Model-Based Experimental Analysis

A Systems Approach to Mechanistic Modeling of Reactive Systems

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Modeling of Product and Process Systems

product & process systems

kinetic phenomena

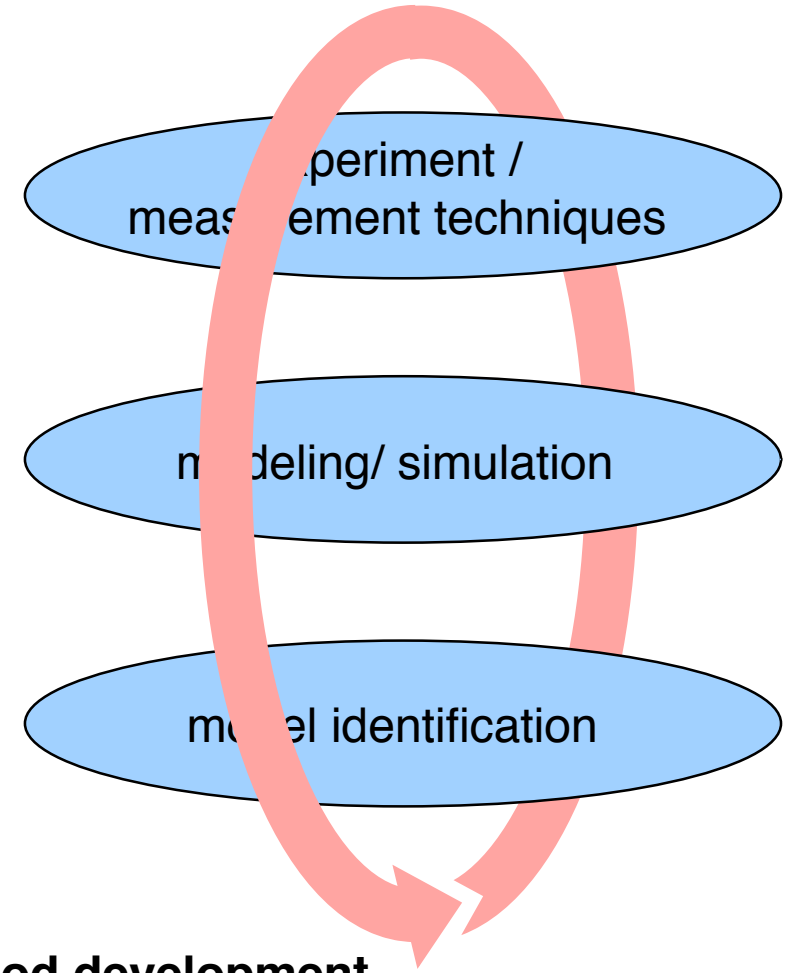
- diffusion
- heat and mass transfer
- interface phenomena
- multiphase transport
- (bio-)chemical reaction
- nucleation ...

multi-phase reaction and separation **processes**

fluid, structured & functional **products**

biological systems

experimental and theoretical methods & tools

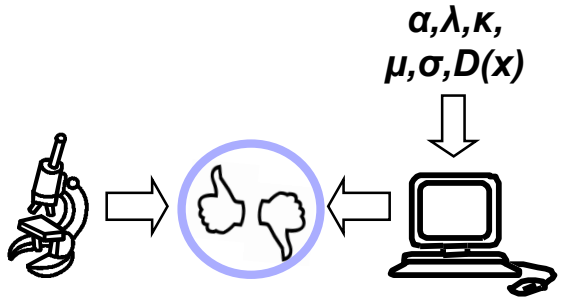


integrated method development
towards a work process of model-based experimental analysis

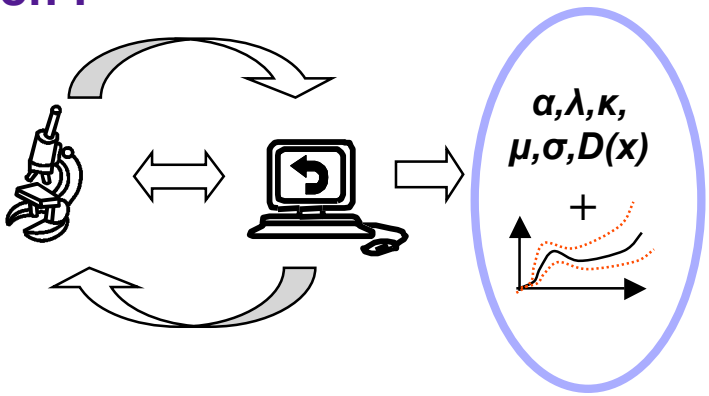
Integration of Modeling and Experimentation

cf. J.V. Beck, *Meas. Sci. Techn.* 9 (1998)

- **common approach in research and industrial practice**
  - coupled phenomena
  - detailed models, numerical case studies
  - comparison of simulation and experimental results
  - **evaluation of the model, but no model identification !**

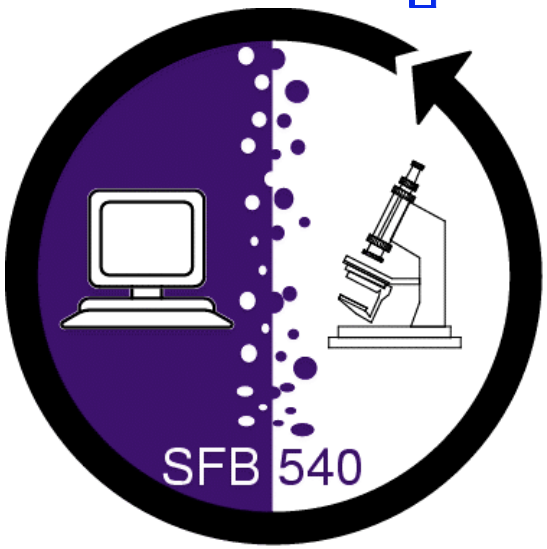


- **suggested future approach**
  - coordinated design of model and experiment
  - model refinement based on experimental evidence
  - accounting for inevitable measurement errors
  - **identification of a valid (mechanistic) model (structure & parameters) !**



model-based experimental analysis – MEXA:
valid models at minimal effort

Model-based Experimental Analysis of Kinetic Phenomena in Fluid Multi-phase Reactive Systems



12 research groups with cross-disciplinary expertise

- biotechnolgy (Ansorge-Schuhmacher)
- biochemical engineering (Büchs)
- reaction engineering (Greiner, Leitner)
- thermal separations (Pfennig)
- transport phenomena (Kneer)
- multiphase fluid dynamics (Modigell)
- computational engineering science (Behr)
- process systems engineering (Bardow, Marquardt)
- numerical mathematics (Reusken)
- scientific computing (Bischof, Bücker)
- NMR imaging (Blümich, Stapf)
- optical spectroscopy (Koß, Lucas, Poprawe)

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Director: W. Marquardt

Research Agenda of CRC 540

development of a work process
Model-based Experimental Analysis (MEXA)
for systematic mechanistic modeling by means of multi-method integration
and
its application to **modeling of multi-phase fluid reaction processes**

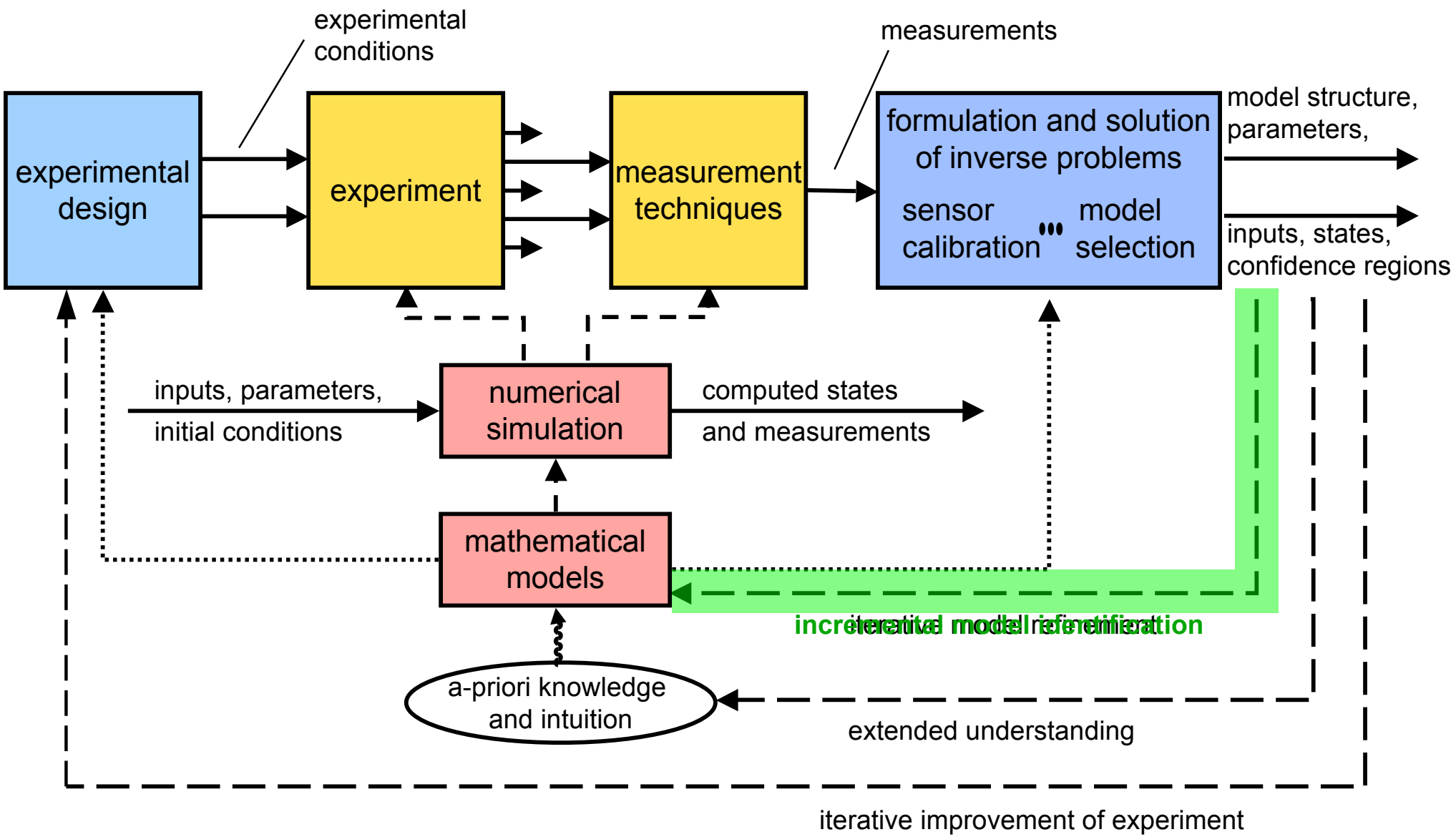
reaction and multi-component diffusion in liquids

transport and (bio-)reaction in heterogeneous systems

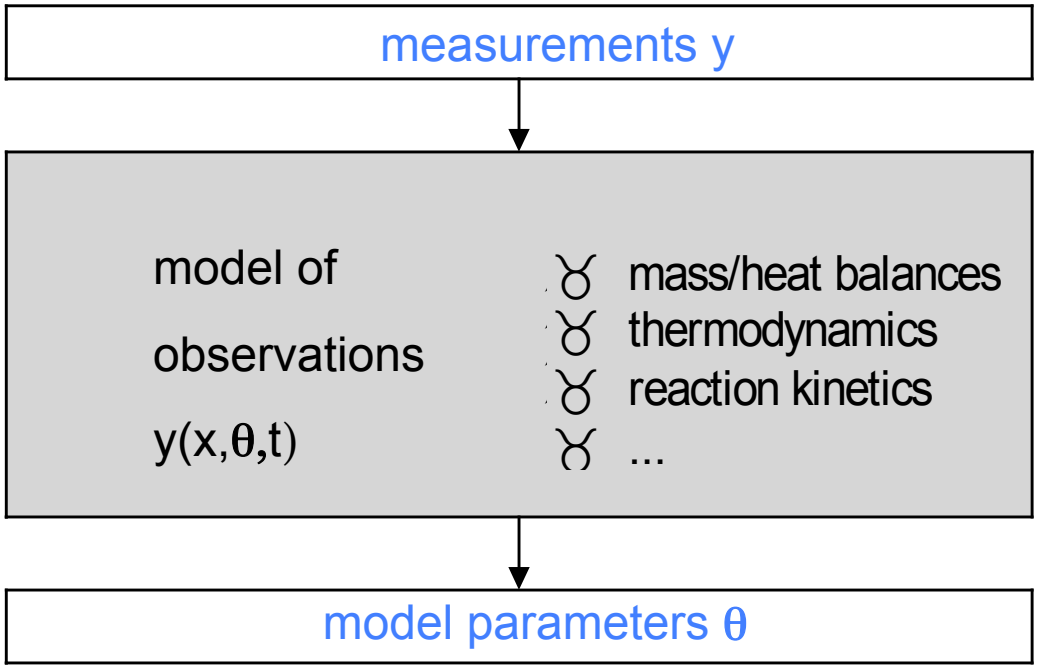
transport and reaction in single dispersed liquid droplets

transport and reaction in liquid falling films

MEXA Methodology



Simultaneous Model Identification



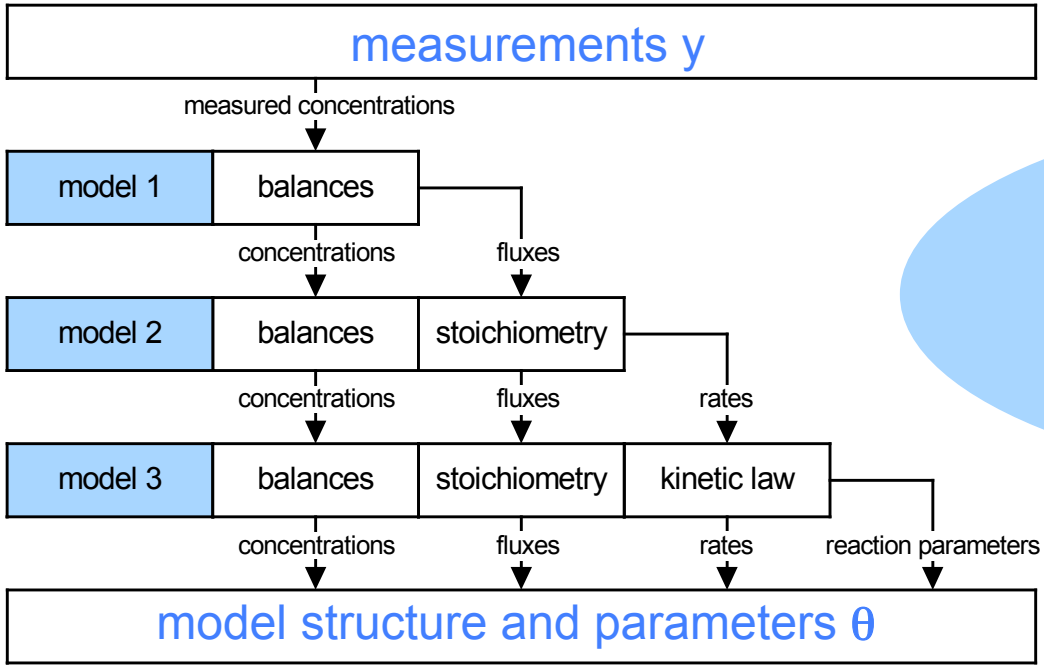
Overall process model y(x,θ,t) is fitted to experimental data:

$$\min_{\grave{e}} \frac{1}{2} \sum_{i=1}^{n} w_i \sum_{j=1}^{m} \left(y(\mathbf{x}, \grave{e}, t_j) - \tilde{y}(t_j) \right)^2$$

s.t. dynamic model & constraints

- What if we do not know any candidate model structure ?
- How to select a suitable model structure ?
- Is bias due to model structure defects or a lack of information content in data ?
- How to deal with very few or very many observations ?
- How to deal with convergence & robustness problems of estimation algorithm?

