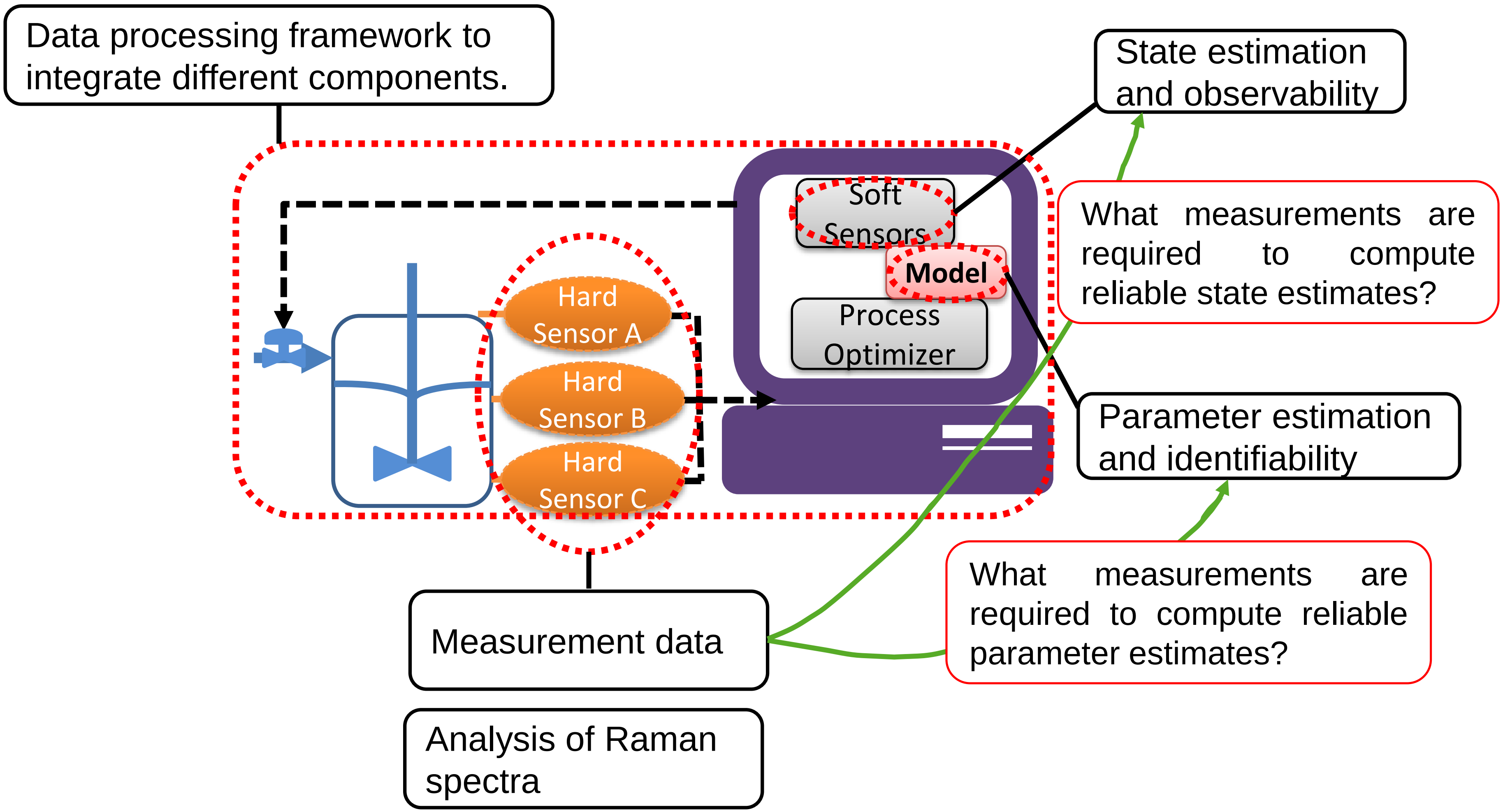
Model identification & parameter estimation techniques for accurate models.

Sensors for robust & reliable online measurements.

Techniques for sensor data analysis.

Robust, online process optimizer to control and optimize the process.

Further Topics



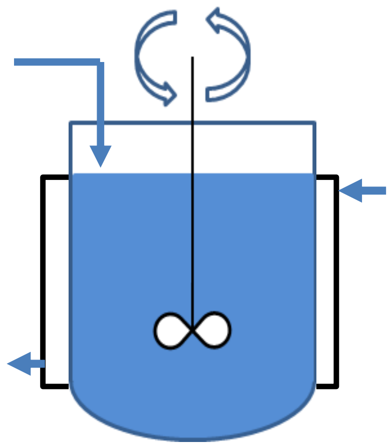
Considered Case Study

4 Monomer semi-batch emulsion polymerization.

Model

- 48 Differential equations
- ~300 Algebraic equations

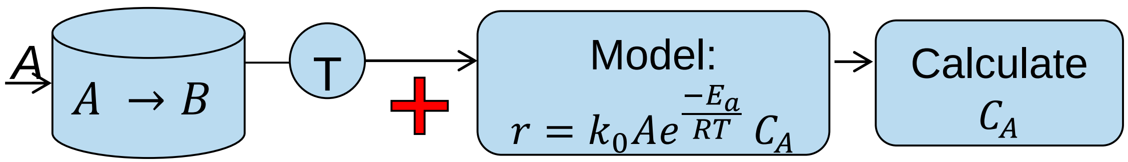
- Mass transfer
- Kinetics of monomer conversion
- Kinetics of polymer chain growth
- Heat transfer



Possible Measurements

- Reactor temperature
- Coolant inlet temperature
- Coolant outlet temperature
- Enthalpy of reaction
- Monomer conversions
- No. Avg. Molecular weight (MN)
- Wt. Avg. Molecular weight (MW)

State Estimation and Observability



State estimation algorithms: Extended Kalman Filter (EKF)

Moving Horizon Estimators (MHE)

Particle Filters

Challenge in polymerization systems: Is it possible to estimate all the states with available measurement data?

Observability:

- Can all states be estimated?
- Which measurements are required to ensure complete state observability?

Observability Analysis in Case Study

- Definition: A state is said to be observable if it can be uniquely determined from measurements in a finite time[1].
- The semi-batch emulsion polymerization system has a high degree of nonlinearity → A nonlinear observability analysis should be performed.
- Nonlinear observability methods not yet tractable for large scale processes.
- Linear observability analysis performed on models linearized at multiple time points.
- Case I: Observability analysis with measurements
  - Reactor temperature
  - Coolant inlet temperature
  - Coolant outlet temperature
  - Enthalpy of reaction
  - Monomer conversions
  - No. Avg. Molecular weight
  - Wt. Avg. Molecular weight

All states are observable!

[1]Kalman, R. E., On the general theory of control systems, *Proc. First IFAC Congress Automatic Control, Moscow*, 1:481-492,1959

Are all measurements available online?

