Simultaneous Estimation of the Heat Transfer Coefficient and of the Heat of Reaction in Semi-Batch Processes

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Outline

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- Modelling of the Process: CSTR and PFR Jacket Models
- System Analysis
- State Estimation by Extended Kalman Filter
  - Tuning of the Estimator
  - Influence of the Model of the Jacket
- Oscillation Calorimetry
  - Traditional data analysis
  - Advanced data analysis by an extended model + EKF
- Summary

Motivation

- Online monitoring of conversion (heat of reaction) in (semi-)batch processes is very important in process monitoring and control and can be performed by calorimetry.
- The heat transfer coefficient is not known a priori or varies over the batch run.
- In heat flux calorimetry, there are often errors in the estimates of the heat of reaction due to a wrong heat transfer coefficient.
- Estimation of heat of reaction **and** heat transfer coefficient is needed.
- Possible approaches:
  - State and parameter estimation by nonlinear filtering
  - Oscillation calorimetry

Motivation

- In practice the following difficulties occur:
  - The jacket of technical reactors normally behaves like a Plug Flow Reactor (PFR), but is modelled as a CSTR,
  - The heat transfer coefficient (k) changes with the batch time, especially in semi-batch processes,
  - The flow rate through the jacket influences the dynamic behaviour of the system significantly.
- These aspects have to be considered in the design of the estimator.

Modelling: Jacket as CSTR

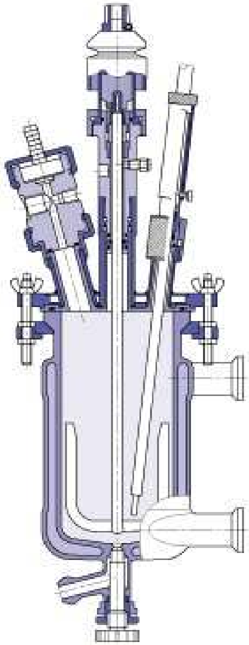
- Reaction calorimetry and state estimation approaches are based on the same physical model.
  - Balances for the reactor and the jacket (CSTR)

$$\text{Vessel}: \frac{dh_R}{dt} = \frac{1}{A_B}\left(\dot{V}_{R,in} - \dot{V}_{R,out}\right)$$

$$\frac{dT_R}{dt} = \frac{\dot{V}_{R,in}}{A_B}\left(T_{R,in} - T_{R,out}\right) + \frac{1}{\rho_R c_{p,R} A_B h_R}\left(\dot{Q}_{R,source} - \dot{Q}_{R,loss}\right) - \dots$$

$$\frac{kA}{\rho_R c_{p,R} A_B h_R}\left(T_R - T_J\right)$$

$$\text{Jacket}: \frac{dT_J}{dt} = \frac{1}{m_J c_{p,J}}\left(\dot{m}_J c_{p,J}\left(T_{J,in} - T_{J,out}\right) + kA\left(T_R - T_J\right)\right)$$



Modelling: Jacket as PFR

- Balances for the reactor and the jacket (PFR)

Vessel : $$\frac{dh_R}{dt} = \frac{1}{A_B}\left(\dot{V}_{R,in} - \dot{V}_{R,out}\right)$$

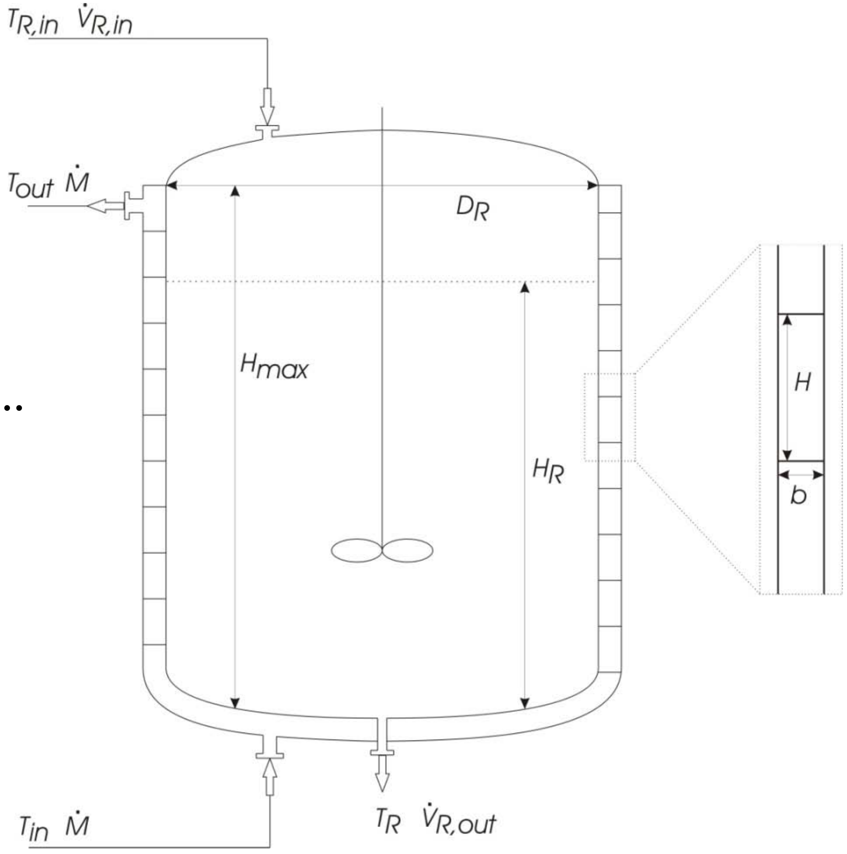
$$\frac{dT_R}{dt} = \frac{\dot{V}_{R,in}}{A_B}\left(T_{R,in} - T_{R,out}\right) + \ldots$$

$$\frac{1}{\rho_R c_{p,R} A_B h_R}\left(\dot{Q}_{R,source} - \dot{Q}_{R,loss}\right) - \ldots$$

$$\frac{kA}{\rho_R c_{p,R} A_B h_R}\left(T_R - T_J\right)$$

Jacket : $$\frac{\partial T_J}{\partial t} = -v\frac{\partial T_J}{\partial z} + \frac{k}{\rho_J c_{p,J} b}\left(T_R - T_J(t,z)\right)$$

$$T_J(t,0) = T_{J,in}$$



CSTR-Model: Pseudo States

- Model for state and parameter estimation:
  - We assume that the level in the reactor h_R is known.
  - The model is extended by pseudo dynamics for the unknown parameters.

$$\frac{dT_R}{dt} = \frac{\dot{V}_{R,in}}{A_B}\left(T_{R,in} - T_{R,out}\right) + \frac{1}{\rho_R c_{p,R} A_B h_R}\dot{Q}_R - \frac{kA}{\rho_R c_{p,R} A_B h_R}\left(T_R - T_J\right)$$

$$\frac{dT_J}{dt} = \frac{1}{m_J c_{p,J}}\left(\dot{m}_J c_{p,J}\left(T_{J,in} - T_{J,out}\right) + kA\left(T_R - T_J\right)\right)$$

$$\frac{d\dot{Q}_R}{dt} = 0$$

$$\frac{dk}{dt} = 0$$

CSTR-Model: Observability Analysis

- Observability of the CSTR-model:
  - The nonlinear observability map is given by:

$$q_O = \begin{bmatrix} y_1 \\ y_2 \\ \dot{y}_1 \\ \dot{y}_2 \end{bmatrix} = \begin{bmatrix} T_R \\ T_J \\ \frac{\dot{V}_{R,in}}{A_B}\left(T_{R,in} - T_{R,out}\right) + \frac{1}{\rho_R c_{p,R} A_B h_R}\left(\dot{Q}_{R,source} - \dot{Q}_{R,loss}\right) - \frac{kA}{\rho_R c_{p,R} A_B h_R}\left(T_R - T_J\right) \\ \frac{1}{m_J c_{p,J}}\left(\dot{m}_J c_{p,J}\left(T_{J,in} - T_{J,out}\right) + kA\left(T_R - T_J\right)\right) \end{bmatrix}$$

  - The system is globally observable, if $q_O^{-1}(x,u)$ can be solved uniquely in terms of the state vector x for $x \in X$, $u \in U$.