Incremental Model Identification



Decompose the model identification and selection problem into fully transparent steps !

- **computationally efficient (minutes rather than days)**
- **numerically robust and fully transparent**
- **a-priori knowledge can be integrated into the identification process**
- **complex and interacting kinetic phenomena can be identified**

Incremental Identification – Some Examples

- **differential methods in reaction kinetics**, e.g. Connors (1990)
  → estimate reaction rate by FD, then estimate kinetic parameters

- **hybrid modeling**, e.g. Psichogios & Ungar (1992), Tholudor & Ramirez (1999)
  → combine first-principles models with neural nets

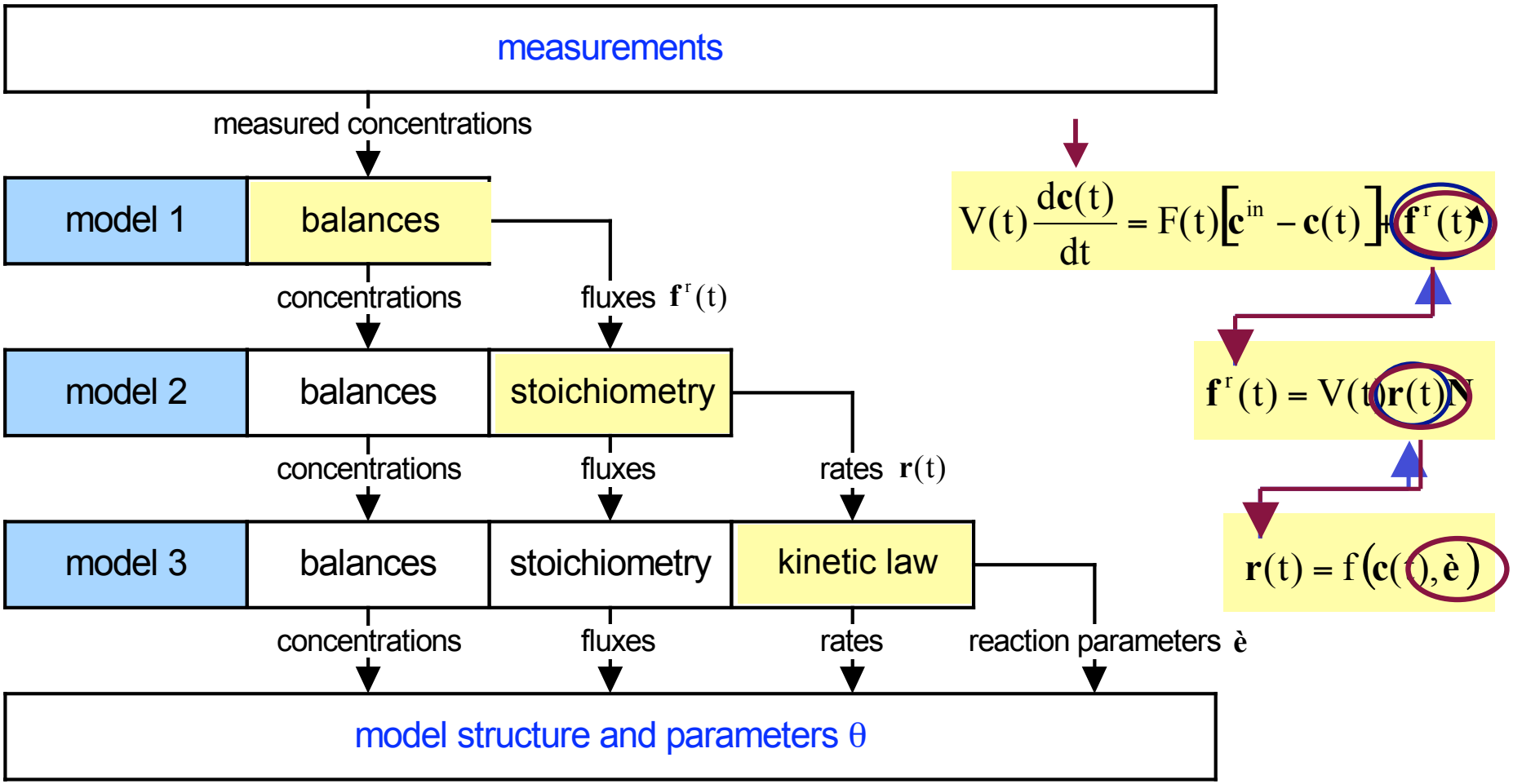
- **inverse problems in population balances**, Mahoney, Doyle & Ramkrishna (2002)
  → calculate growth rate as model-based data, correlate with states

- **derived from intuition and physical insight**
- **often ad-hoc methods**
- **problem-specific solutions**

a generice principle ?

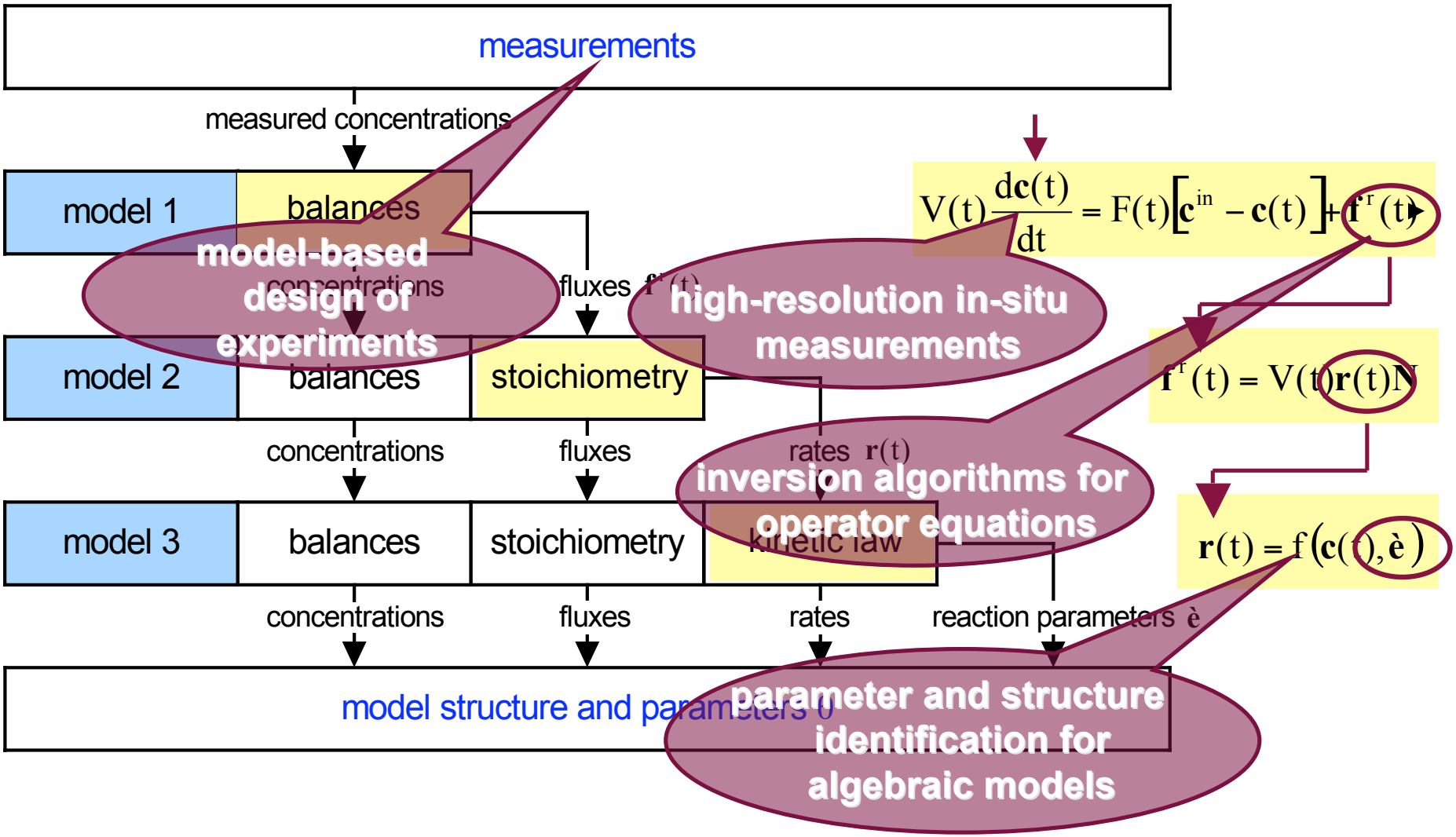
Incremental Model Development

Illustration with reaction kinetic identification in a CSTR



(Marquardt, 1998)

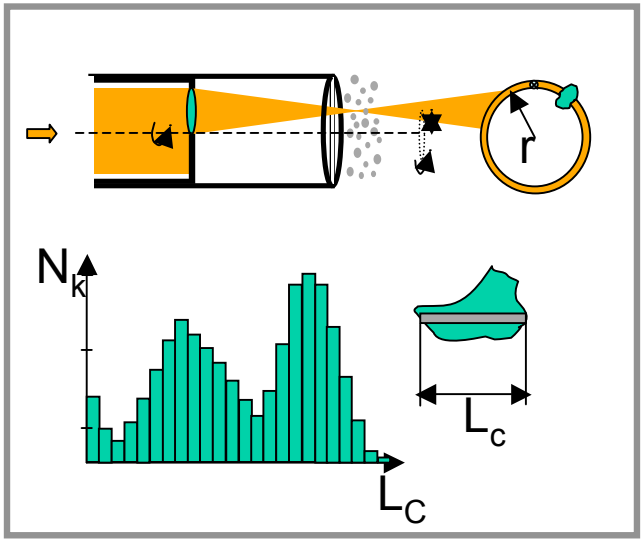
What are the ingredients for implementation ?



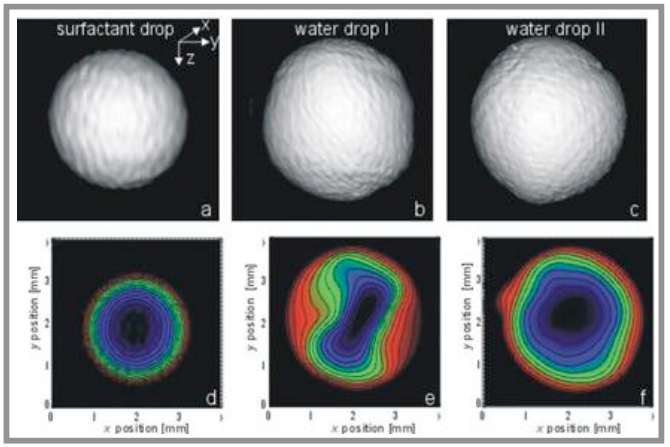
non-invasive, in-situ measurements of field data

- **observation of qualitative behavior**
- **quantitative characterisation of kinetic phenomena**

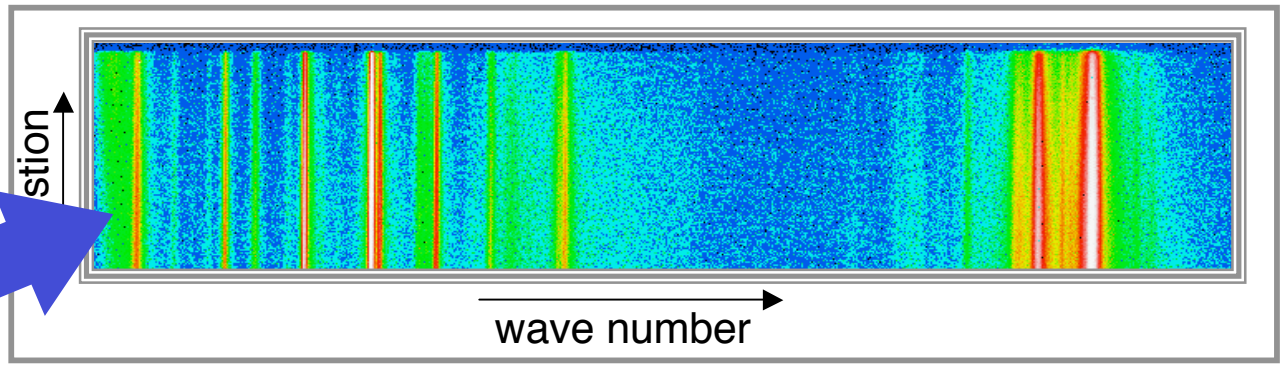
particle size distribution (FBRM)
Kail, Briesen, Marquardt et al., LPT



velocity profiles in a levitated droplet, NMR imaging, Blümich et al., CRC 540



conencentrations on a line,
Raman spectrosc
Koß, Lucas et al.



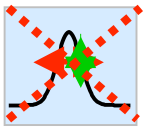
interpretation of the primary measurement data, calibration, quantification of measurement errors !