Measurements	Commonly used Analysis techniques	Analysis time*
MN, MW	GPC	40 min
Monomer Conversions	GC, HPLC	25 min

* Does not include time for sample preparation

MN, MW, Monomer conversion require significantly long analysis times!

→ Cannot be used for online estimation.

Observability analysis with online measurements

- Reactor temperature
- Coolant inlet temperature
- Coolant outlet temperature
- Enthalpy of reaction

Not all states are observable!

States in Emulsion Polymerization

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States in a semi-batch emulsion polymerization model

Subsystem I

States related to material and energy balance, e.g.

- Moles of unreacted monomer
- Reactor temp.

Subsystem II

States related to molecular properties of polymer, e.g.

- Moments of molecular weight

Observable only if online polymer property measurements are available[1,2].

Also shown by our observability analysis.

[1]Kozub D. J., Macgregor J. F., *Chemical Engineering Science*, 47 (5): 1047-1062, 1992.

[2]Schuler H., and Suzhen Z., *Chemical Engineering Science*, 40(10): 1891-1904, 1985.

Observability of Material and Energy Balance

Case II: Observability of only material and energy balance equations tested with reduced model and measurements:

- Reactor temperature
- Coolant inlet temperature
- Coolant outlet temperature
- Enthalpy of reaction

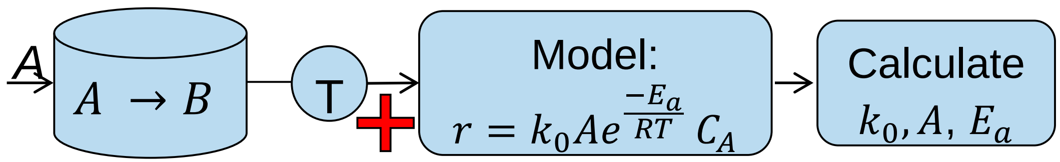
Not all states are observable!

Case III: Material and energy balance with measurements:

- Reactor temperature
- Coolant inlet temperature
- Coolant outlet temperature
- Enthalpy of reaction
- Monomer conversions

All states are observable! → Monomer conversion
estimation of materia

Parameter Estimation and Identifiability



Parameter estimation algorithms: Weighted Least Squares
Maximum Likelihood Estimation
} Offline methods

Parameter estimation may also be carried out online using with state estimation algorithms (e.g. EKF)

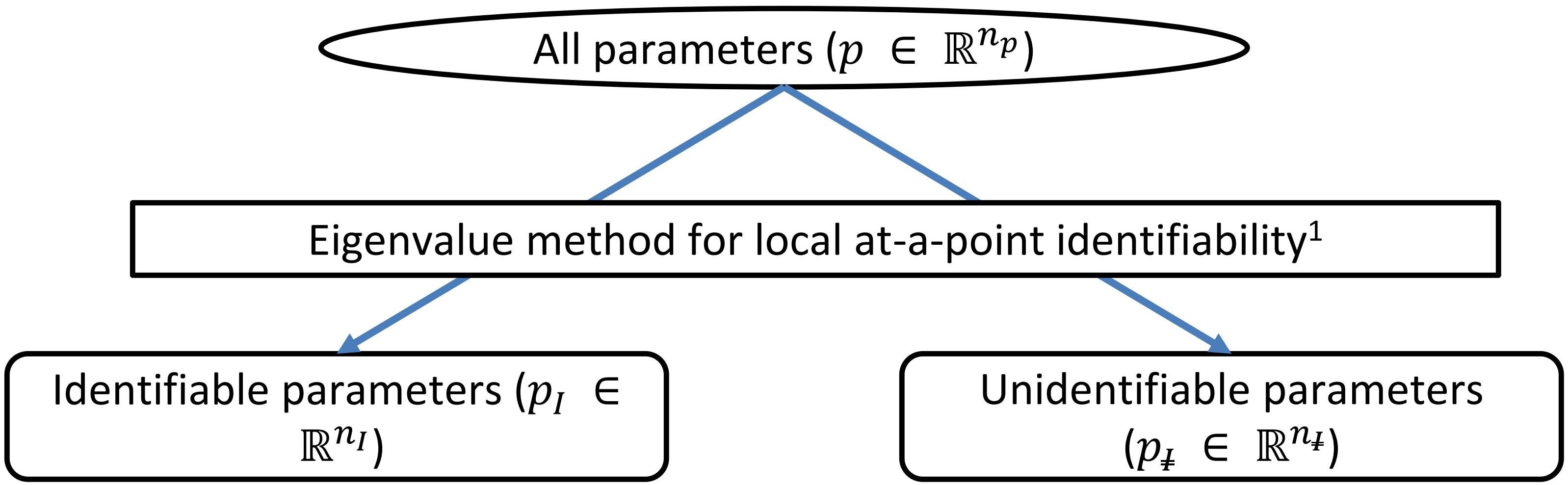
Challenge in polymerization systems: Is it possible to estimate all the parameters with available measurable data?

Identifiability:

- Can parameters be uniquely determined from available measurements?

Identifiability Analysis- Definition and Method

- Identifiability definition: A parameter is said to be identifiable if it can be uniquely determined from available measurement and input data.
- Several methods exist to check parameter identifiability. However, only local, at-a-point methods are tractable for large scale systems.



[1]Quaiser, T., Data- and model-based identification of biochemical processes, *PhD Thesis*, Ruhr-Universität Bochum, 2011

Identifiability Analysis in Case Study

Identifiability Analysis with measurements

- Reactor temperature
- Coolant inlet temperature
- Coolant outlet temperature
- Enthalpy of reaction

\+

- Monomer conversions

Type	Parameter
Mass transfer	$K_{MW_S}, K_{MW_B}, K_{MW_{AA}}, K_{MW_{AAm}}, K_{WP_S}, K_{WP_B}, K_{WP_{AA}}, K_{WP_{AAm}}, \boldsymbol{V_{kd_1}}, \boldsymbol{V_{kd_2}}, \boldsymbol{V_{kd_3}}, \boldsymbol{V_{kd_4}}, \boldsymbol{V_{kp_1}}, \boldsymbol{V_{kp_2}}, \boldsymbol{k_{abs}}, \boldsymbol{k_{des}}$
Reaction Kinetics	$k_{I0}, k_{intra0}, E_{intra}, k_{p1ter0}, E_{p1ter}, \boldsymbol{k_{p2ter0}}, E_{p2ter}, k_{p3ter0}, E_{p3ter}, k_{p4ter0}, \boldsymbol{E_{p4ter}}, a_S, a_B, \boldsymbol{k_{p0_3}}, \boldsymbol{k_{p0_4}}, \boldsymbol{r_{12}}, r_{13}, r_{14}, \boldsymbol{r_{21}}, r_{23}, r_{24}, r_{31}, r_{32}, \boldsymbol{r_{34}}, r_{41}, r_{42}, \boldsymbol{r_{43}}, k_{t0_3}, k_{t0_4}, k_{f0_1}, k_{f0_2}, k_{f0_3}, E_{f_3}, k_{f0_4}, E_{f_4}$
Heat transfer	$\boldsymbol{kA}, \boldsymbol{kA_{v1}}, \boldsymbol{kA_{v2}}$

Number of identifiable parameters is 19 out of 54

Parameter Estimation – Solution to Suboptimal Estimates

Parameter estimation is typically multimodal optimization problem.