Observability Analysis

- The inversion of the nonlinear observability map yields:

$$T_R = y_1$$
$$T_J = y_2$$
$$\dot{Q}_R = \rho_R c_{p,R} V_R \left(\dot{y}_1 - \frac{\dot{V}_{R,in}}{A_B} \left(T_{R,in} - T_{R,out}\right) \right) + \left(m_J c_{p,J} \dot{y}_2 - \dot{m}_J c_{p,J} \left(T_{J,in} - T_{J,out}\right)\right)$$
$$k = \frac{1}{A\left(T_R - T_J\right)} \left(m_J c_{p,J} \dot{y}_2 - \dot{m}_J c_{p,J} \left(T_{J,in} - T_{J,out}\right)\right)$$

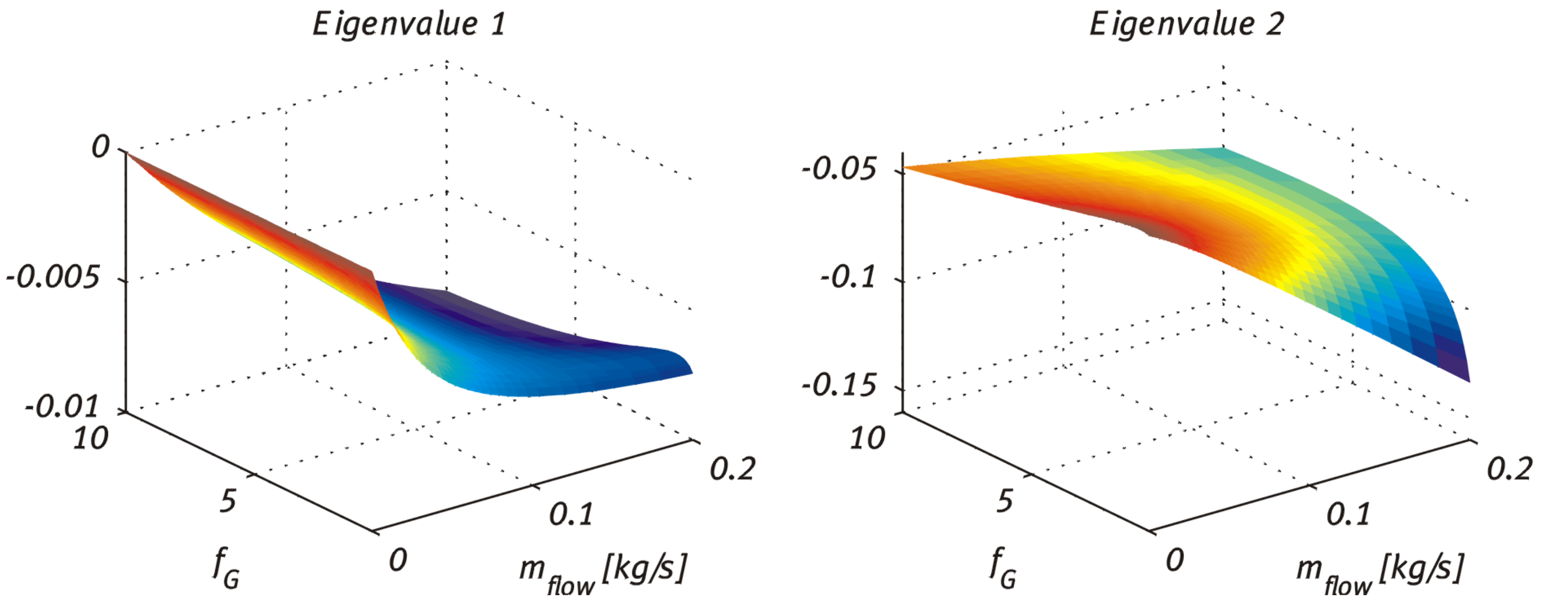
$\Rightarrow$ The given system is globally observable for all

$T_R \neq T_J$ and $A \neq 0 \Leftrightarrow V_R \neq 0$

- It can be shown that this result holds also for the PFR-model if it is discretized by orthogonal collocation with $T_J = T_{J,out}$.

System Analysis

- Dynamic behaviour of the system
  - eigenvalues of the linearised system



- f_G = scaling factor, m_{flow} = normalised jacket flow rate
- Eigenvalues depend strongly on the jacket mass flow!

State Estimation: Extended Kalman Filter

- **Algorithm:**
  - Notation:
    $\hat{x}_{k+1,k}$ estimated state at $t=t_{k+1}$ based on measurements up to $t=t_k$
  - Correction:

$$K_k = P_{k,k-1} H_{k,k-1}^T \left(H_{k,k-1} P_{k,k-1} H_{k,k-1}^T + R \right)^{-1}$$

$$\hat{x}_{k,k} = \hat{x}_{k,k-1} + K_k \left(y_k - h\left(\hat{x}_{k,k-1} \right) \right)$$

$$P_{k,k} = \left(I - K_k H_{k,k-1} \right) P_{k,k-1}$$

  - Prediction:

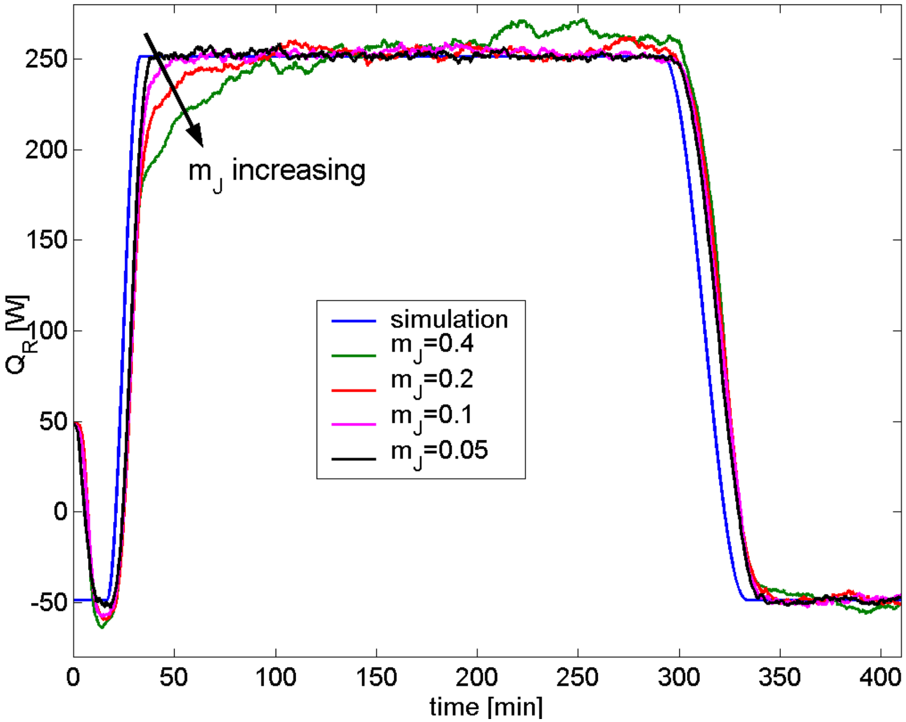
$$\hat{x}_{k,k+1} = F\left(\hat{x}_{k,k}, u_k \right)$$