Problems of Established Analysis Methods

Most established spectral analysis methods such as

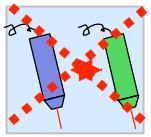
- **PCA, PLS or**
- **classical least squares**

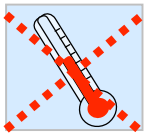
are linear and cannot model all nonlinear effects that occur in real mixtures:



Non-linear effects due to molecular interaction

Reactive mixtures: restricted extrapolability





Replacement of measurement device or varying temperatures

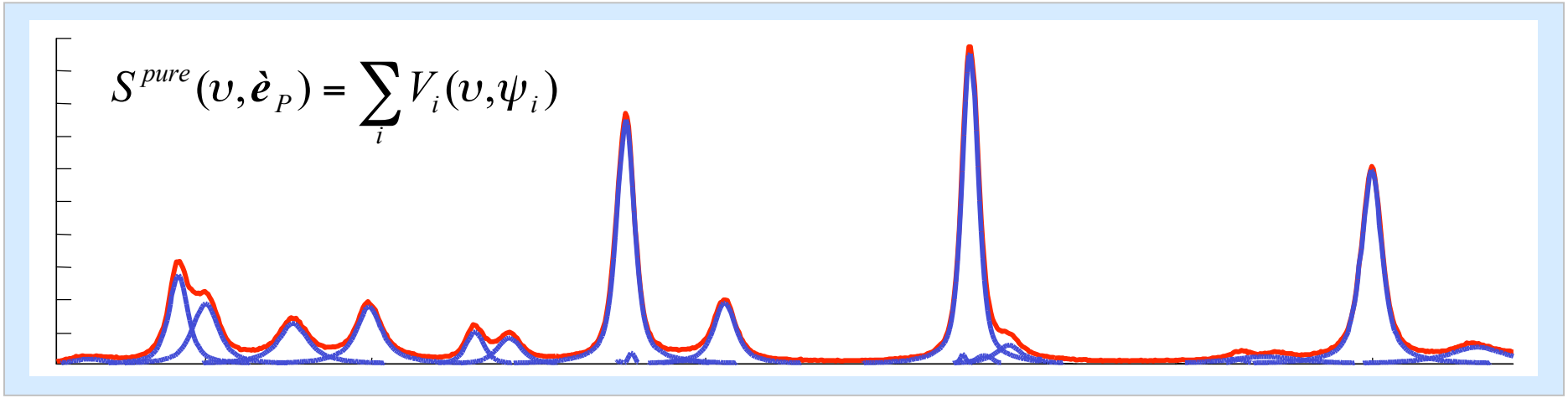
Indirect Hard Modeling – General Idea

Indirect Hard Modeling: a nonlinear spectral analysis approach

Development of a rigorous mathematical model of the spectrum

Consideration of physical effects of the spectrum through phenomenological modeling

Step 1: Modeling of pure component spectra (during calibration)

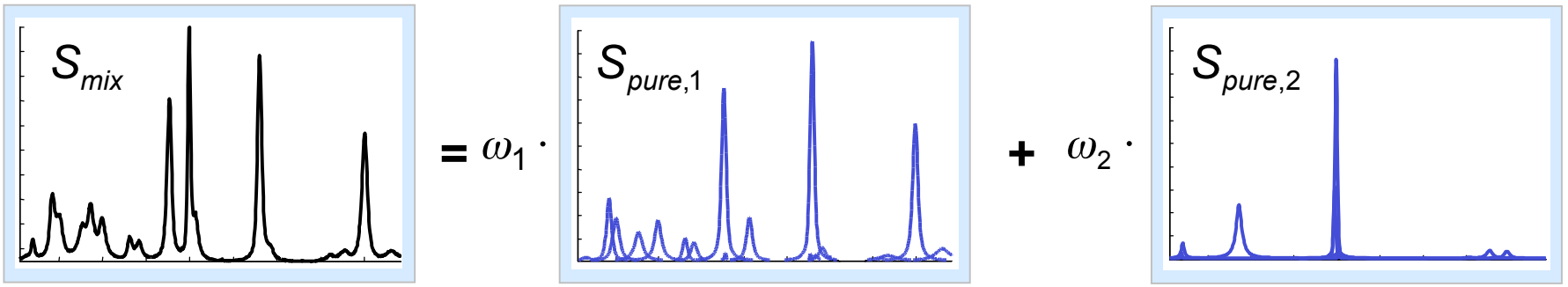


Generation of **pure component models** using **automatic peak fitting algorithm**

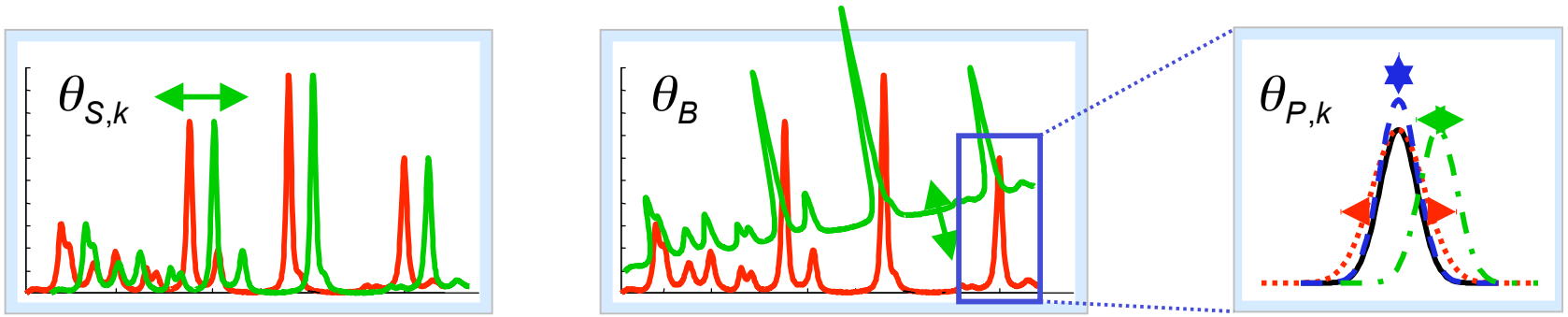
(Alsmeyer et al., *Applied Spectroscopy,* 58 (8), 2004)

Model of the Mixture Spectrum

Step 2: Mixture spectrum is modeled as**:**
Linear combination of parameterized non-linear pure component models



All non-linear effects are modeled phenomenologically:

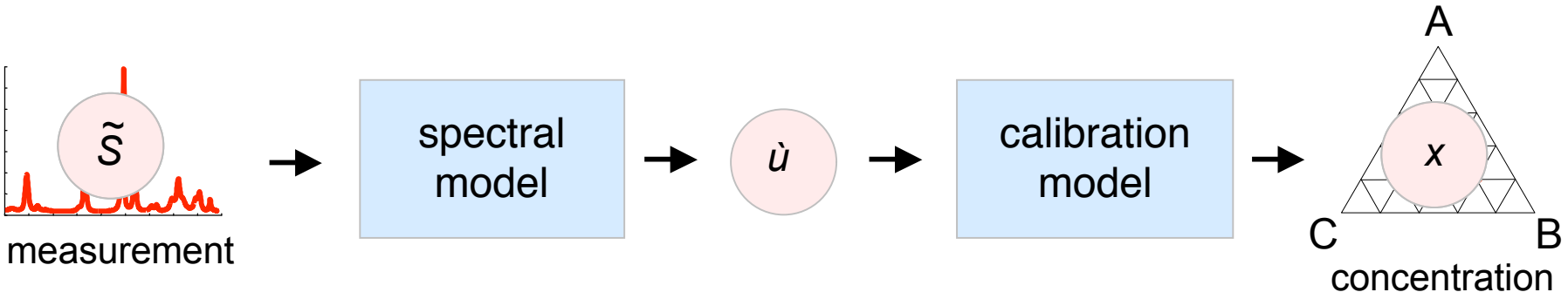


Spectral shifts | Baseline variations | Peak variations

Full spectral model:

$$S(\upsilon) = B(\theta_B) + \sum_k \omega_k \cdot S_k^{pure}(\upsilon, \theta_{P,k}, \theta_{S,k})$$

Calibration



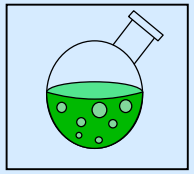
Linear, physically motivated calibration model

(**Non-linear effects** are **corrected** by the spectral model)

$$\frac{\omega_i}{\omega_j} = K_{i,j}\frac{x_i}{x_j} \quad \Rightarrow x_i = \frac{1}{\sum_k \frac{\omega_k}{\omega_i}\frac{1}{K_{k,i}}}$$

Highly reduced amount of calibration measurements

Theory: **One** calibration measurement
Practice: **Few** measurements



Good extrapolability:

Calibration in concentration subspace is possible

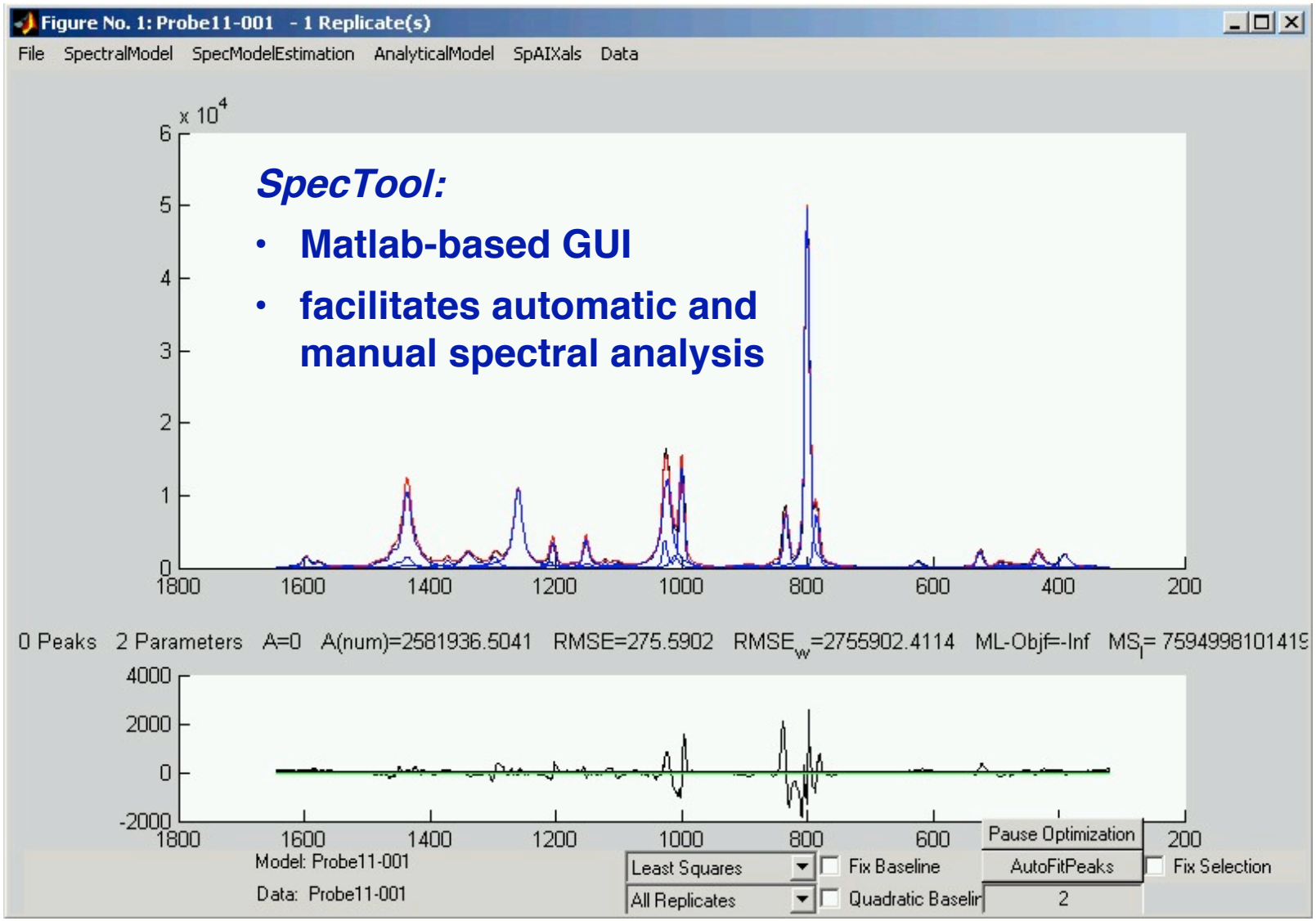
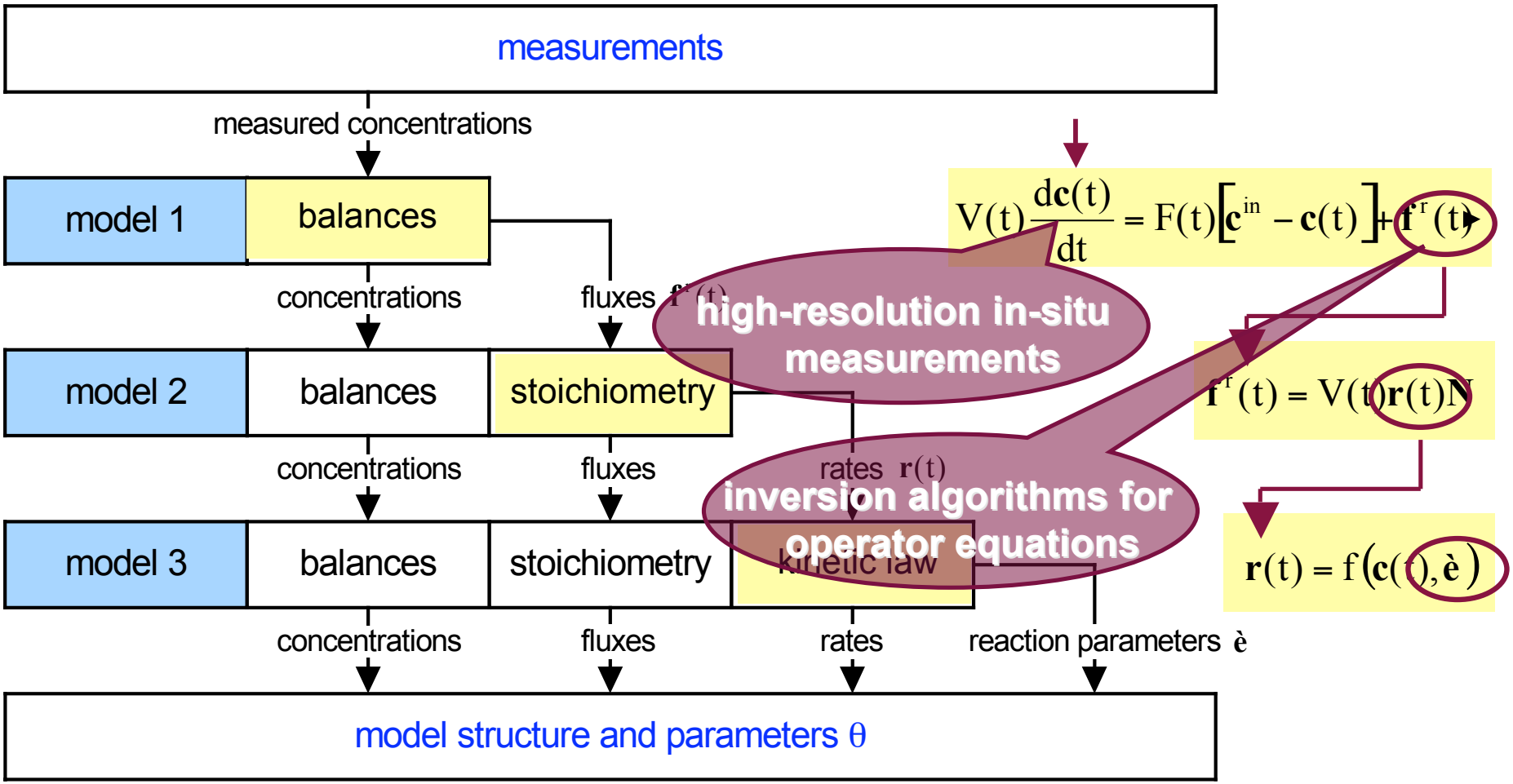