Reasons for model-experiment mismatch:

- Wrong experimental data.
- Wrong model structure.
- Suboptimal estimation. → Can be solved through deterministic global optimization.

Case study:

- batch reaction
- 6 states
- 3 unknown parameters: (k_{2f}, k_{3f}, k_4)

xI (Measurements):

$$x_I = x_A + \frac{2}{21} x_B + \frac{2}{21} x_D$$

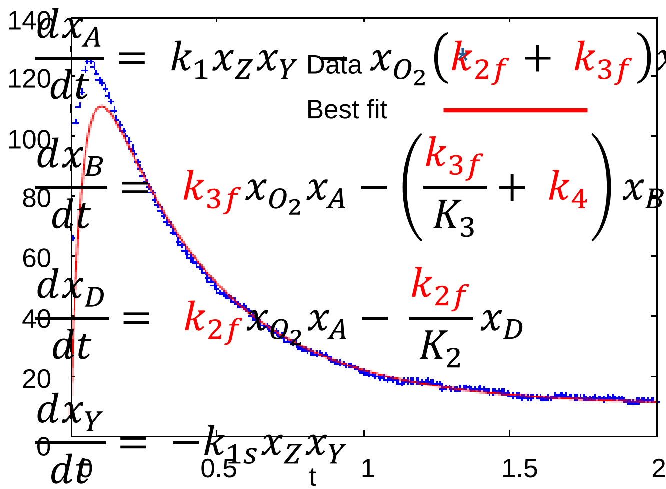
$$\frac{dx_A}{dt} = k_1 x_Z x_Y - x_{O_2}(k_{2f} + k_{3f}) x_A + \frac{k_{2f}}{K_2} x_D + \frac{k_{3f}}{K_3} x_B - k_5 X^2{}_A$$

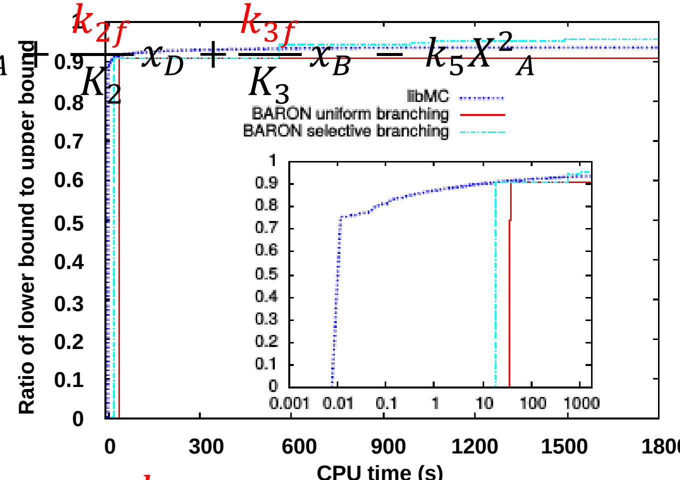
$$\frac{dx_B}{dt} = k_{3f} x_{O_2} x_A - \left(\frac{k_{3f}}{K_3} + k_4\right) x_B$$