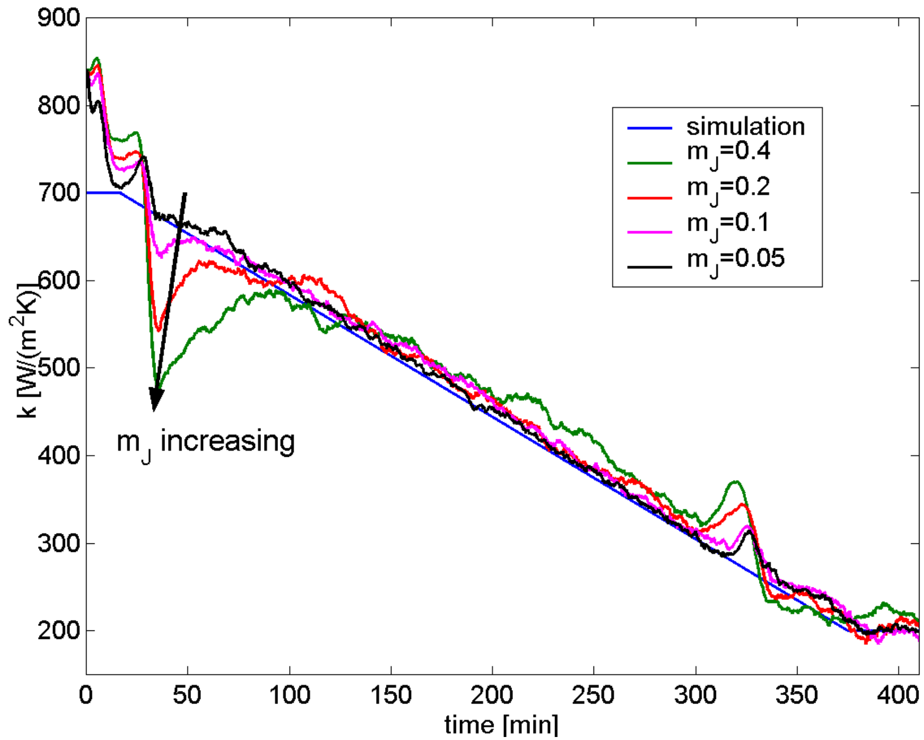
$$P_{k,k+1} = A_{k,k} P_{k,k} A_{k,k}^T + Q \quad \text{with} \quad A_{k,k} = \left. \frac{\partial f}{\partial x} \right|_{\hat{x}_{k,k}} \quad \text{and} \quad H_{k,k-1} = \left. \frac{\partial h}{\partial x} \right|_{\hat{x}_{k,k-1}}$$

  - Only (at best) local stability!

Tuning of the EKF

- Simulation of a reactor ($V=10\ l$) with a PFR-jacket
- Estimator based on CSTR-model for the jacket



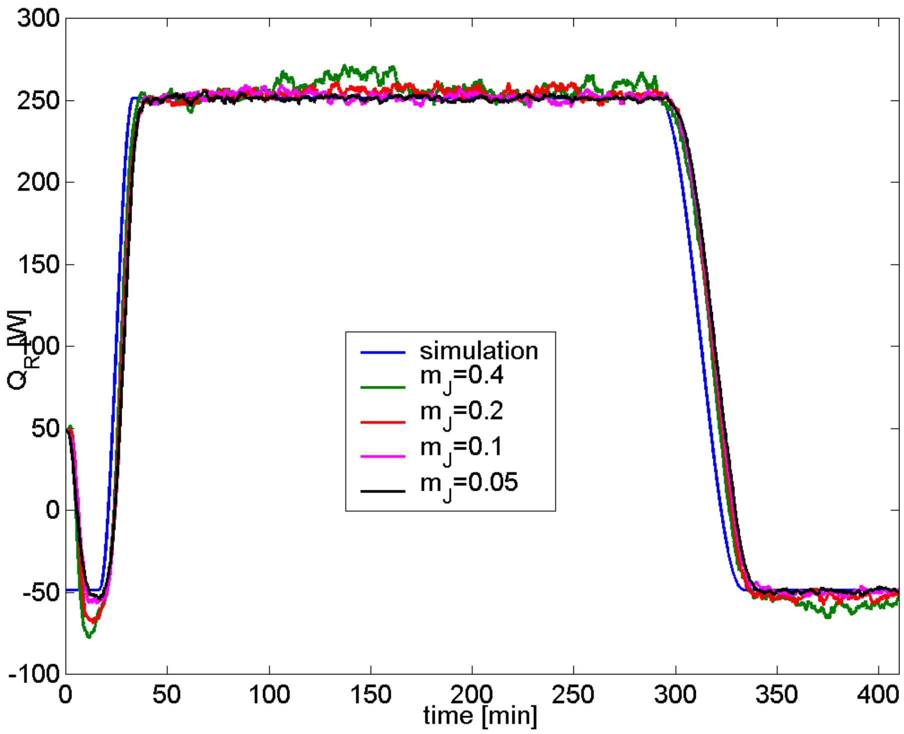


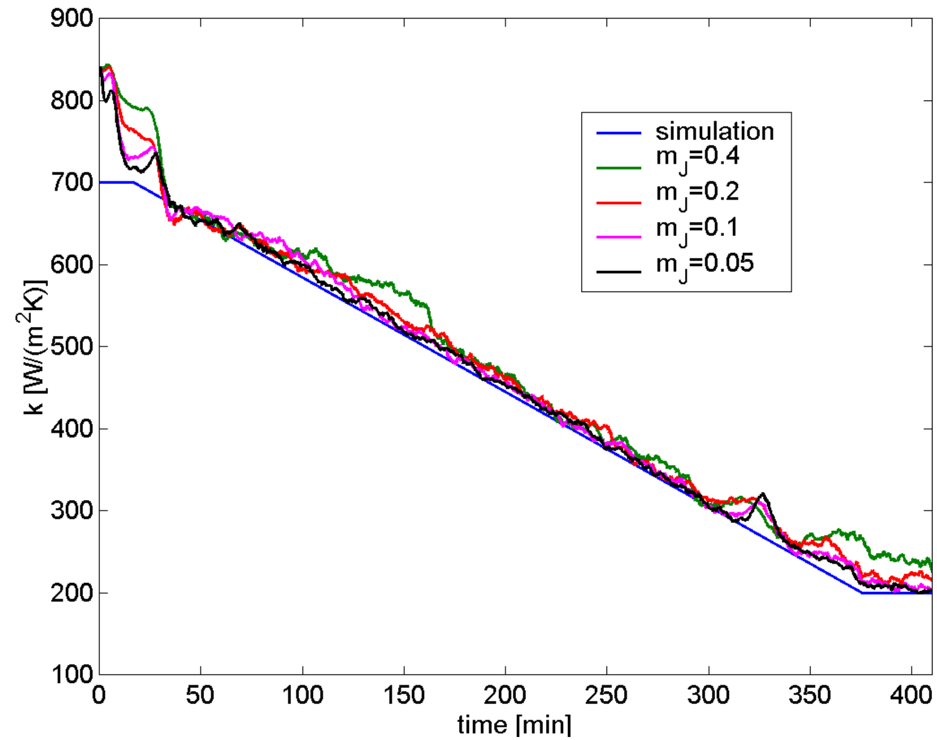
- For a constant covariance matrix Q:
  Performance depends on the jacket mass flow rate.