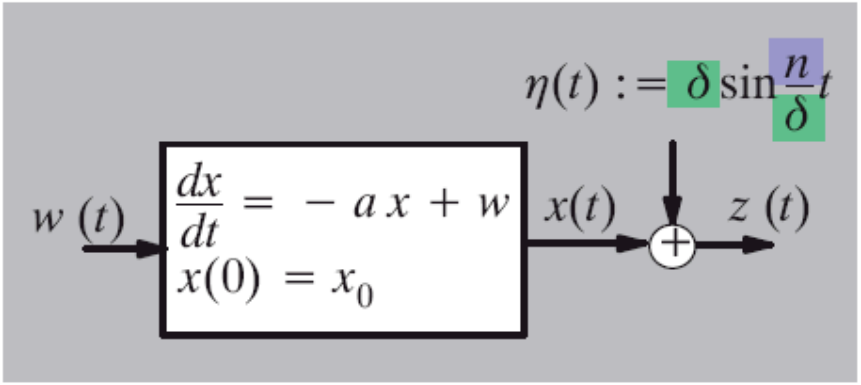
Figure No. 1: Probe11-001 - 1 Replicate(s)
File SpectralModel SpecModelEstimation AnalyticalModel SpAIXals Data
x 10^4
6
5
4
3
2
1
0
1800 1600 1400 1200 1000 800 600 400 200
SpecTool:
• Matlab-based GUI
• facilitates automatic and manual spectral analysis
0 Peaks 2 Parameters A=0 A(num)=2581936.5041 RMSE=275.5902 RMSE_w=2755902.4114 ML-Objf=-Inf MS_I= 7594998101419
4000
2000
0
-2000
1800 1600 1400 1200 1000 800 600 200
Model: Probe11-001
Data: Probe11-001
Least Squares
All Replicates
Fix Baseline
Quadratic Baselir
Pause Optimization
AutoFitPeaks
2
Fix Selection

What are the ingredients for implementation ?



Inversion Problems are Ill-Posed

illustrating linear example (scalar):



a problem is well–posed if

- a solution exists
- the solution is unique
- small errors in the data lead to small errors in the solution

(Hadamard, 1923)

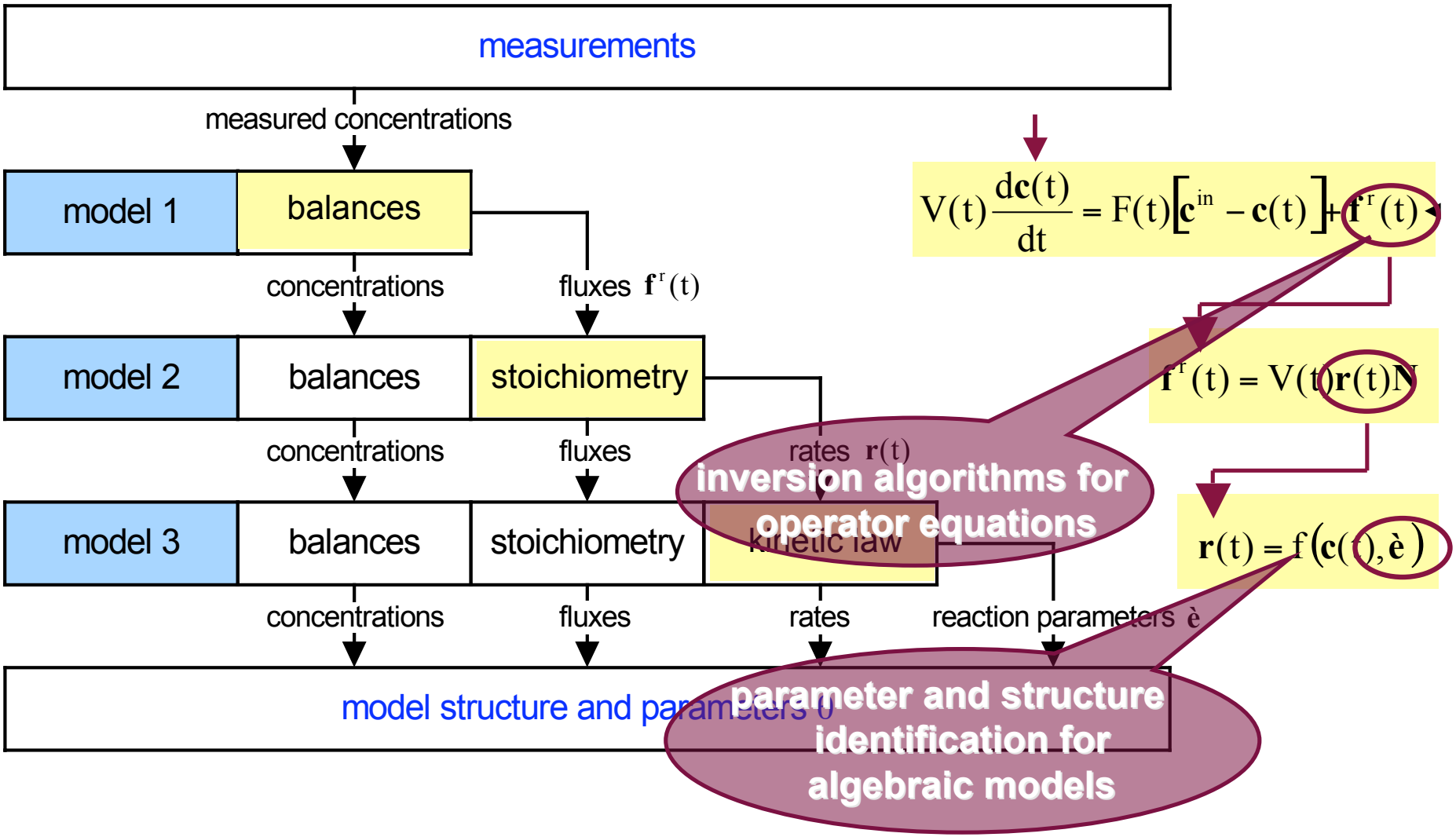
Issues in Flux Estimation

- decide on a set of measurements for best identifiablility
  - at least as many measurements as unknown fluxes (Hirschorn, 1979)
  - ➢ methods for quantification of identifiability (e.g. Asprey, 2003)

- balance information content of measurements and resolution of the flux parameterization
  - choose spatial and temporal resolution of flux function
  - ➢ adaptive discretization methods (e.g. Binder et al. 2000)

- compromise between bias and variance in estimates
  - balanced choice of discretization, early stopping and regularization
  - systematic methods for the selection of regularization operators and multiple regularization parameters (e.g. Ascher, Haber, 2001, Engl et al., 1996, Belge et al. 2002)

What are the ingredients for implementation ?



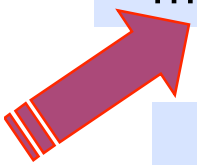
Flux Model Selection Problem

find the most appropriate functional representation

for the correlation of fluxes and states: structure and parameters

generate candidate model structures

- experience-based, qualitative reasoning, kinetic power laws (Schaich et al., 2001)
- molecular scale modeling (Barrett & Prausnitz, 1975, Liu et al., 1998)
- multivariate regression & data mining (Bates & Watts, 1988, Hastie et al. 2001)



parameter estimation and model structure selection

- error-in-variables formulation (Britt & Lücke, 1978, Boggs et al. 1992)
- inference approach, decision tree & statistical tests (Verheijen, 2003)
- Bayesian a-posteriori probability tests (Stewart et al., 1998)
- combinatorial search (McKay et al., 1997, Skrifvars et al., 1998)

Function Approximation

Given: noisy data set S

$$S = \{(\mathbf{x}_i, \tilde{y}_i) \in R^d \times R\}_{i=1}^{M} \qquad \tilde{y}_i = f(\mathbf{x}_i) + \varepsilon$$

Recover unknown function $f \in V$ from data S "as good as possible"

$$\min_{f_N \in V_N} \frac{1}{M} \sum_{i=1}^{N} \left(f_N(\mathbf{x}_i) - \tilde{y}_i\right)^2 + \lambda \Phi(f)$$

Binder *et al.* (2000)

Ascher and Haber (2001)

Restriction to finite dimensional subspace V_N

Tikhonov regularization term enforcing smoothness of f_N

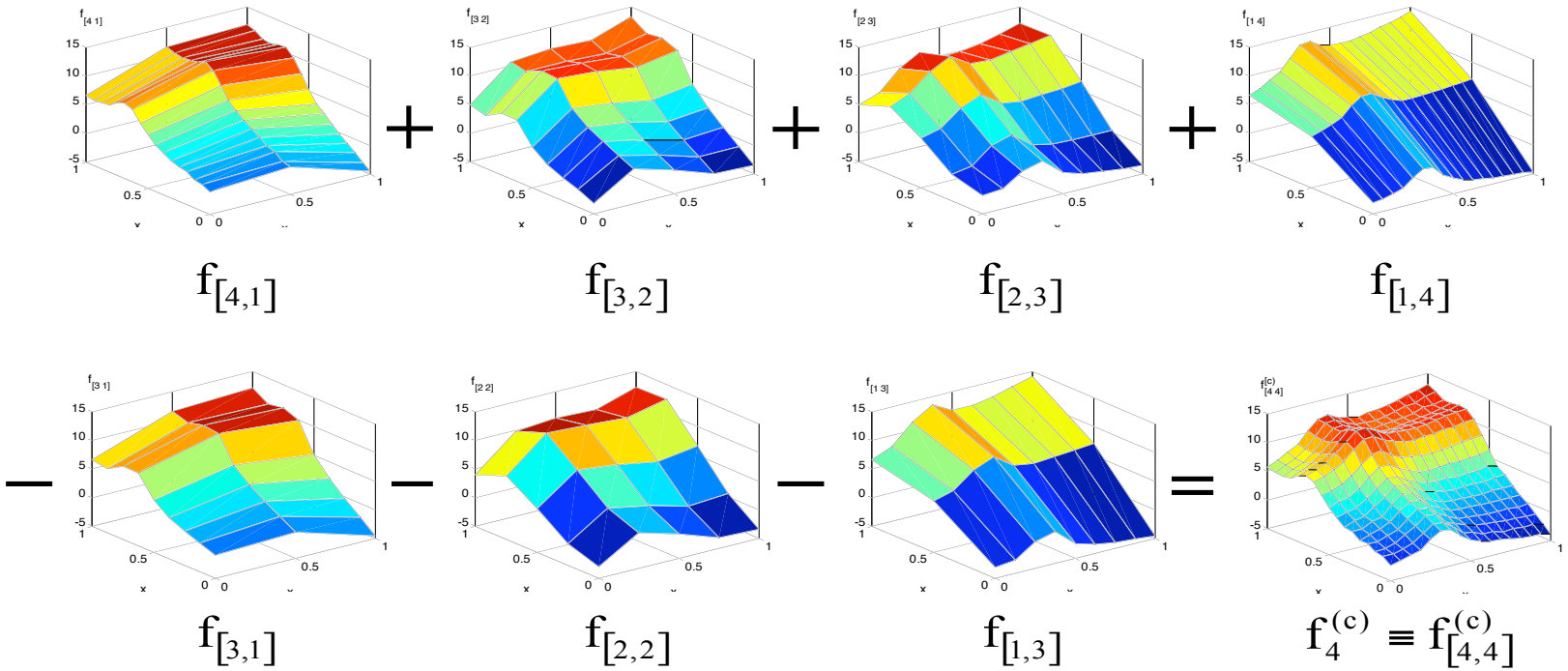
Desirable properties

- linear scaling with number of data points (avoid „curse of dimensionality“)
- properly exploit information content in data (avoid under-/overfitting)

Sparse Grid Approximation

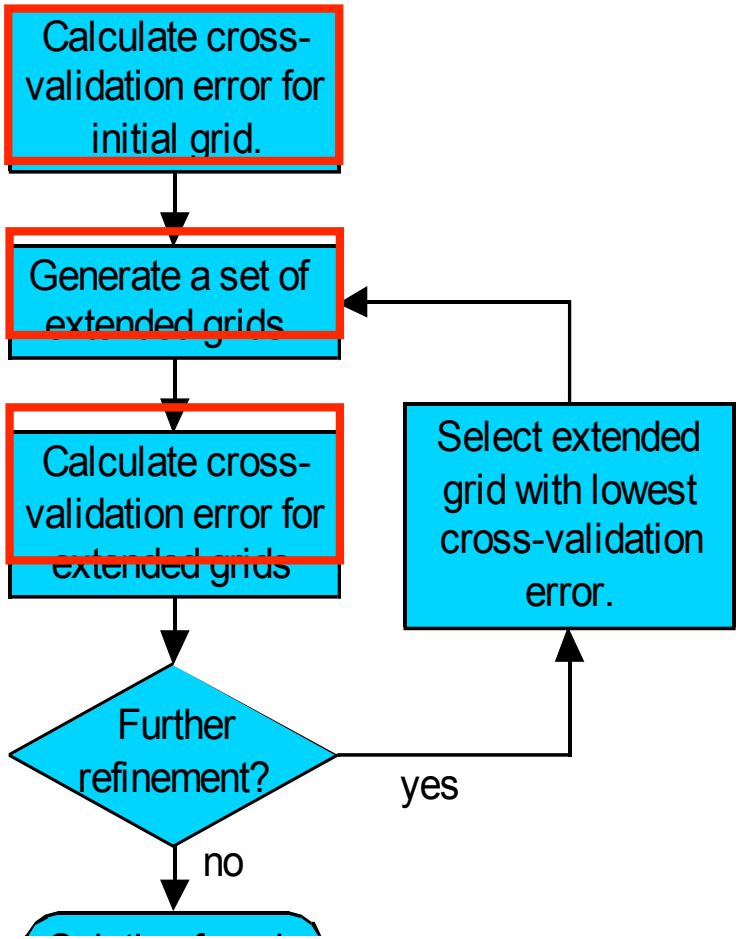
(Garcke et al., 2001; Brendel & Marquardt, 2003)