$$\frac{dx_D}{dt} = k_{2f} x_{O_2} x_A - \frac{k_{2f}}{K_2} x_D$$

$$\frac{dx_Y}{dt} = -k_{1s} x_Z x_Y$$

$$\frac{dx_Z}{dt} = -k_1 x_Z x_Y$$





unknown parameters

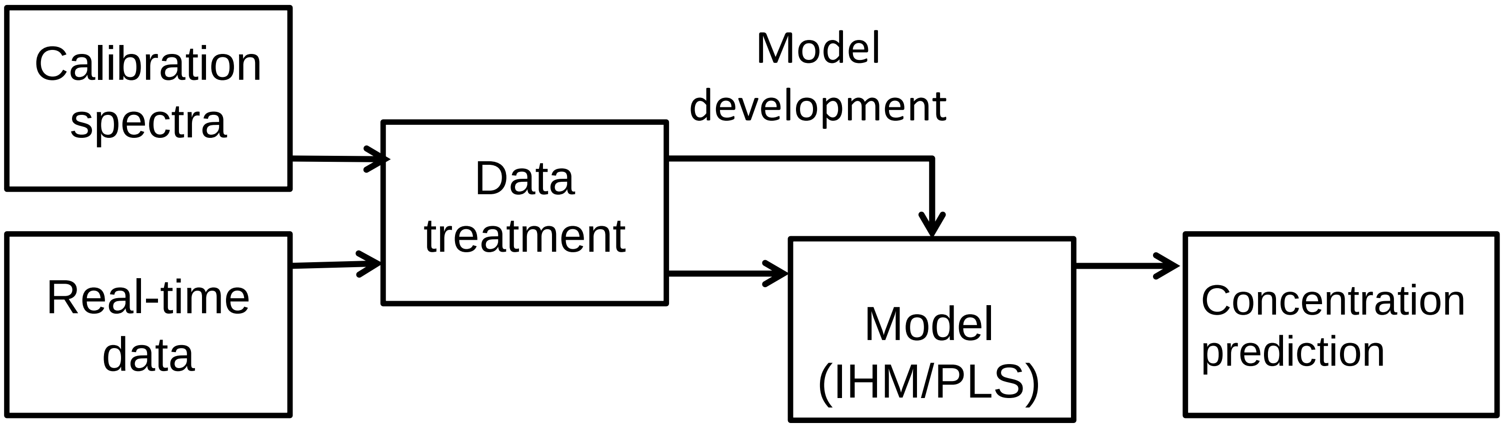
Mitsos et al., SIOPT, 2009

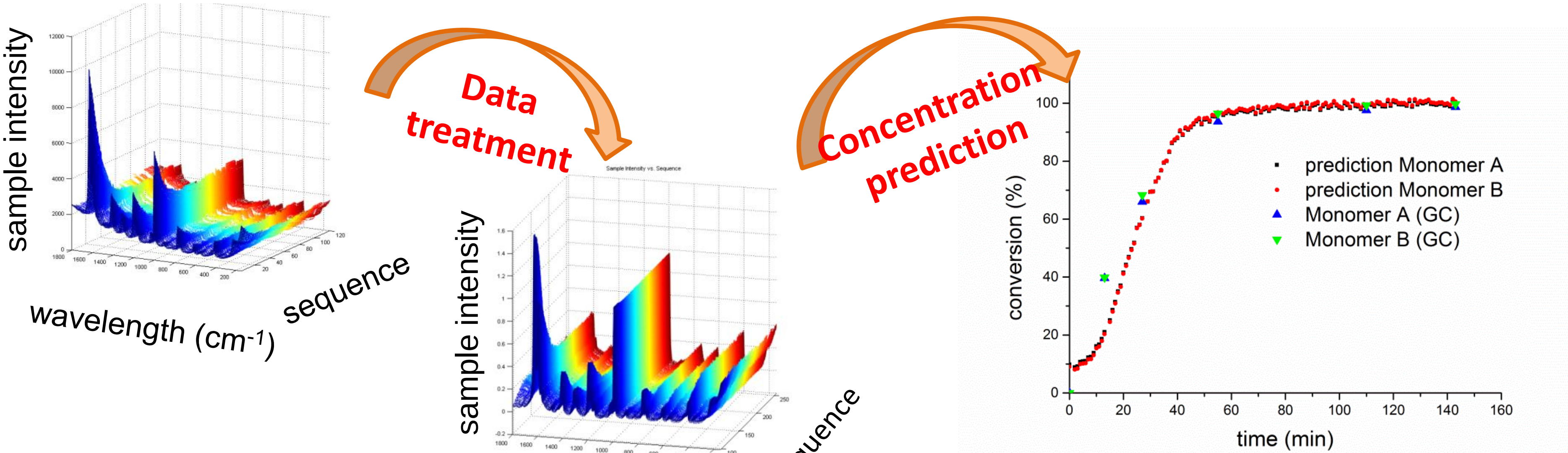
Taylor et al. J. Phys. Chem. A, 2004

Standard optimization methods not promising, specialized ones tractable.

Challenge holds also for other optimization problems, e.g. offline and online optimization.

Raman Spectra → Conversion Prediction (Batch process)

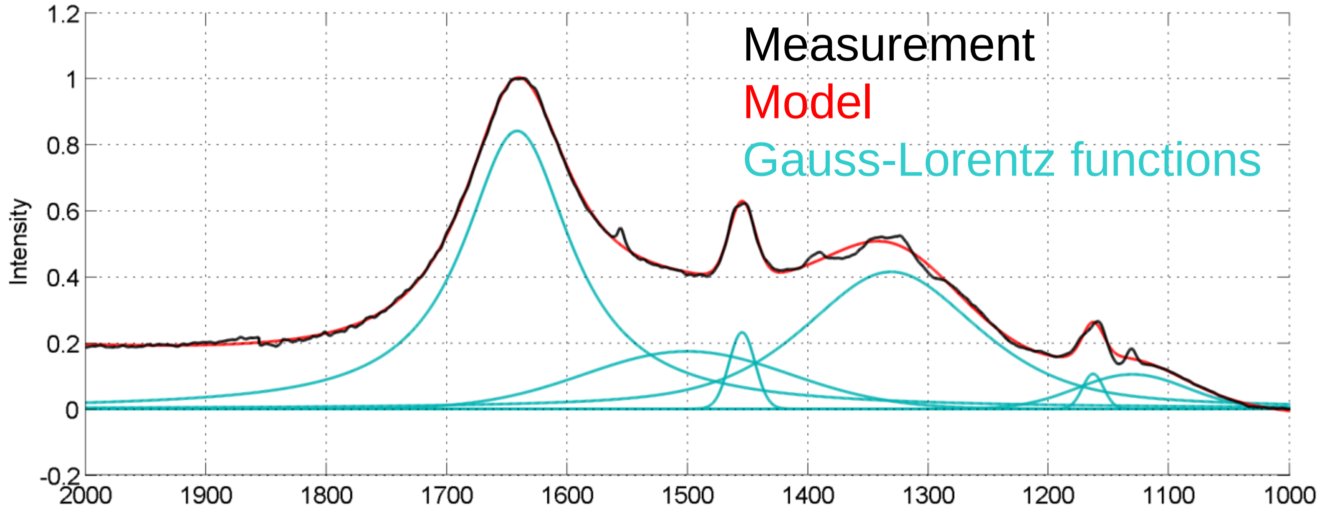




Results from University of Cambridge

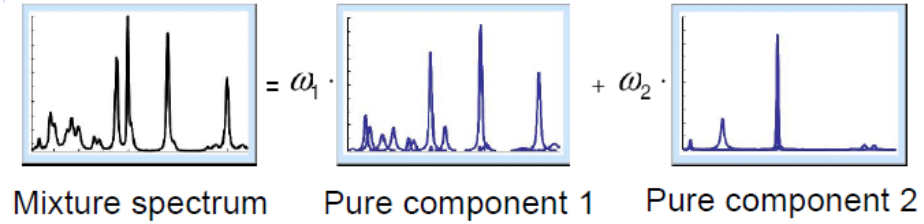
Indirect Hard Modeling[1] (IHM)

Model Raman Spectra of reaction components using Gauss – Lorentz functions.



A model of the mixture (A) is created as weighted sum of the pure component models (A'_k)