Tuning of the EKF

- Adaptation of the covariance matrix of the model error Q to the mass flow rate through the jacket:

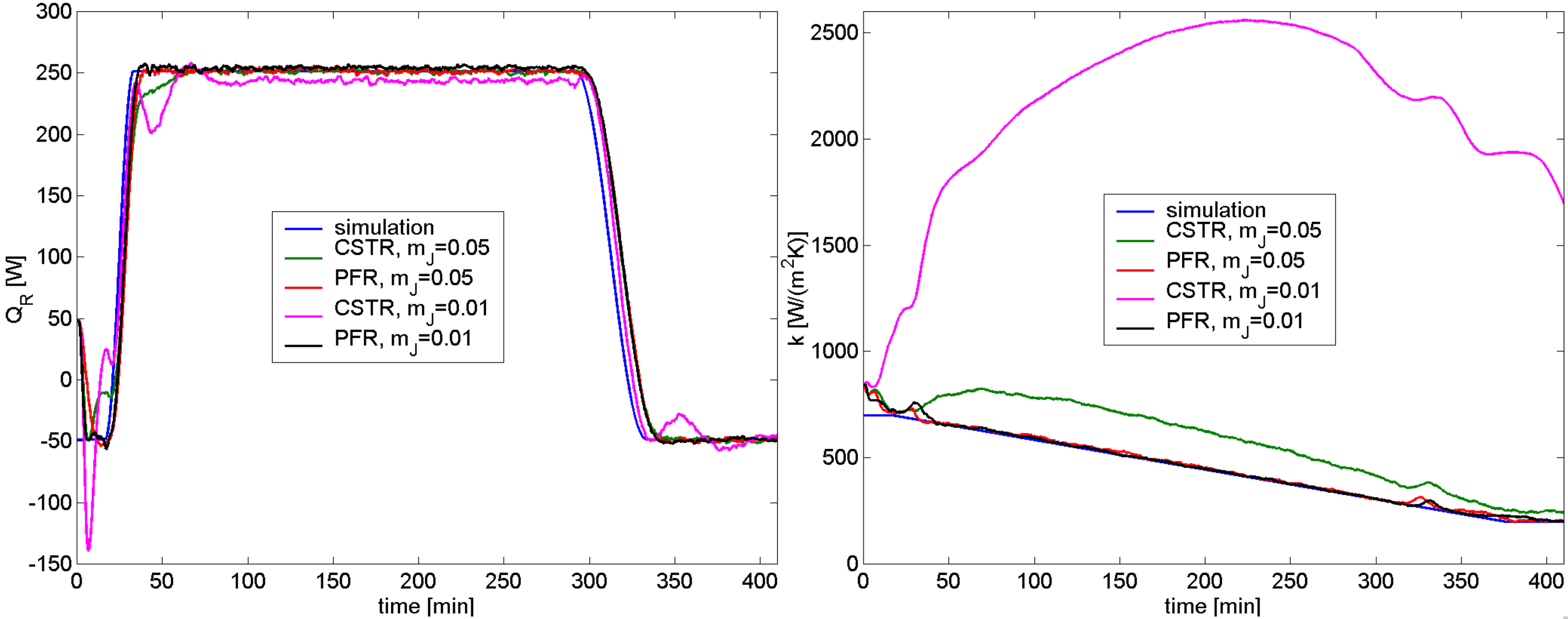
$$Q_{3,3} \sim \dot{m}_J$$





Influence of the Model of the Jacket

- Simulation of a system with a PFR-jacket
- EKF estimation for with CSTR or PFR-model



- Large reactors or low flow rate of the coolant:
  Results are poor if a CSTR model of the jacket is used.

Summary

- For simultaneous estimation of the heat of reaction and the heat transfer coefficient in a CSTR, it has been shown that:
  - the real behaviour of the jacket (CSTR or PFR) must be taken into account,
  - the estimator dynamics have to be adapted to the dynamic behaviour of the reactor,
  - adaptation of Q similar to the change of the eigenvalues by the jacket mass flow rate results in satisfactory estimations.

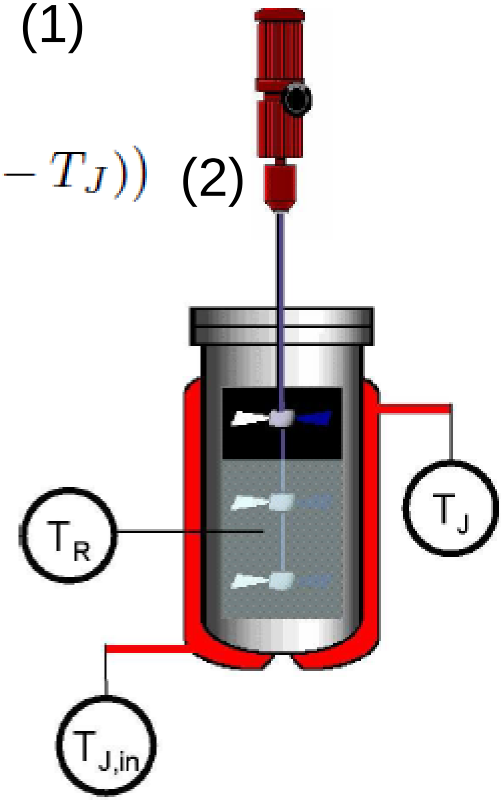
Recap: Calorimetry

Energy balances:

Reactor: $$\frac{dT_R}{dt} = \frac{1}{C_{p,R}}(\dot{Q}_R + kA(T_J - T_R) + \dot{Q}_{misc}) \quad (1)$$