- hierarchical d-dimensional finite-element discretization of unknown function with d-linear hat functions, Yserentant (1992)
- significant reduction of number of parameters by successive approximation on subgrids and subsequent linear combination to a sparse grid approximation



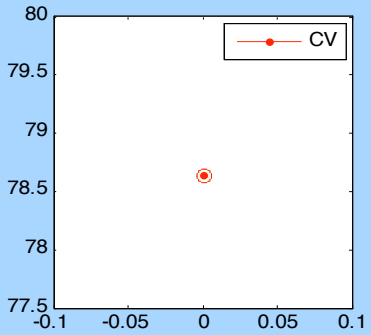
- approximation quality close to full grid approximation

Incremental Grid Refinement



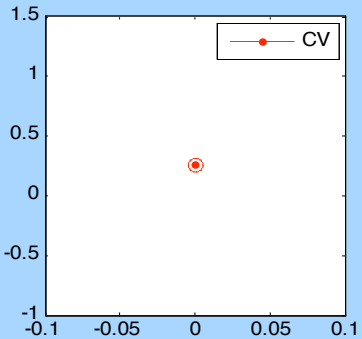
Initial grid:

Level: [-1 -1]



Extended grids:

Level: [0 -1]



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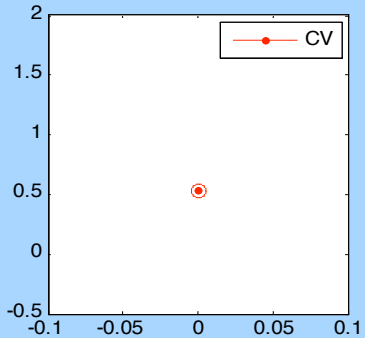

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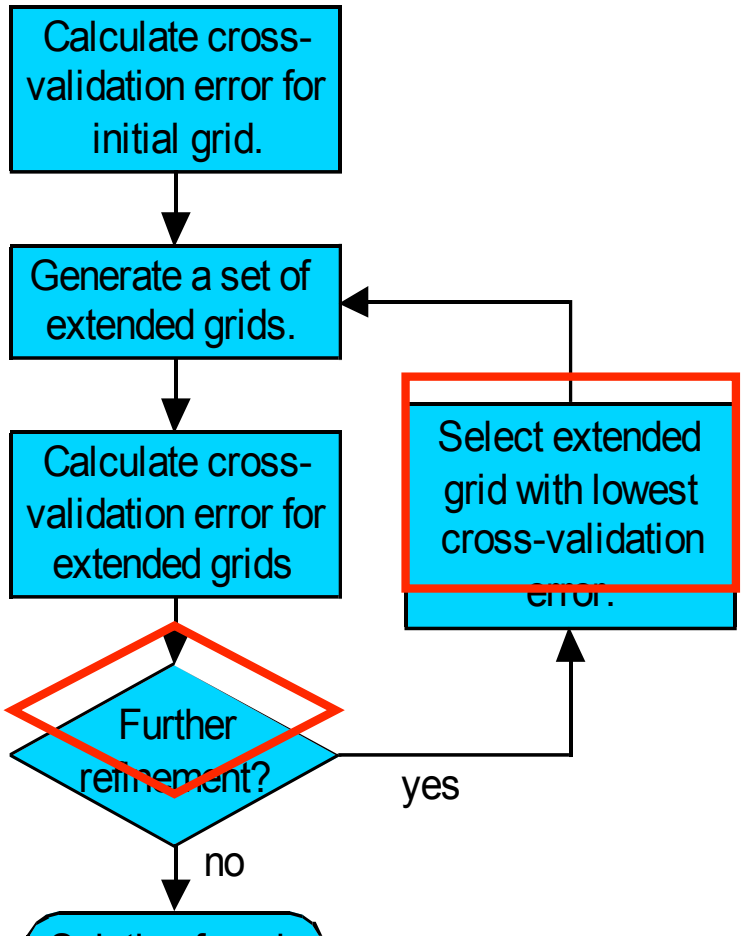

Initial grid:

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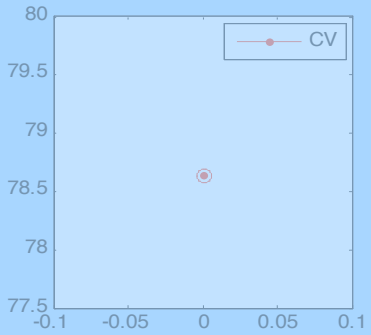

Extended grids:

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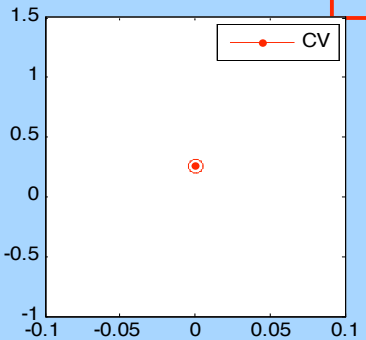

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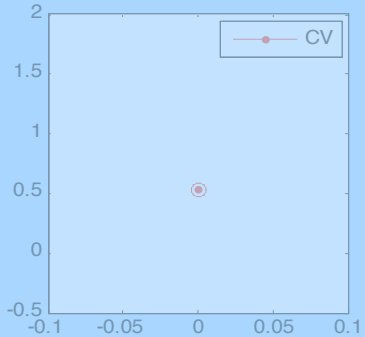
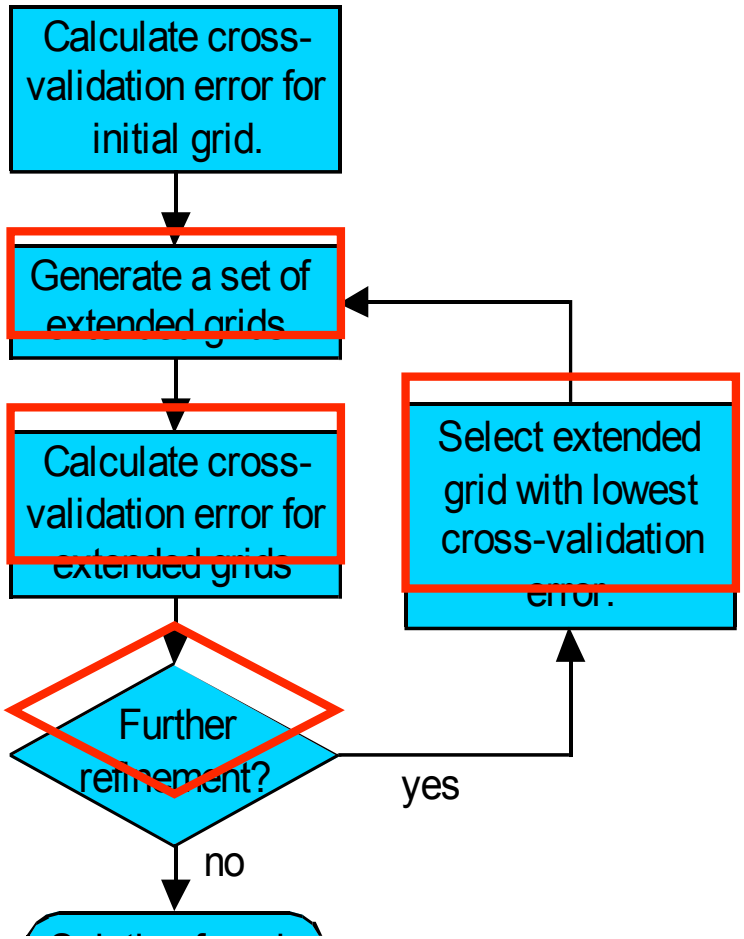
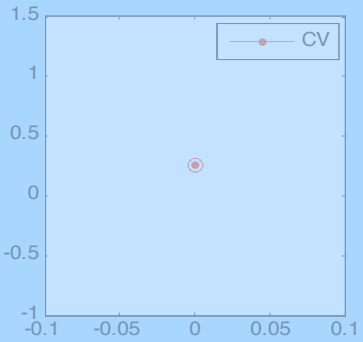


Incremental Grid Refinement



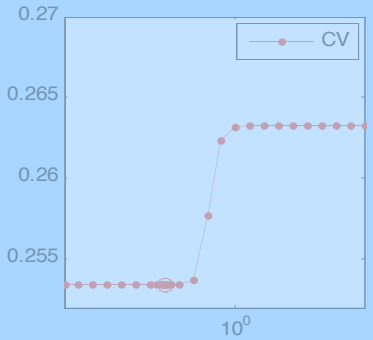
Current grid:

Level: [0 -1]



Extended grids:

Level: [1 -1]



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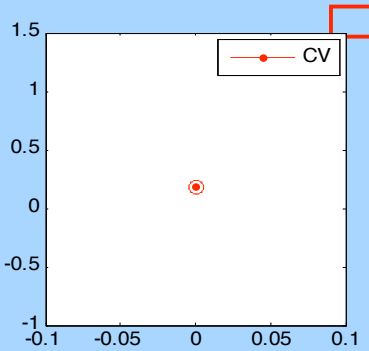
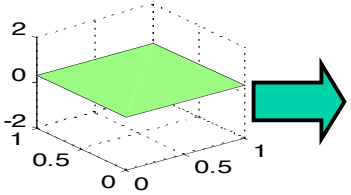


Incremental Grid Refinement

Initial grid

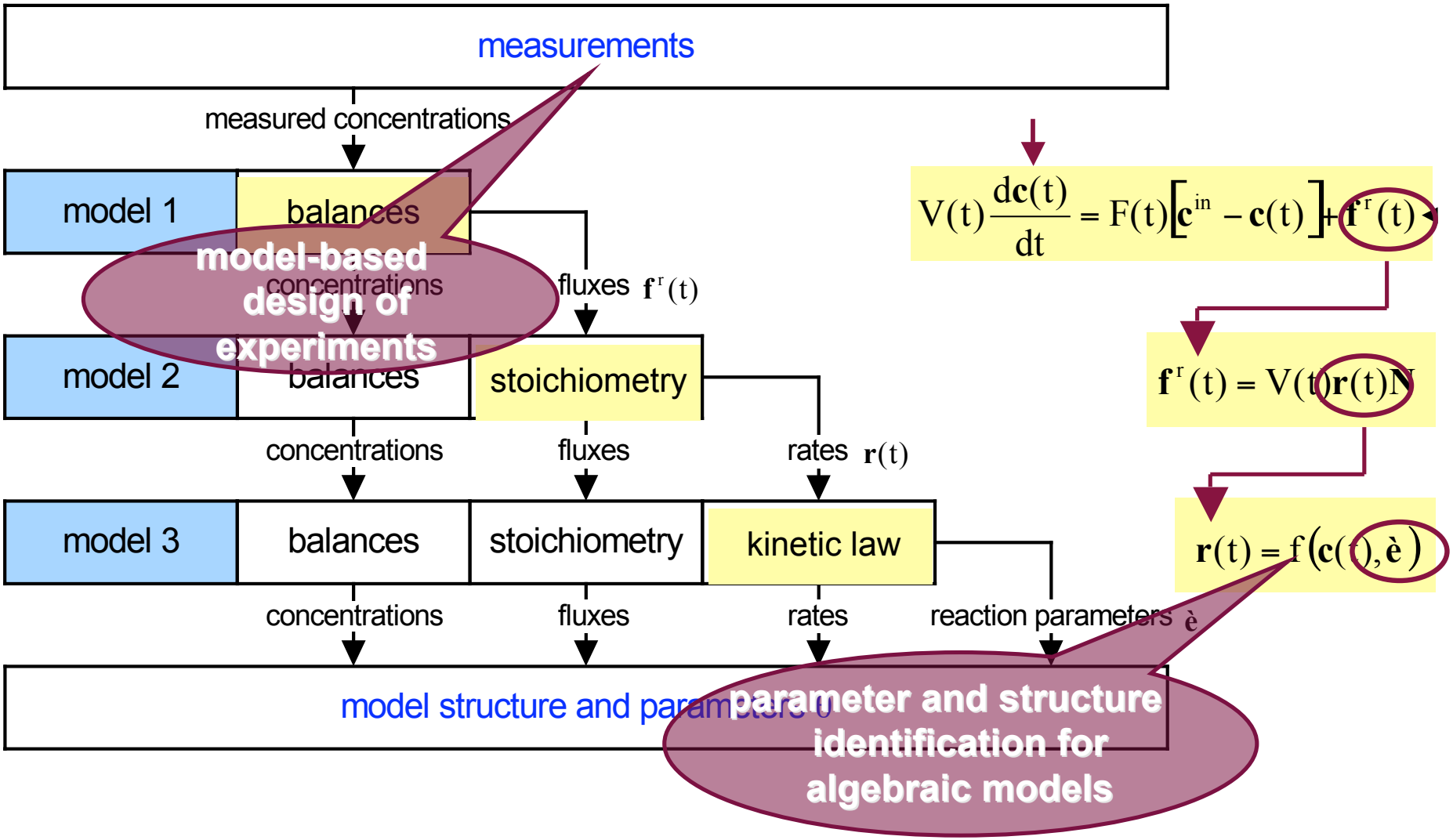


?