$$A(\tilde{v}, x) = \sum_{k=1}^{K} \omega_k(x) A'_k\,(\tilde{v})$$

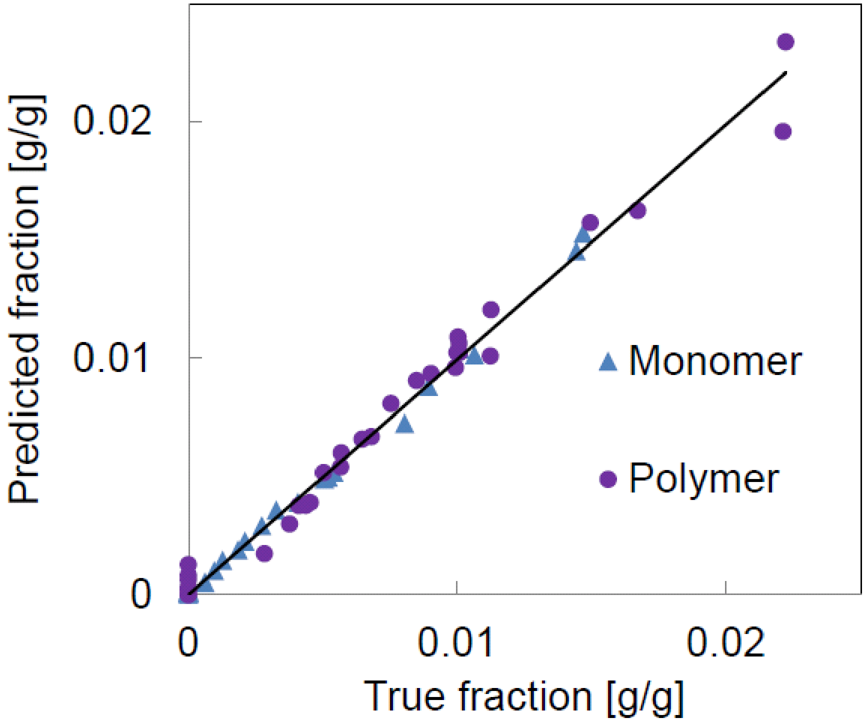


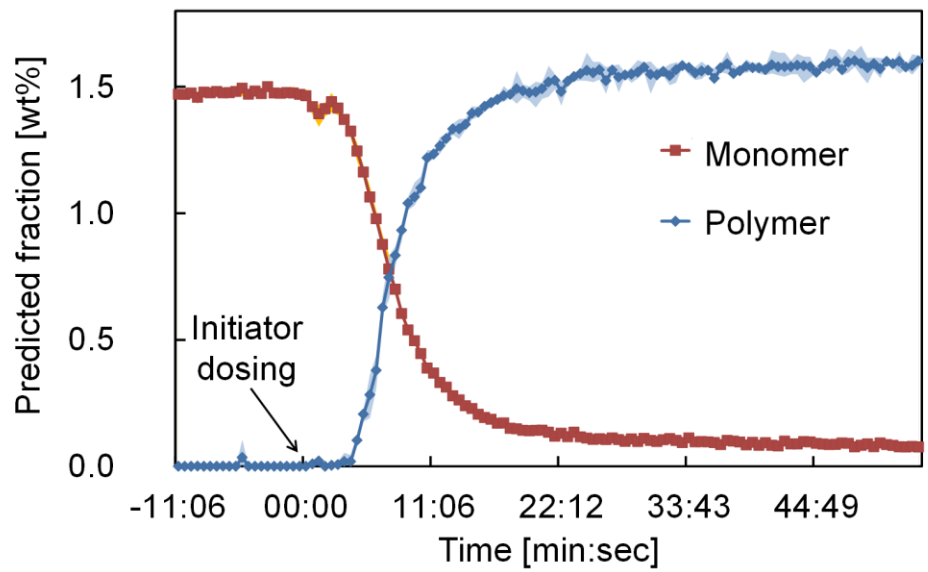
[1]Alsmeyer, F., Koß, H.-J., Marquardt, W., *Journal of Applied Spectroscopy*, 58,975-985,2004.

Concentration Estimation from Raman Spectra (Batch Process)

Models calibrated using multivariate regression in PEAXACT[1] software.

Calibrated models are then used for concentration estimation





Monomer RMSECV=0.00028 R^2=0.9967

Polymer RMSECV=0.00084 R^2=0.9835

[1]PEAXACT, S-PACT GmbH

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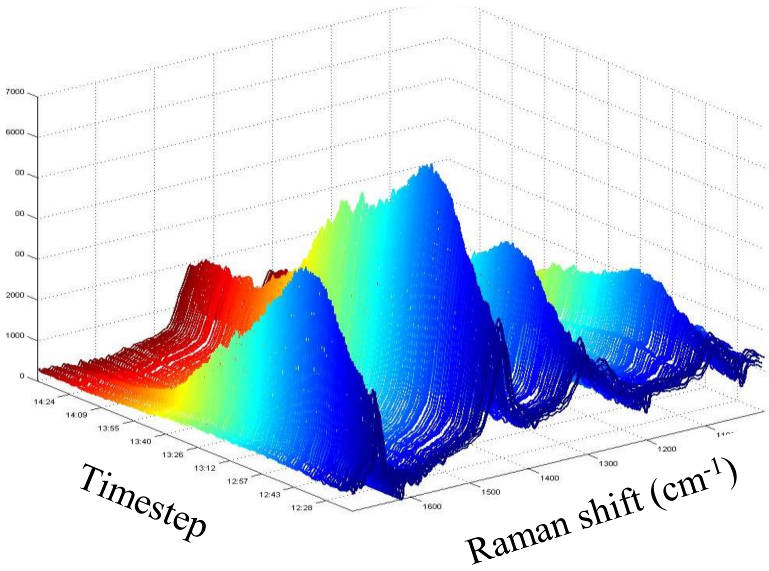
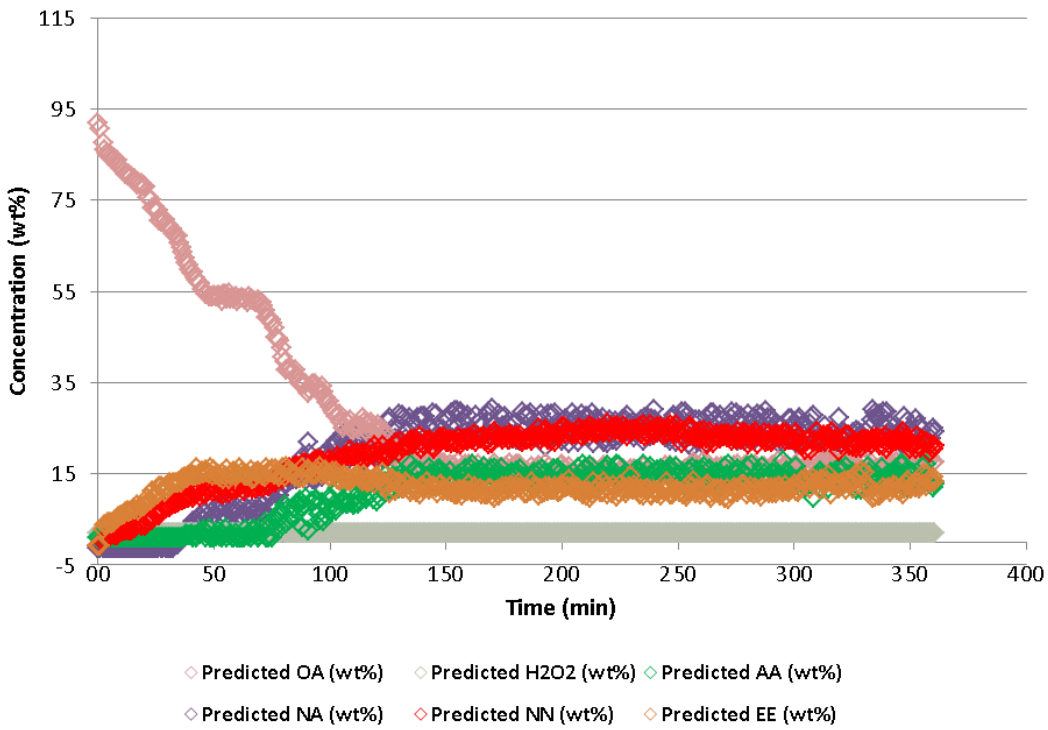
Monitoring Oxidation of Oleic Acid using Raman Spectra (Batch Process)





EU Project - OPTICO

Raman Spectra → Particle Size Prediction (Batch Process)