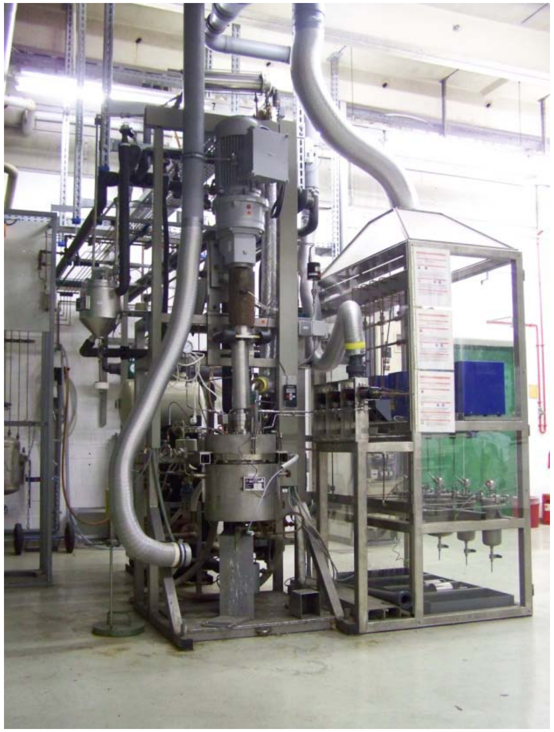
Jacket: $$\frac{dT_J}{dt} = \frac{1}{C_{p,J}}\left(-kA(T_J - T_R) + \dot{m}\, c_p\, (T_{J,in} - T_J)\right) \quad (2)$$

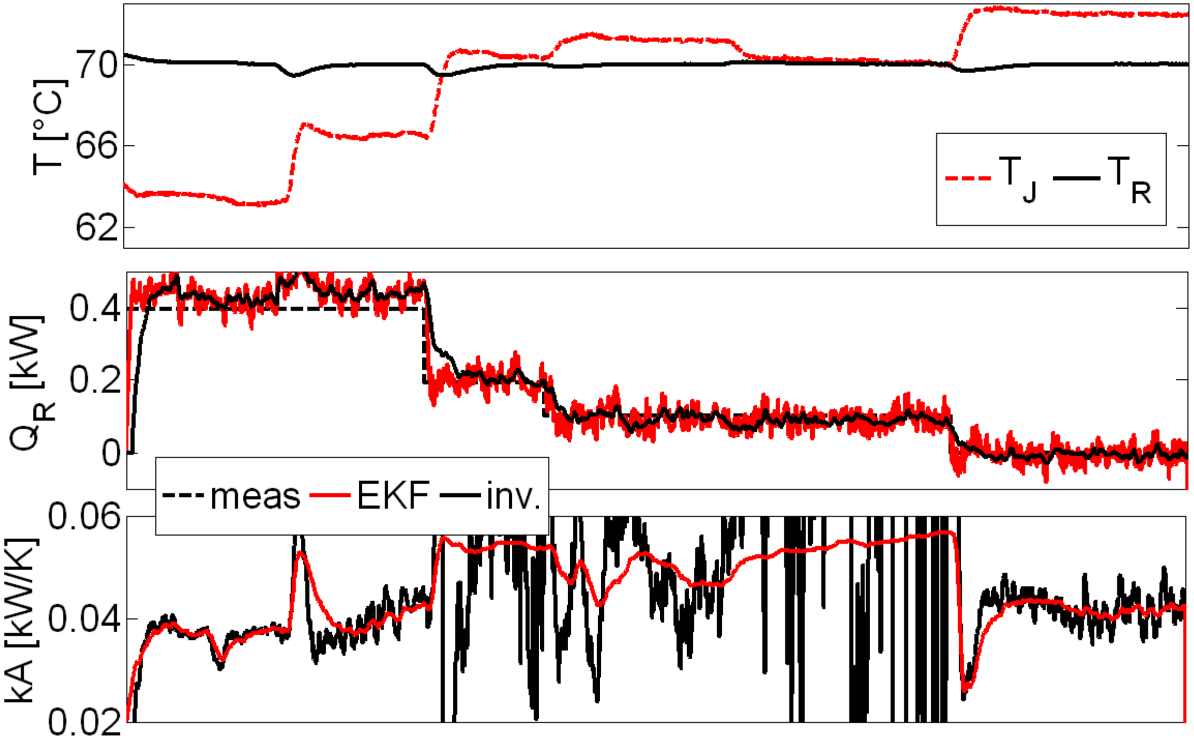
$$\frac{d\dot{Q}_R}{dt} = 0 \qquad \frac{d(kA)}{dt} = 0$$



- In case *kA* is known and constant, Eq. (1) is sufficient.
- Otherwise, Equation (2) has to be added.
- The model can be exploited by
  - Direct Inversion
  - Extended Kalman Filter

Calorimetry – Results at a Pilot Scale Reactor





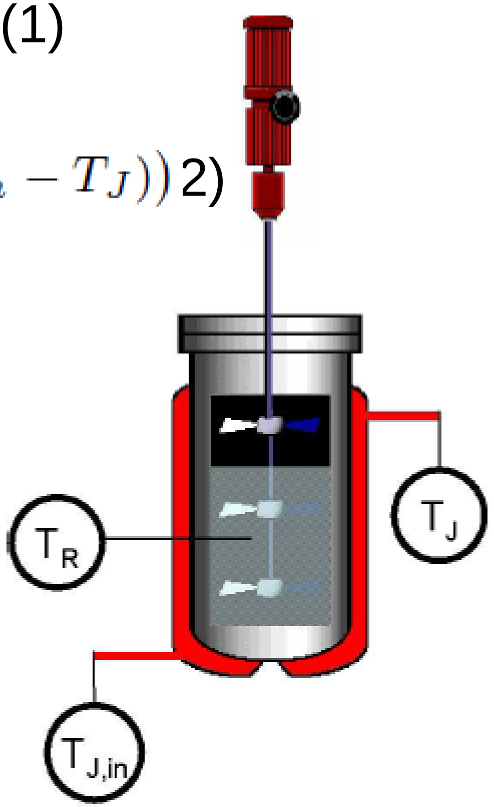
Heat Balance Calorimetry: Limitations

Energy balances:

Reactor: $$\frac{dT_R}{dt} = \frac{1}{C_{p,R}}(\dot{Q}_R + \mathbf{kA}(T_J - T_R) + \dot{Q}_{misc}) \quad (1)$$

Jacket: $$\frac{dT_J}{dt} = \frac{1}{C_{p,J}}(-\mathbf{kA}(T_J - T_R) + \dot{m}\, c_p\, (T_{J,in} - T_J)) \quad 2)$$