- number of tested grids scales linearly with number of dimensions instead of exponentially
- curse of dimensionality in selecting the discretization is avoided
- no guarantee to find optimal discretization

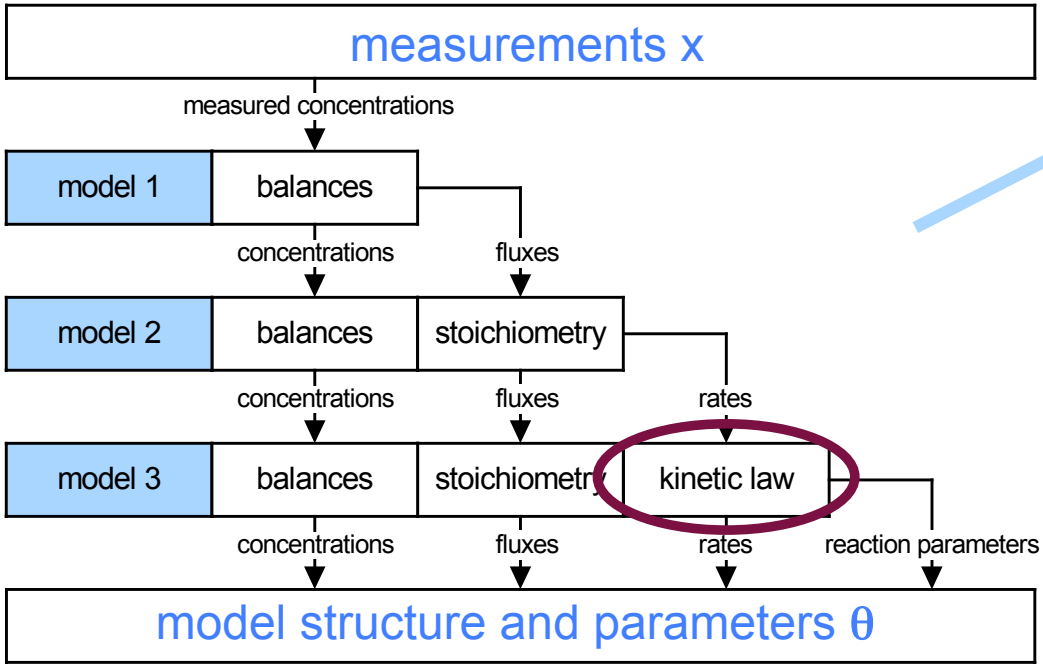
Final grid

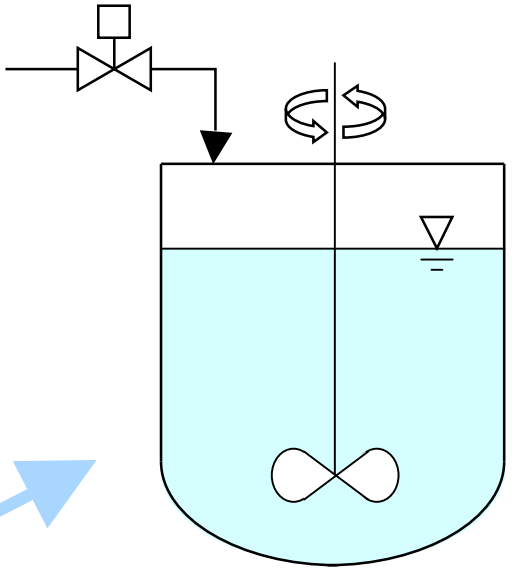
What are the ingredients for implementation ?



Modeling of Reaction Kinetics

- no. of reactions, stoichiometry and kinetics unknown
- isothermal semi-batch CSTR experiments
- concentration measurements (ex-situ, e.g. GC; in-situ, e.g. Raman/IR spectroscopy)
- a number of simulated semi-batch reactor experiments, 60 min (cases: noise, sampling …)



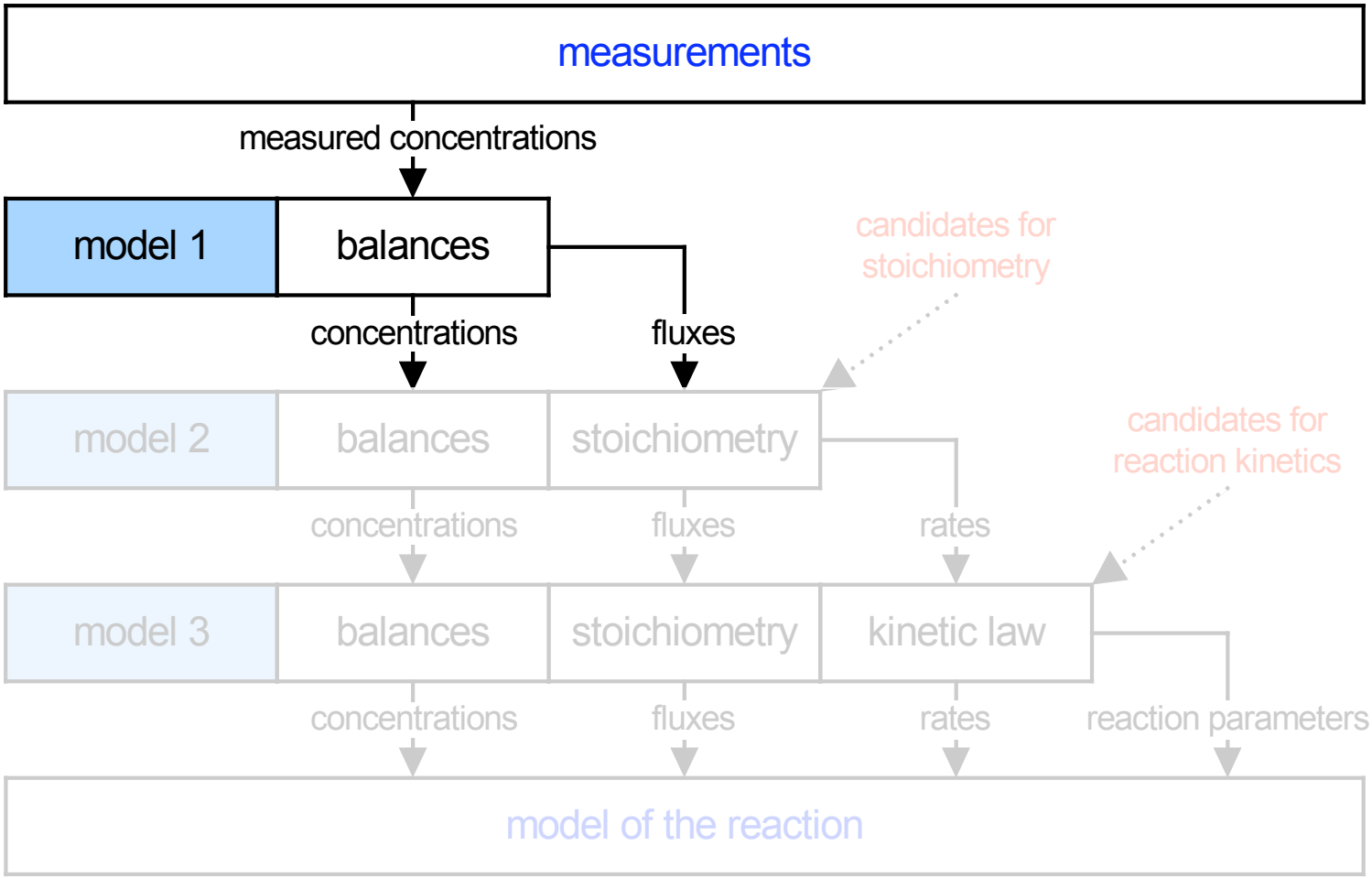


Acetoacetylation of Pyrrole with Diketene

1: P + D → PAA
2: D + D → DHA
3: D → OL
4: PAA + D → F

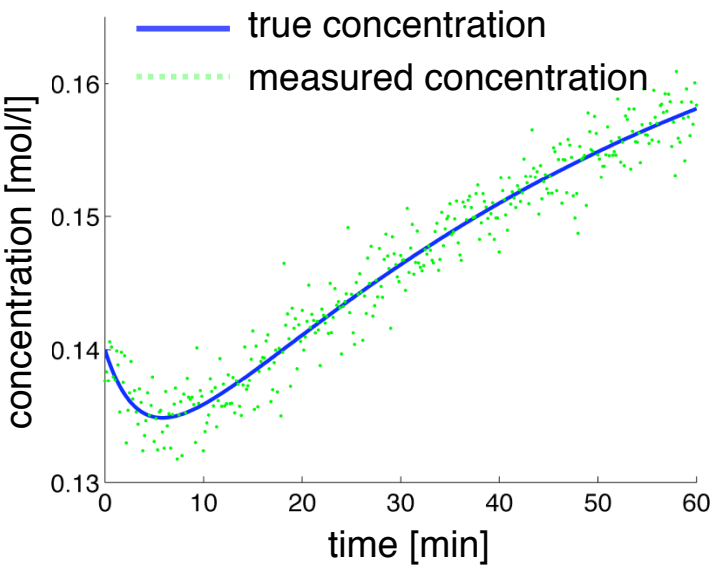
(Brendel, Bonvin, Marquardt, 2006)

Incremental Model Identification



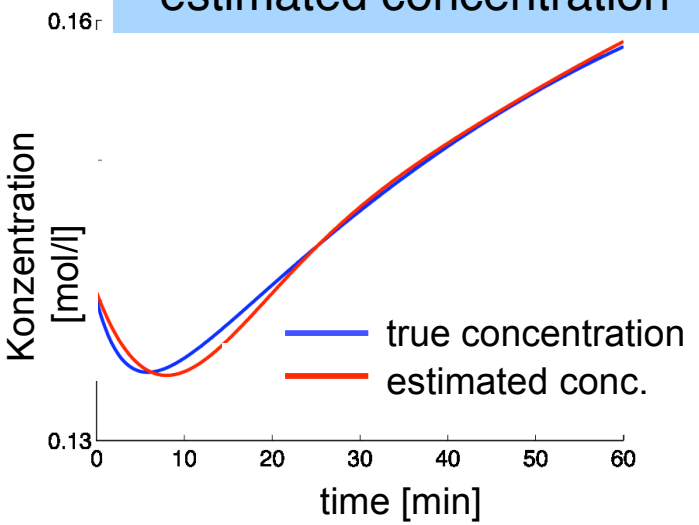
Concentration Measurements → Reaction Fluxes

concentration measurements



regularisation

estimated concentration

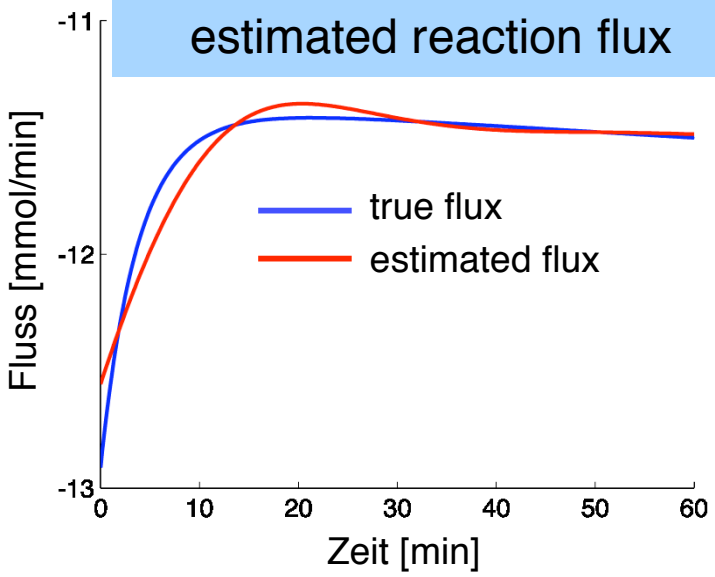


inverse problem:

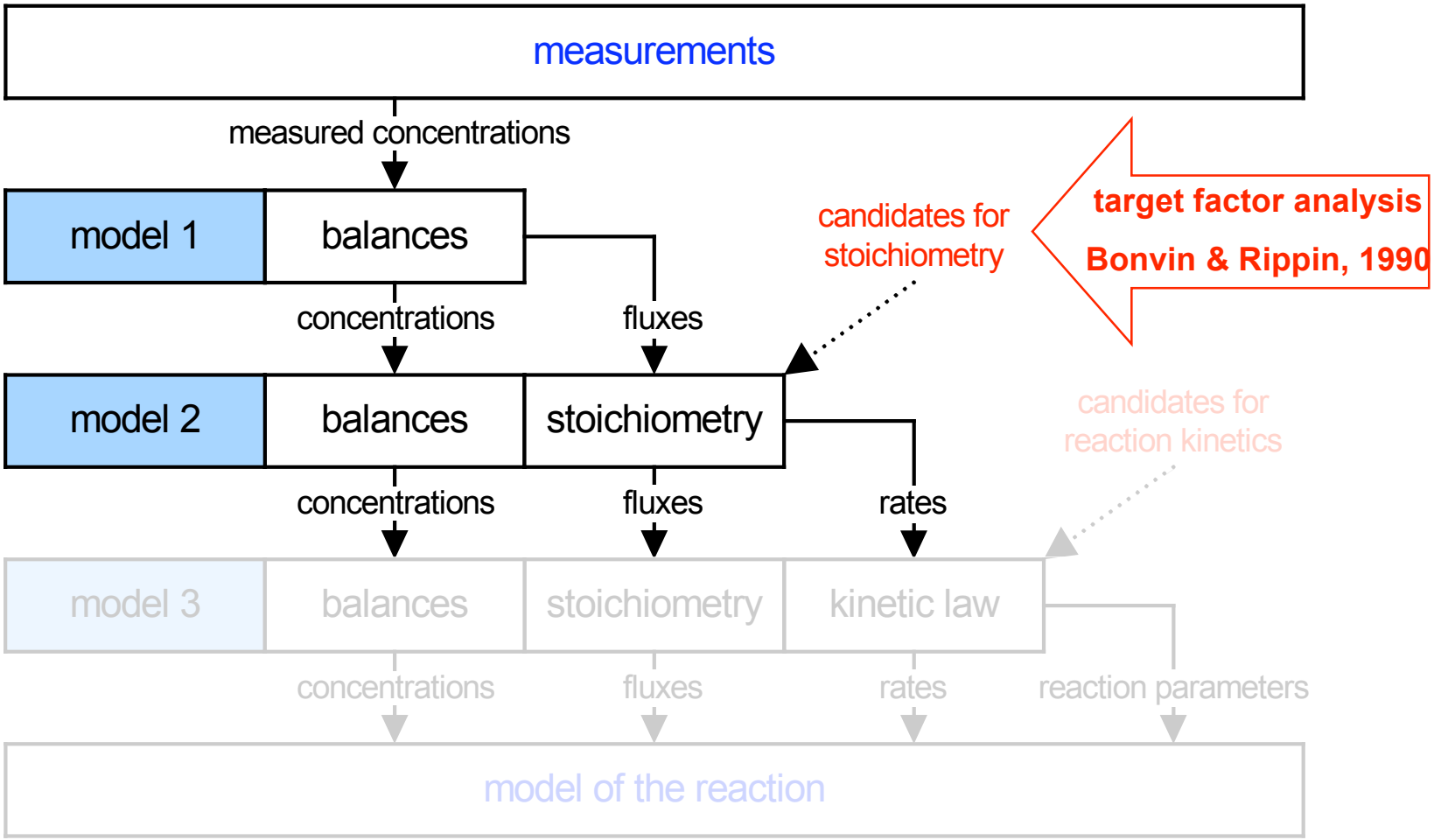
$$f_i^r(t) = V(t)\frac{dc_i(t)}{dt} - F\left[c_i^{in} - c_i(t)\right]$$

mass balances (semi-batch)

estimated reaction flux



Incremental Model Identification



Target Factor Analysis

Propose an R x S dimensional stoichiometric matrix $\boldsymbol{N_p}$ with

R = number of reactions

S = number of involved species

Set up a B x S dimensional data matrix $\boldsymbol{D}$

B = Number of observations

$\boldsymbol{D}$ = can be expressed as $\boldsymbol{X}*\boldsymbol{N}$, where $\boldsymbol{X}$ is a matrix indicating the extents of the R reactions and $\boldsymbol{N}$ is the correct stoichiometric matrix