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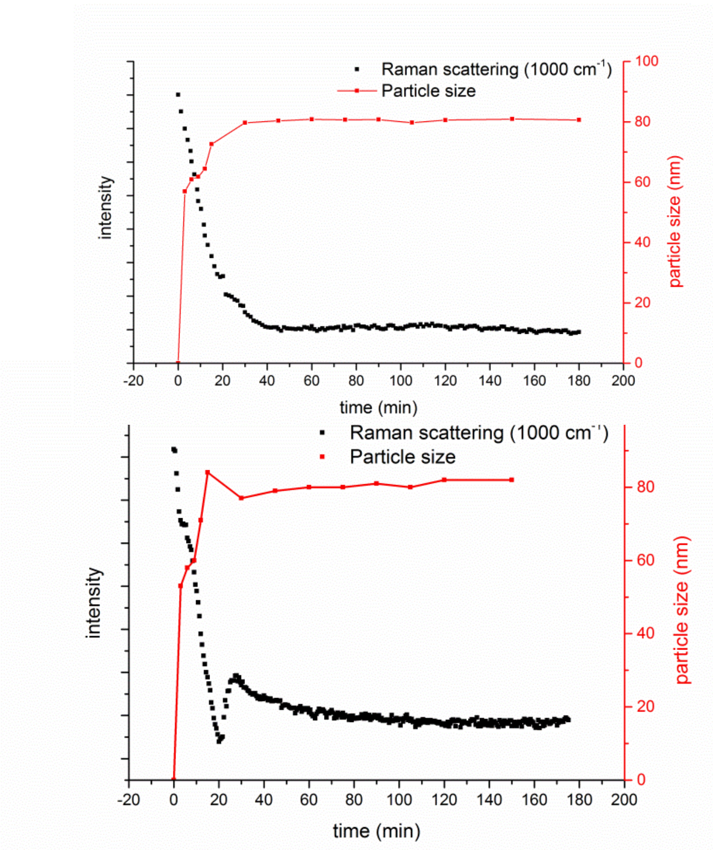
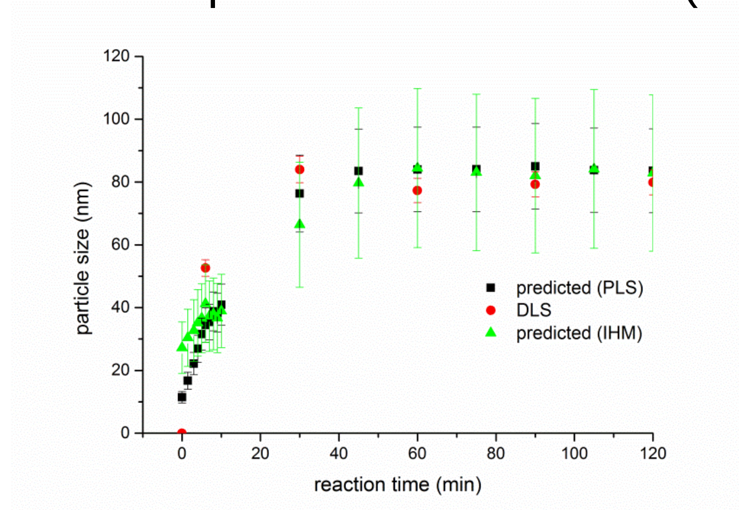


Comparison of particle size (red) and intensity profile (black) during experiments.

- Model for prediction of particle size is trained on the intensity change at 1000 cm^{-1}.
- Model shows good agreement to offline particle measurement (DLS).



Comparison particle size of PLS, IHM and offline measured with DLS.

Results from University of Cambridge

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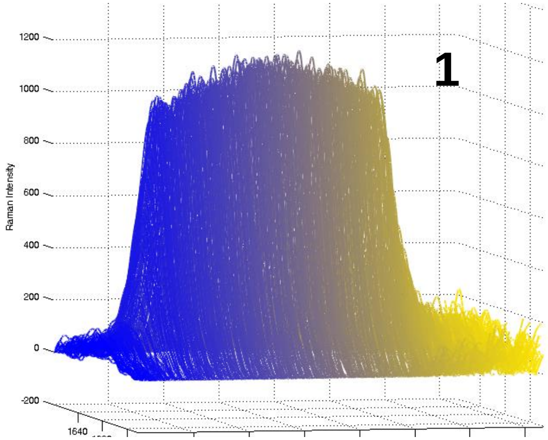
Raman Spectra → Conversion and Particle Size (Semi-batch Process)

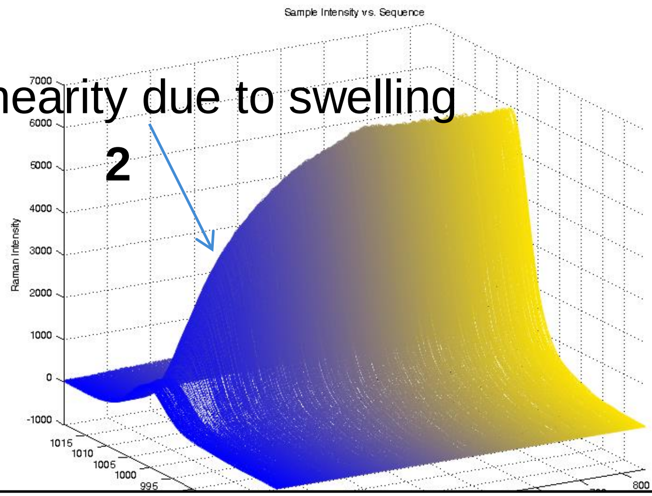


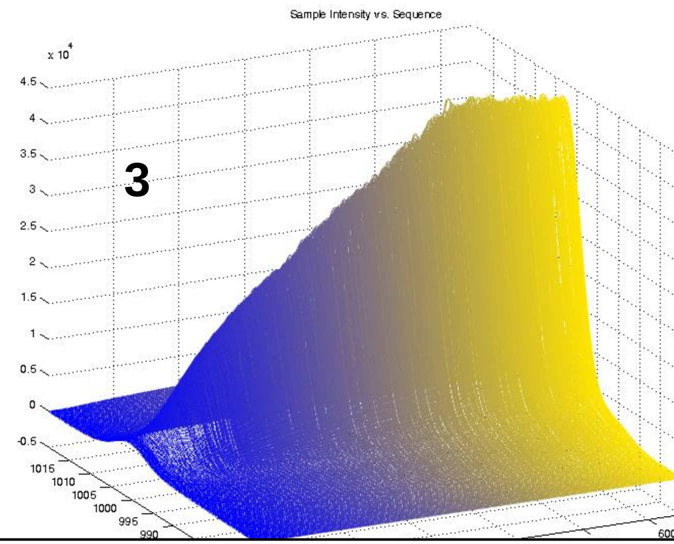
1 Raman intensity for the double bond of styrene under starved conditions