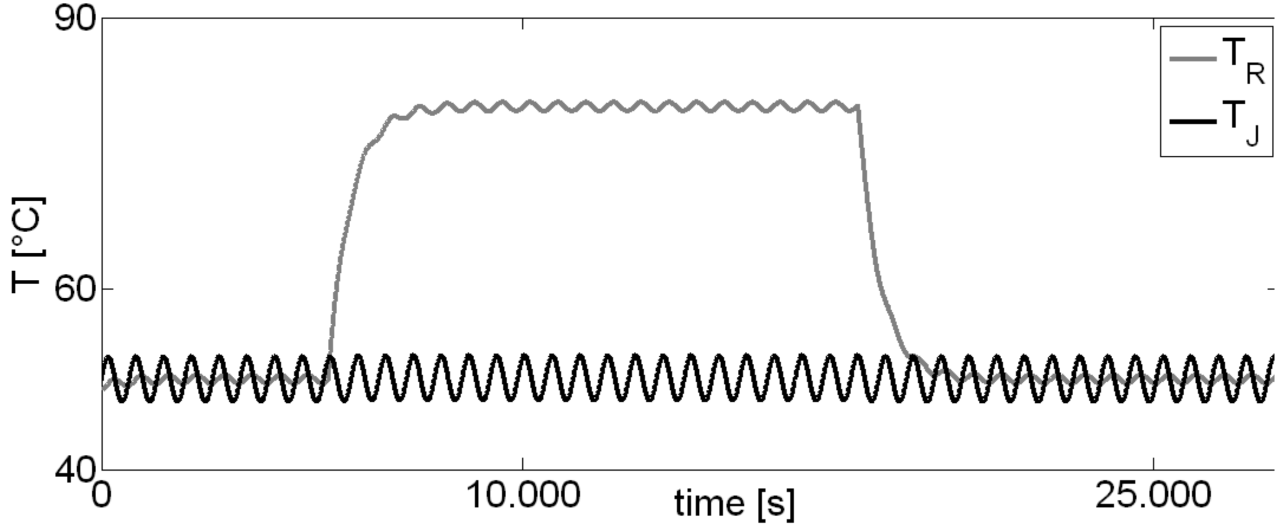
$$\frac{d\dot{Q}_R}{dt} = 0 \qquad \frac{d(kA)}{dt} = 0$$



- **Eq. (2) can only be exploited for sufficiently large | $T_{J,in} - T_J$ | !**

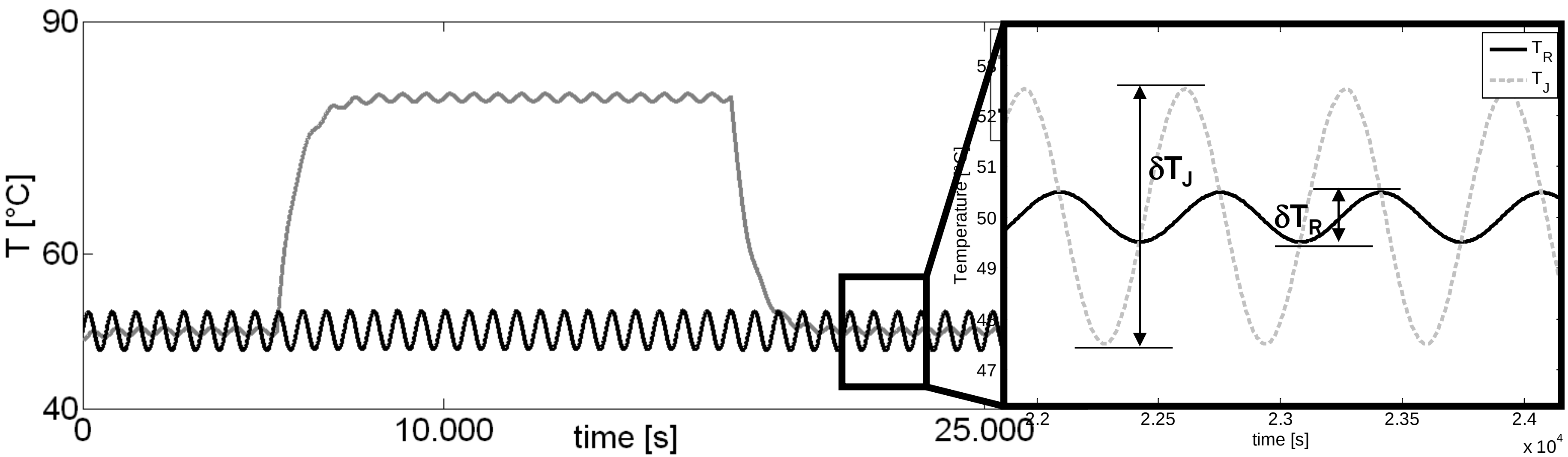
Temperature Oscillation Calorimetry (TOC)

- Many, especially small (laboratory) reactors are operated at large jacket flowrates → temperature difference in the jacket is too small..
- ***Idea:***
  Add a sinosoidal signal to the reactor temperature T_R



- **Compute kA from the frequency response between the two harmonic signals**

TOC: Formulae by Tietze (1996)



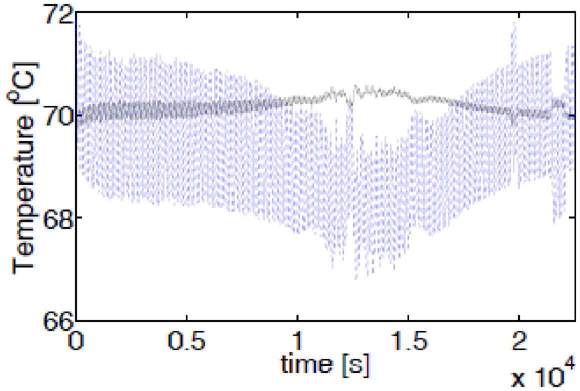
- *kA* computation: $$kA = \frac{C_p\ \omega}{\tan\left(\arccos\left[\frac{\delta T_R}{\delta T_J}\right]\right)}$$
- Q_R computation: From reactor heat balance

Drawbacks of the Method by Tietze

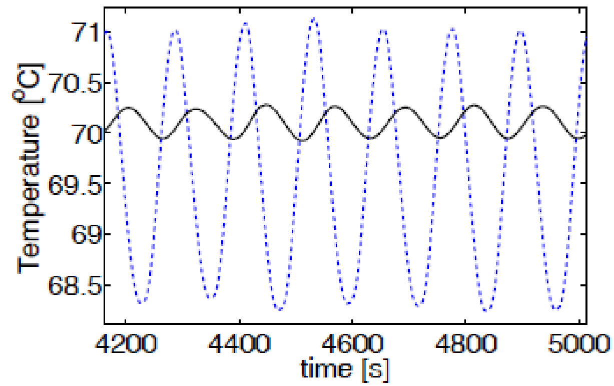
- The method works well in a stationary situation where the temperatures are approximately constant and kA does not change fast.
- Good estimation of the amplitudes is difficult in transient situations

  → Large deviations, slow convergence

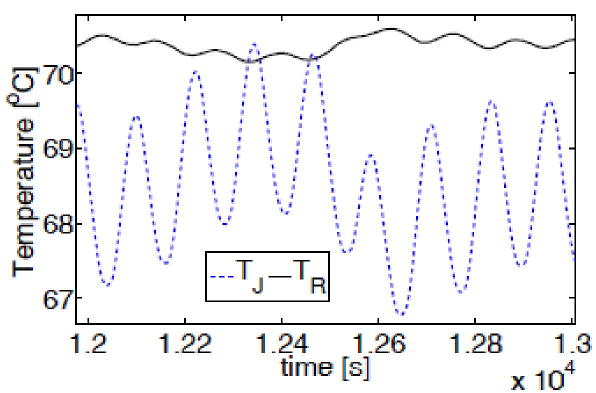
Typical Signals in TOC



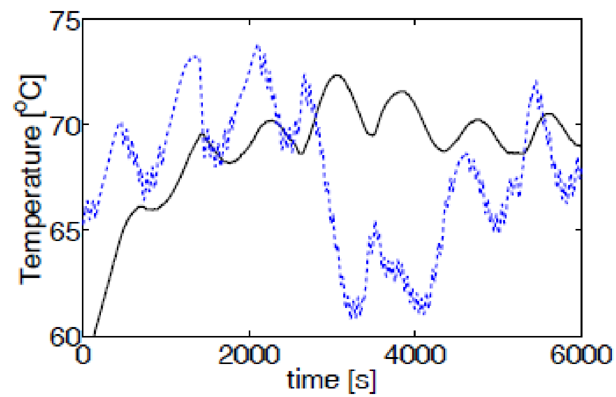
(a) Trajectories of T_R and T_J in calorimeter