Singular Value Decomposition → $\boldsymbol{D} = \underbrace{\boldsymbol{U}*\boldsymbol{S}}_{\boldsymbol{X_a}} * \underbrace{\boldsymbol{V^T}}_{\boldsymbol{N_a}}$,

where the rows of $\boldsymbol{N_a}$ represent a basis for the observed stoichiometric space

Discard those stoichiometries of $\boldsymbol{N_p}$ that cannot be approximately described as linear combination of the rows of $\boldsymbol{N_a}$

Stoichiometry

$$f_D^r = V(-r_1 - 2r_2 - r_3 - r_4)$$

$$f_P^r = V(-r_1)$$

$$f_{PAA}^r = V(r_1 - r_4)$$

$$f_{DHA}^r = V(r_2)$$

$$\mathbf{f}^r = V\mathbf{r}\mathbf{N}$$

Determination of reaction rates

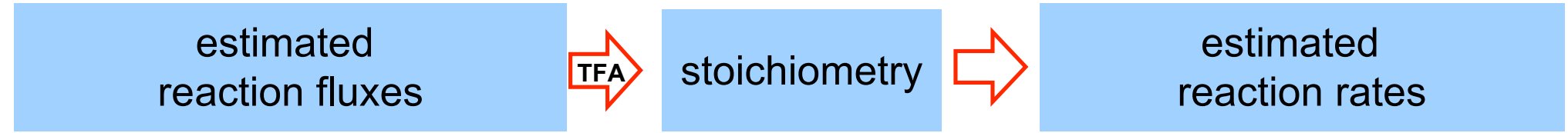
- BLS problem

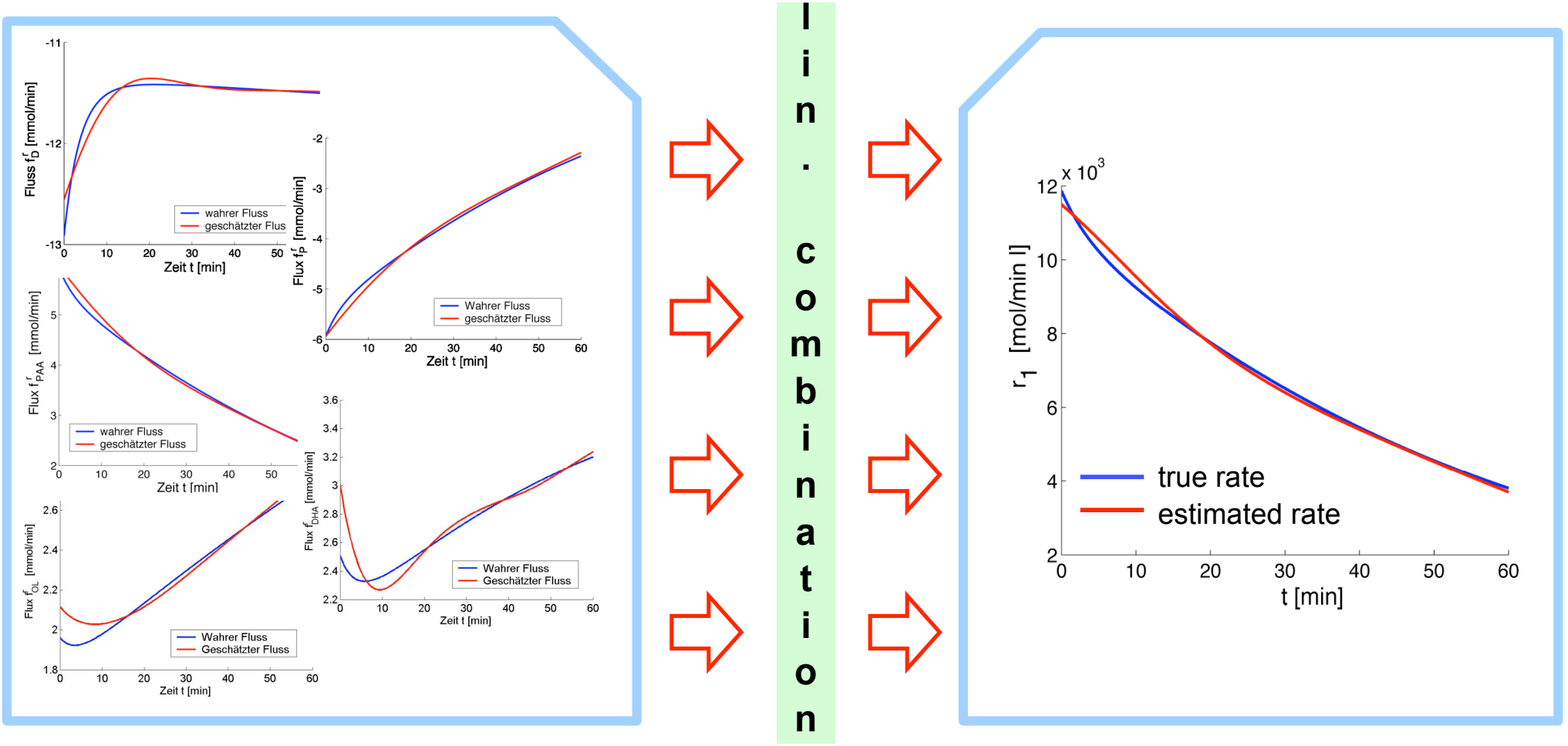
$$\mathbf{r} = \arg\min \left\| \mathbf{N}\mathbf{r} - \frac{1}{V}\mathbf{f}^r \right\|$$

$$\text{s.t.} \quad \mathbf{r}_l \le \mathbf{r} \le \mathbf{r}_u$$

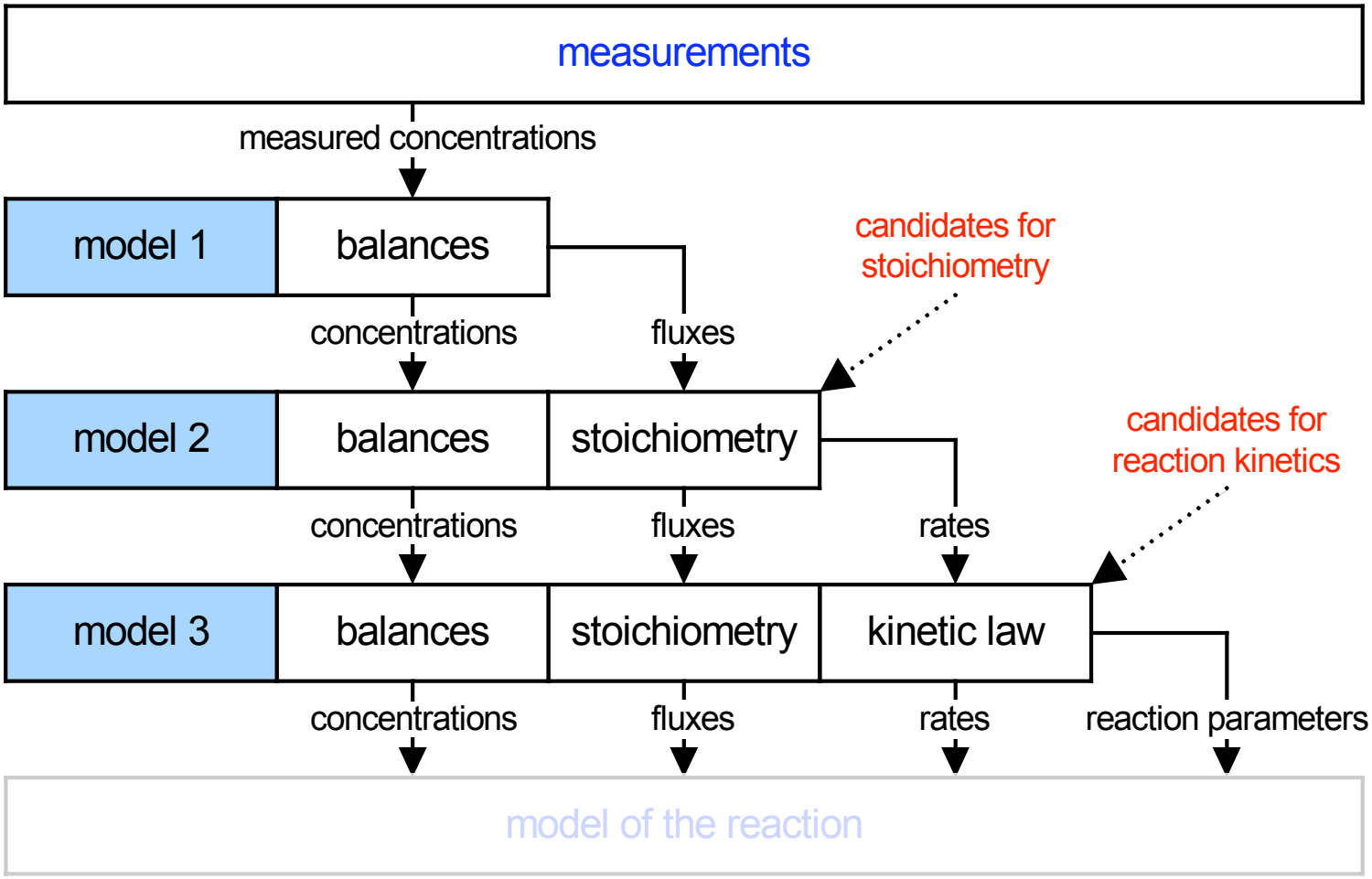
- $\mathbf{f}^r$: reaction fluxes
- $\mathbf{r}$: reaction rates
- $\mathbf{N}$: stoichiometric matrix
- V: volume

Reaction Fluxes → Stoichiometry, Reaction Rates





Incremental Model Identification



Structure Identification of Reaction Rate Function

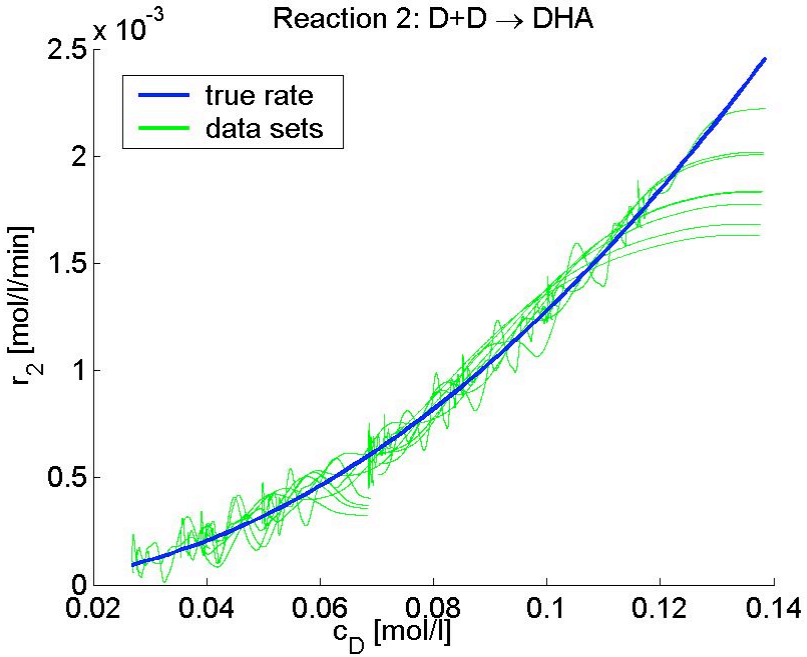
correlation of concentrations and rates

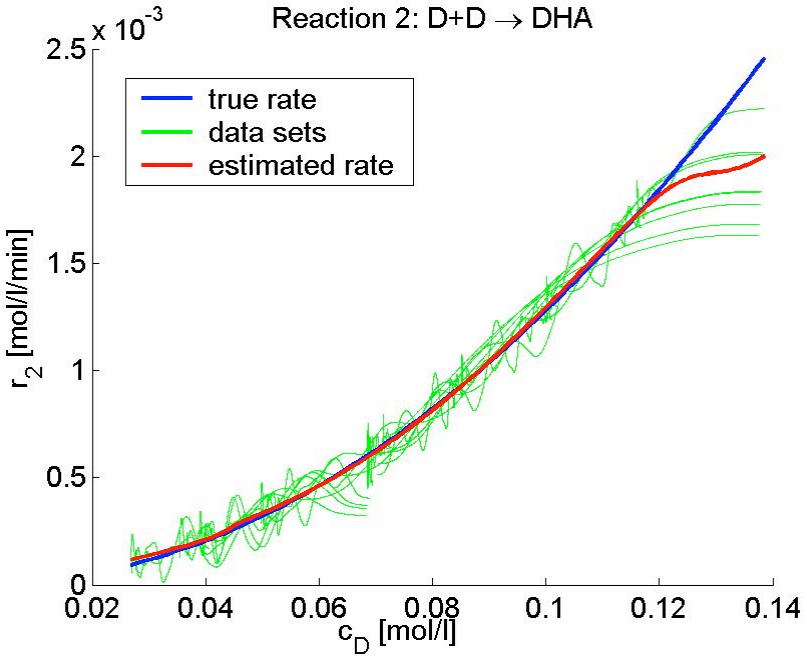
$$\hat{c}(t) \rightarrow \hat{r}(c)$$
$$\hat{r}(t) \rightarrow \hat{r}(c)$$