2 Raman intensity for the aromatic ring of styrene under starved conditions

3 Raman intensity for the aromatic ring of styrene without reaction.

Nonlinearity due to swelling

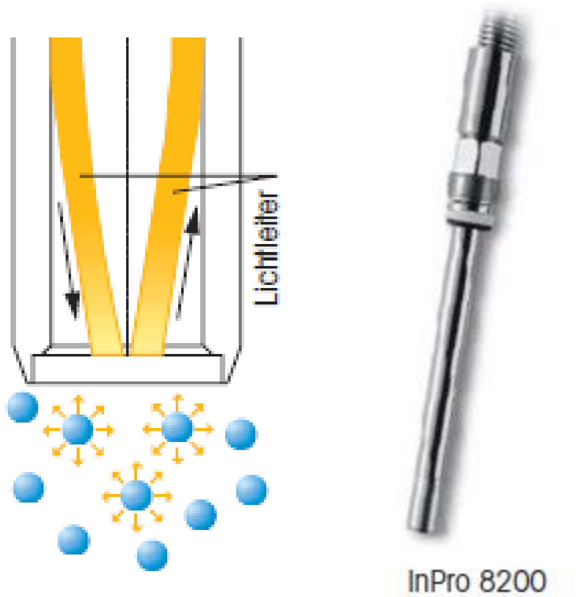




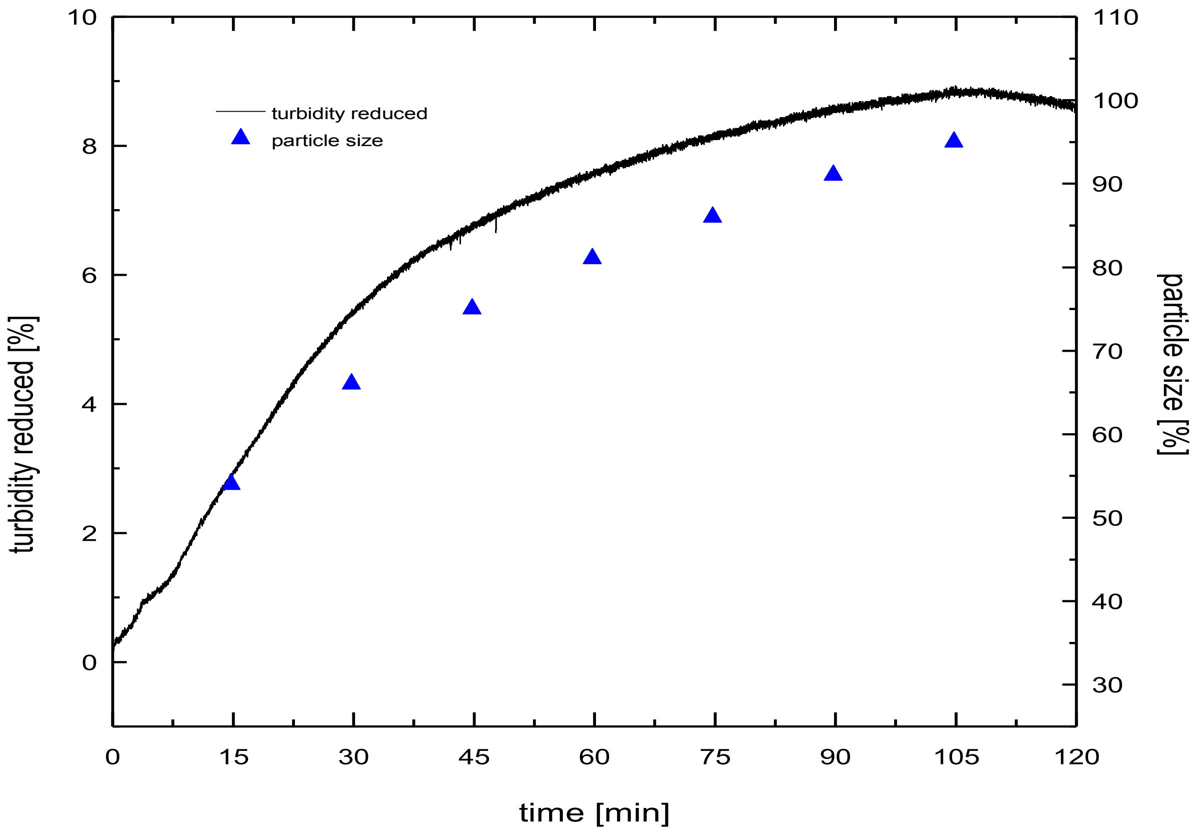
Estimation of concentration and particle size challenging

Results from University of Hamburg

Turbidity → Particle Size Prediction (Semi-batch Process)



Turbidity sensors by Mettler Toledo

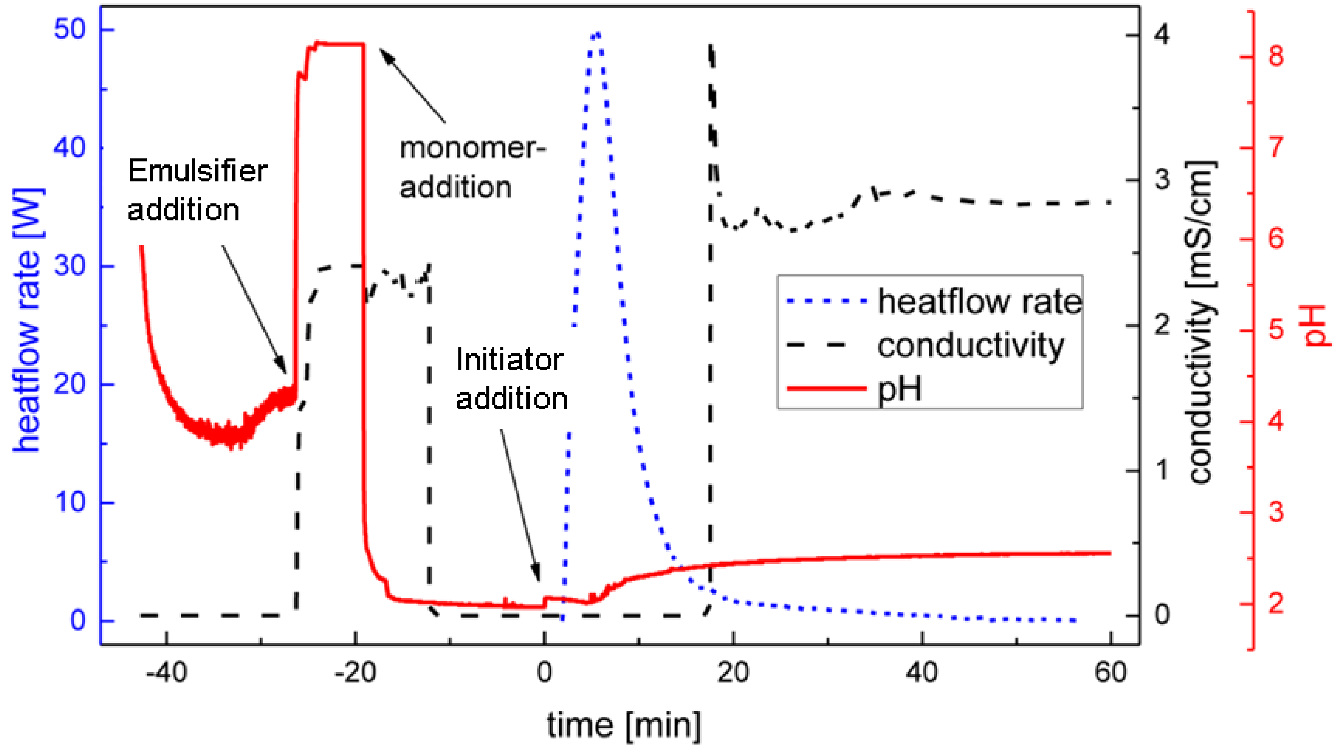


- Turbidity measurements in semi-batch reaction without effects of dosing.
- With sensor calibration turbidity leads directly to particle size.