(b) Detail enlargement of (a)



(c) Another detail enlargement of (a)



(d) Trajectories of T_R and T_J in a $1\ l$ glass reactor

New Approach to TOC

Alternative evaluation schemes by Wolfgang Mauntz:

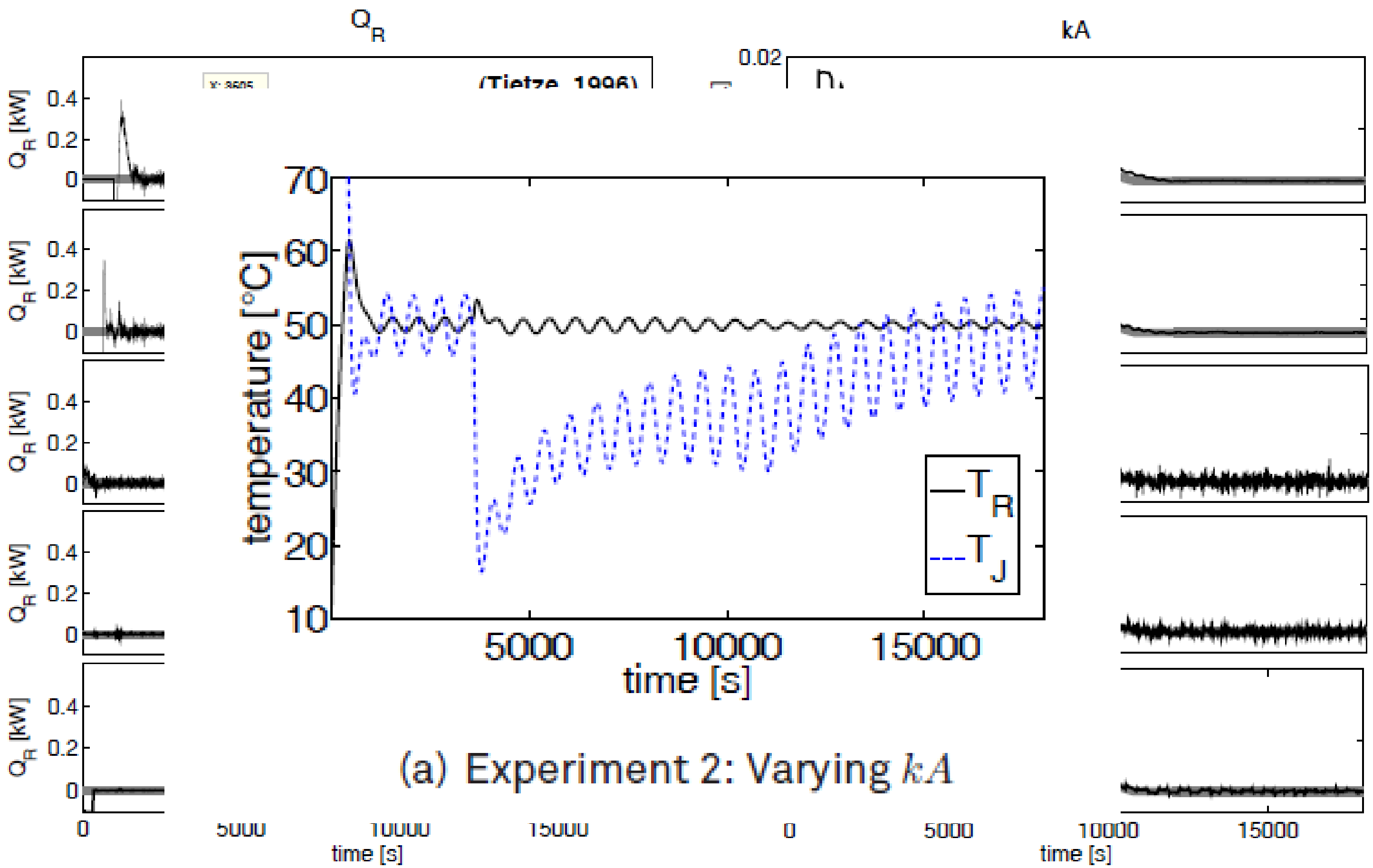
- Representation of the signal by sine plus drift
- Least-squares estimate in moving data window
- Use reactor heat balance + its time derivative (2nd order)

$$\frac{dT_R}{dt} = \frac{1}{C_{p,R}}(\dot{Q}_R + kA(T_J - T_R))$$

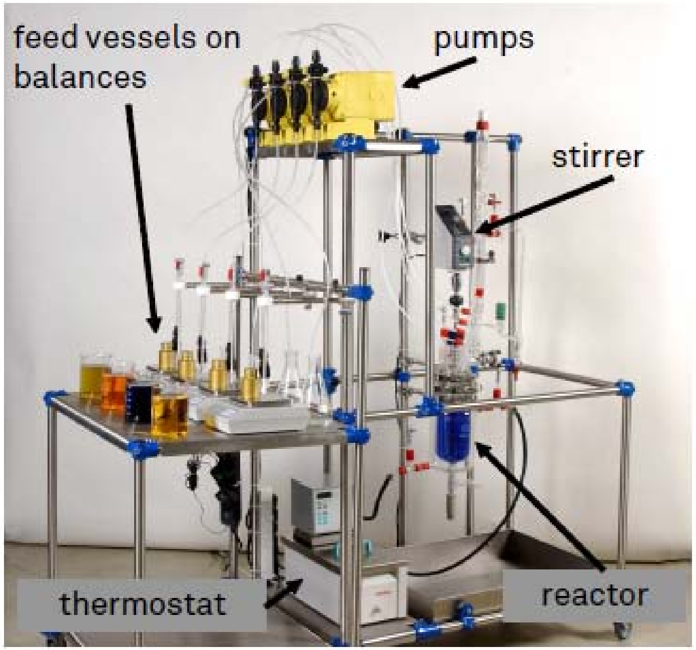
$$\frac{d\frac{d\,T_R}{dt}}{dt} = \frac{1}{C_{p,R}}\left[kA\left(\frac{d\,T_J}{dt} - \frac{d\,T_R}{dt}\right)\right]$$

- Moving horizon estimator

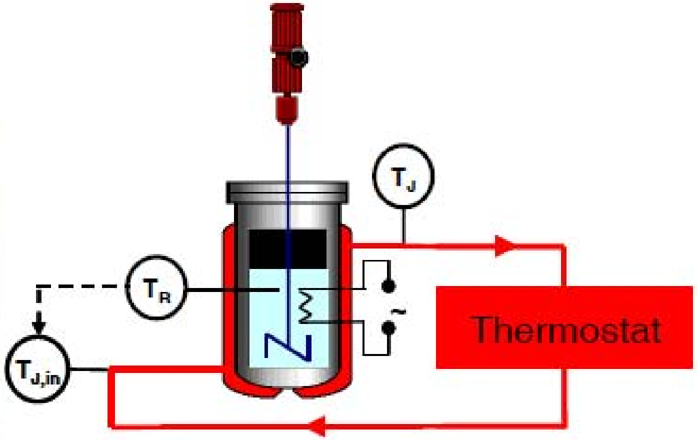
Comparison of Different Schemes



Experimental Reactor



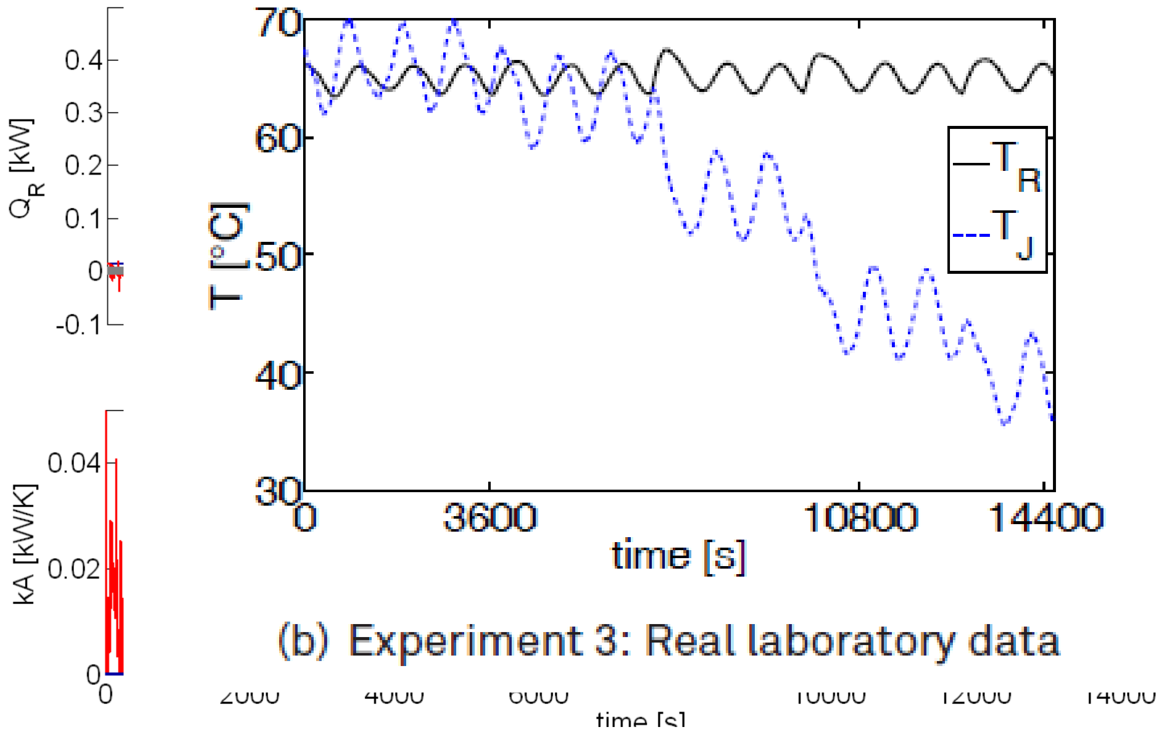
(a) Photograph



(b) Sketch

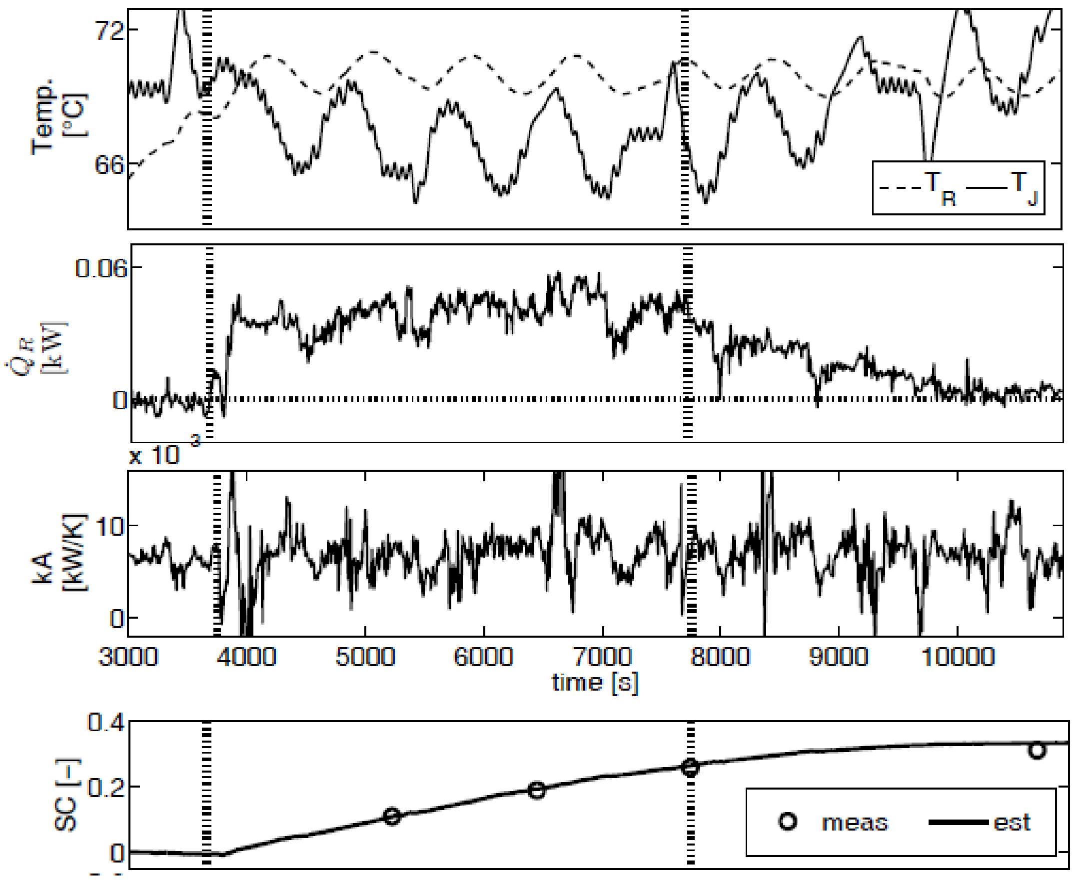
Figure 5.10: The 1 l reactor setup

New TOC: Experimental Results



(b) Experiment 3: Real laboratory data

Experimental Results: Polymerization



Results obtained with TOC and 2nd order model in the EKF for a co-polymerization of styrene and butyl acrylate in the 1l reactor

Summary and Conclusions

- Reaction calorimetry is a widely used method to estimate conversion in (semi-)batch processes
- EKF can estimate Q_R and kA simulataneously
- If the jacket is not behaving as a CSTR (low flow rates, large reactors), a PFR model of the jacket should be used in the estimator
- If the temperature difference between the jacket inlet and outlet is small (high jacket flow rate compared to the volume), traditional calorimetry fails
- Temperature Oscillation Calorimetry is a solution to this problem
- Data analysis using a second order derivative model in a EKF performs very well also in transient situations
- The exitation signal can be optimized → triangular shape