Results from University of Hamburg

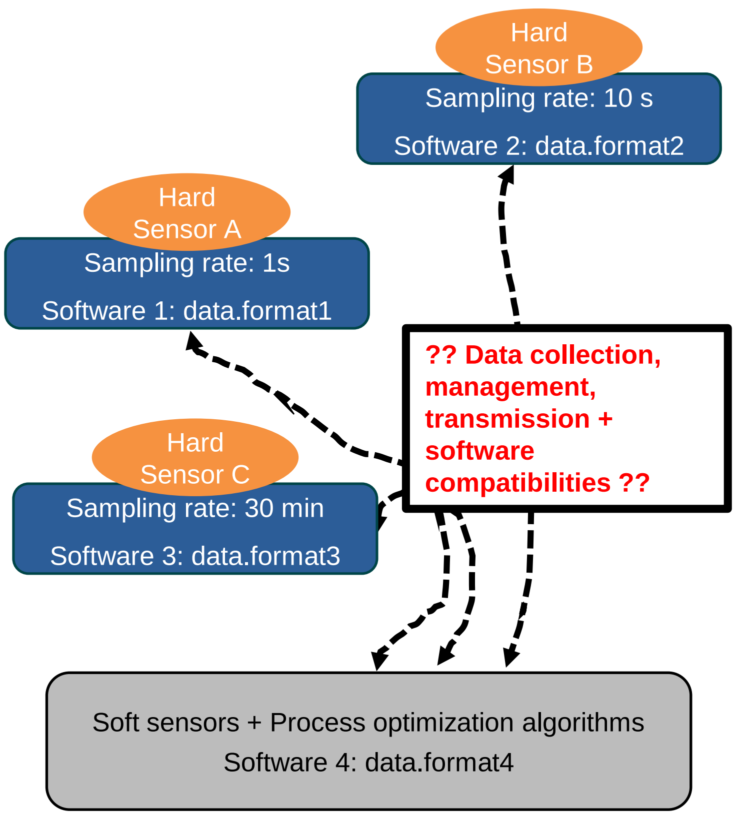
Other Sensors Investigated by Collaborators

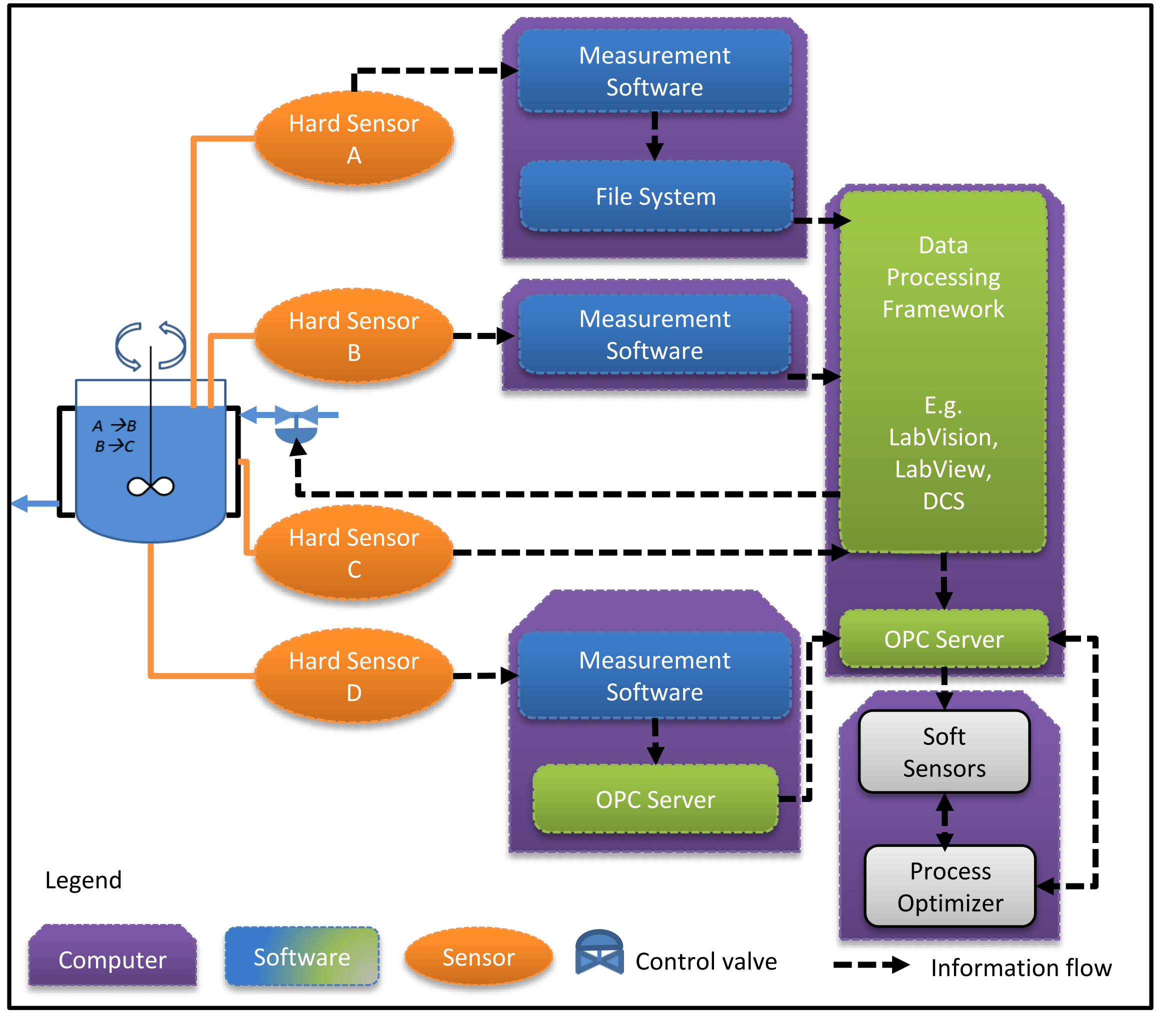
- Conductivity measurements
- pH measurements



Results from University of Hamburg

OPC-based Data processing framework





Summary

- Moments of polymer chain length are unobservable without online measurements of polymer property.
- Linear observability analysis shows that online concentration measurements are required for observability of energy and material balance equations.
- More parameters are identifiable when concentration measurements are used along with calorimetric measurements.
- Problem of suboptimal estimation can be solved by using deterministic global optimization techniques.
- Raman spectra can be used to predict online monomer conversions and particle size.

Future Directions

- Modeling – developing models that correlate measurements (conductivity, pH, turbidity) to process states (particle sizes, polymer property).
- Sensor development – sensor for online monitoring of polymer properties.
- Nonlinear techniques for testing observability and identifiability.

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Thank you for your attention!

Questions?