- band of noisy data sets
- physical insight

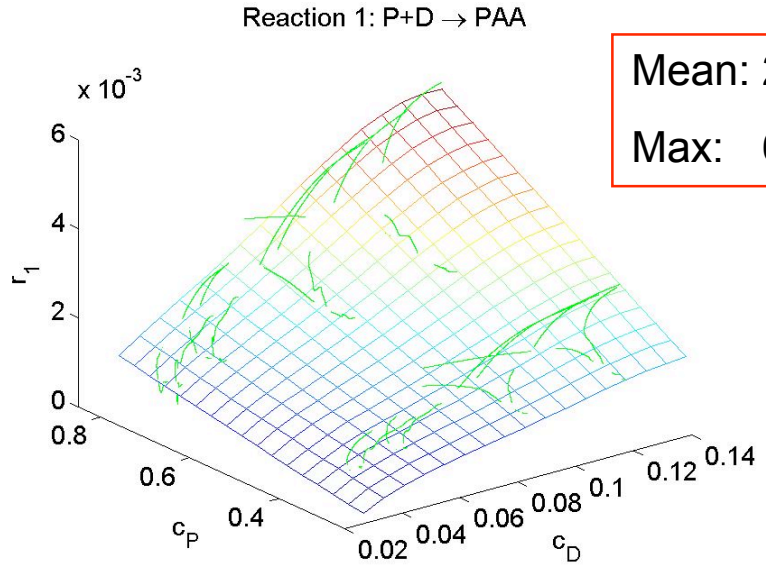
correlation with data-driven methods

- NN with Bayesian regularization



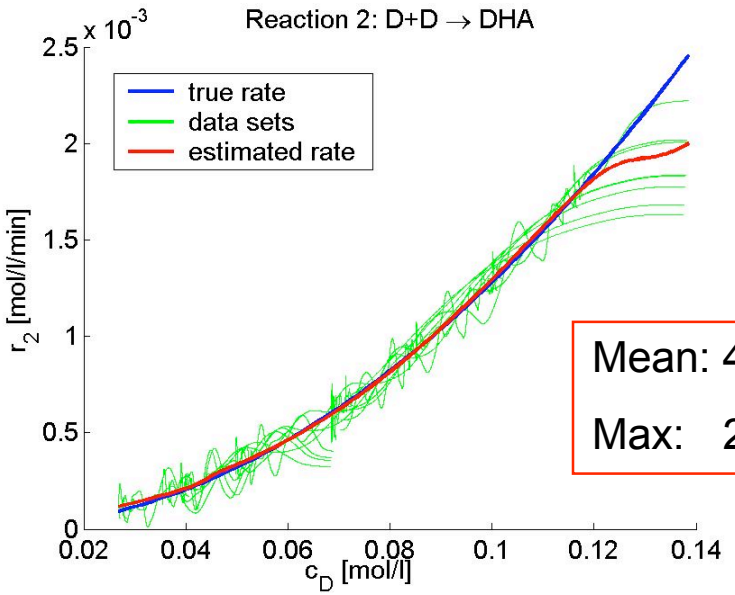


Estimated Reaction Rate Functions



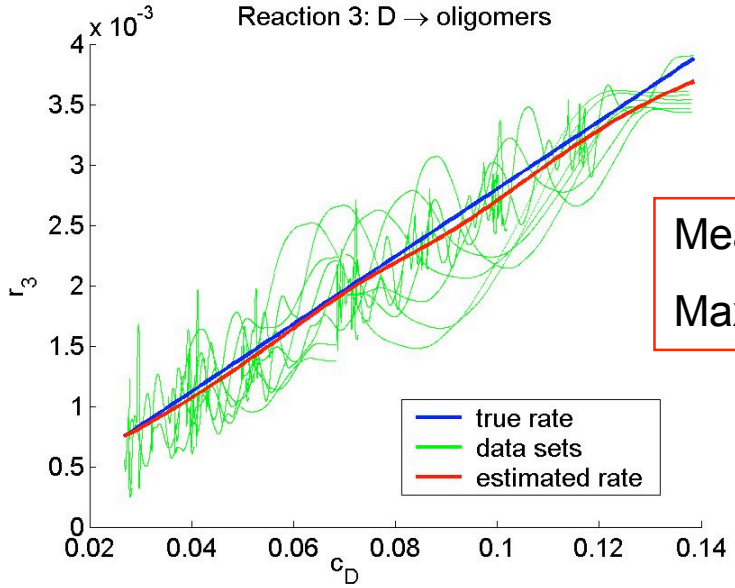
Mean: 2.7%

Max: 6%



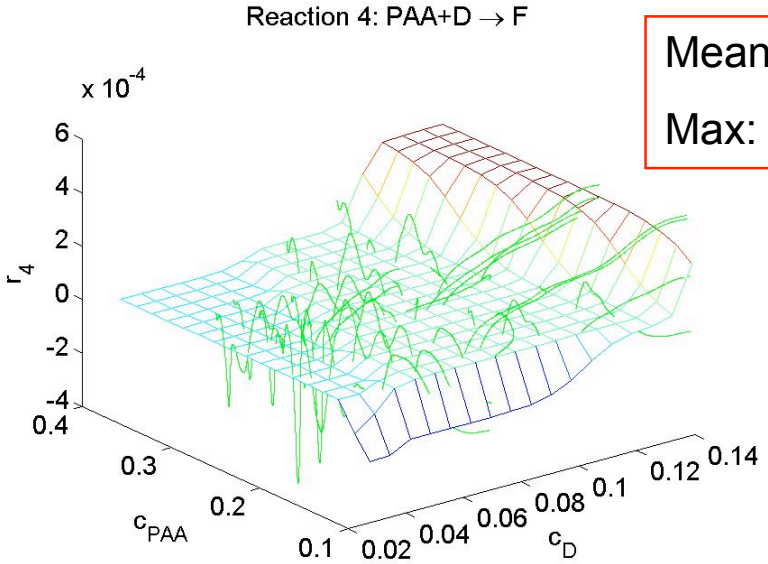
Mean: 4.2%

Max: 25%



Mean: 2.9%

Max: 5%



Mean: 167%

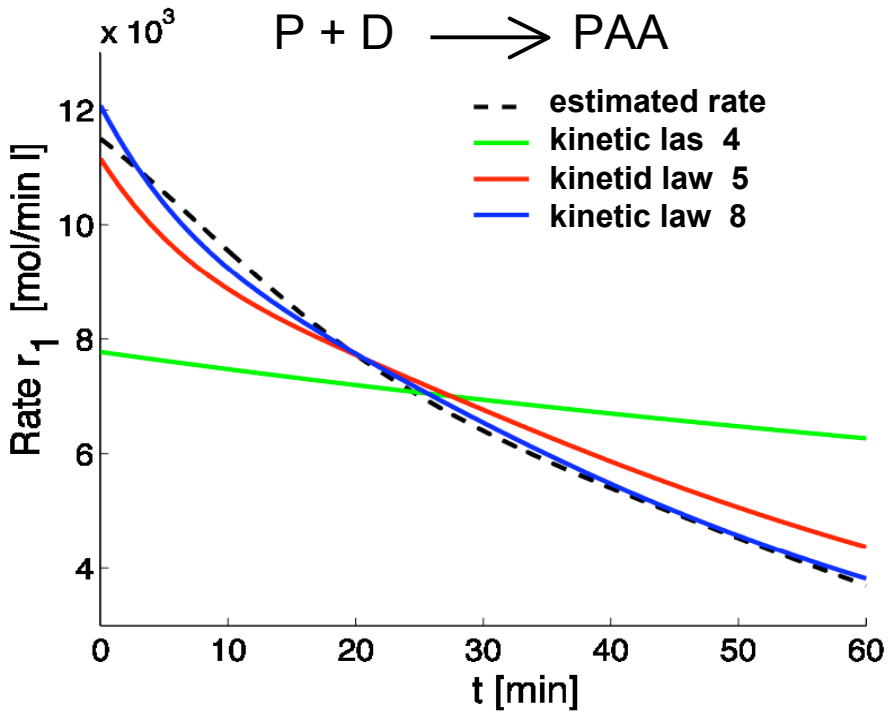
Max: 2958%

Identification of Reaction Kinetic Laws

regression of estimated rates and concentrationens for each individual reaction

$r(t)$ → $r(k,c)$

$c(t)$ → $r(k,c)$



candidate kinetic laws

$r_1^{(1)} = k_1^{(1)}$

$r_1^{(2)} = k_1^{(2)} c_D$

$r_1^{(3)} = k_1^{(3)} c_P$

$r_1^{(4)} = k_1^{(4)} c_K$

$r_1^{(5)} = k_1^{(5)} c_D c_P$

$r_1^{(6)} = k_1^{(6)} c_P c_K$

$r_1^{(7)} = k_1^{(7)} c_D c_K$

$r_1^{(8)} = k_1^{(8)} c_D c_P c_K$

$r_1^{(9)} = k_1^{(9)} c_D c_P^2$

$r_1^{(10)} = k_1^{(10)} c_D^2 c_P$

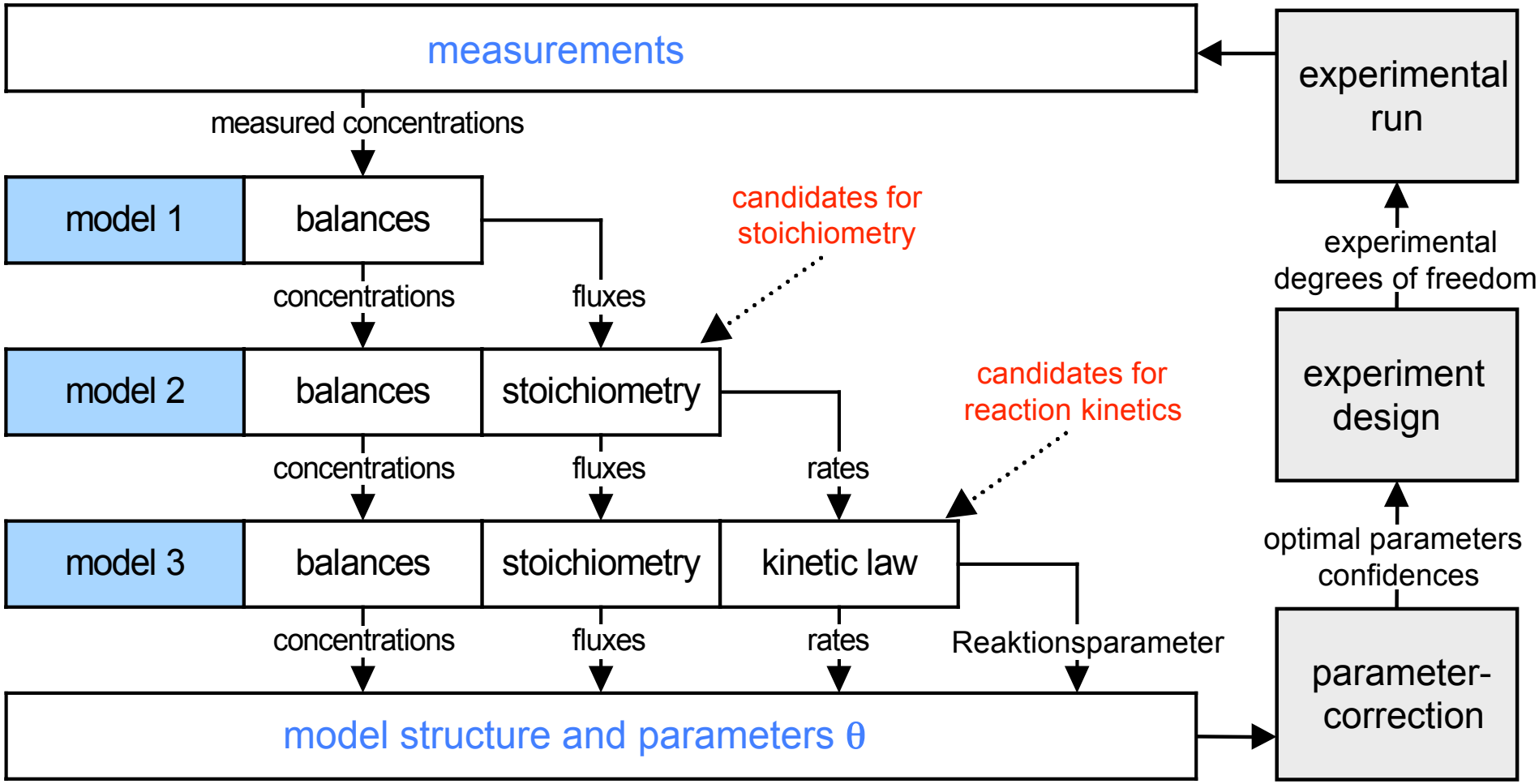
?

simultaneous identification for promising model candidates

⇒

best model structure with statistically optimal parameters

Iterative Model Improvement

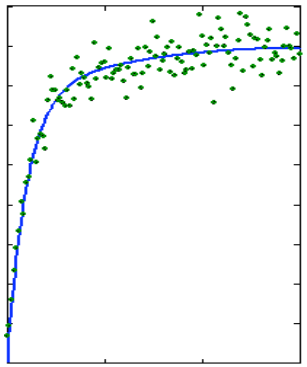
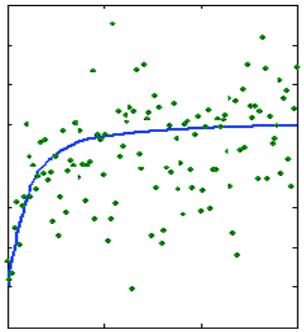


Iterative Improvement - Example

scenario: 3600 data points in each experiment (T_s=1 s), 5% noise

	stoichiometry	kinetic law	validation	parameters
1	1: P + D → PAA 2: D + D → DHA 3: D → OL	$r_1 = k_1 c_D c_P c_K$ $r_2 = k_2 c_D$ $r_3 = k_3$	–	N/A
⇩	experimental design for model discrimination			
2	1: P + D → PAA 2: D + D → DHA 3: D → OL	$r_1 = k_1 c_D c_P c_K$ $r_2 = k_2 c_D^2 c_K$ $r_3 = k_3 c_D$	✔	Parametersatz k_1, k_2, k_3
⇩	experimental design for best model parameters			
3	1: P + D → PAA 2: D + D → DHA 3: D → OL	$r_1 = k_1 c_D c_P c_K$ $r_2 = k_2 c_D^2 c_K$ $r_3 = k_3 c_D$	✔	Verbesserter Parametersatz k_1, k_2, k_3
STOP	requested accuracy achieved			

Incremental vs. Simultaneous Model Identification

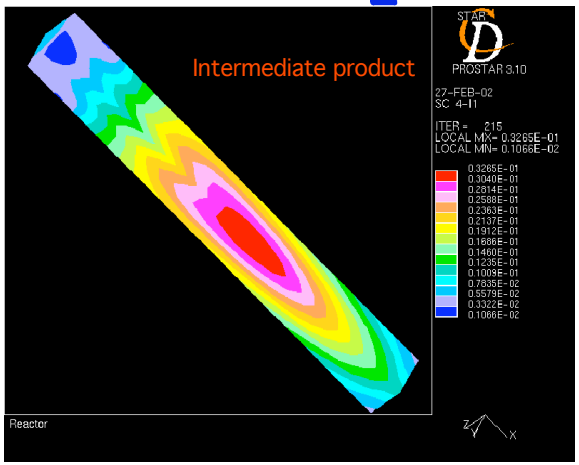
	Incremental Method				Simultaneous Method			
	Reso-lution	# Models	CPU Time	Ident. Corr.	Reso-lution	# Models	CPU Time	Ident. Corr.
STDV = 2% 	3 sec	1	30 min	100 %	3 sec	3600	1.7 d	100 %
	30 sec	≈ 1	15 sec	100 %	30 sec	3600	3.8 h	100 %
	5 min	≈ 7	6 sec	100 %	5 min	3600	1.7 h	100 %
STDV = 10% 	3 sec	≈ 100	40 min	100 %	3 sec	3600	1.6 d	100 %
	30 sec	≈ 160	3 min	50 %	30 sec	3600	3.9 h	50 %
	5 min	≈ 200	2 min	10 %	5 min	3600	1.1 h	10 %

60 min

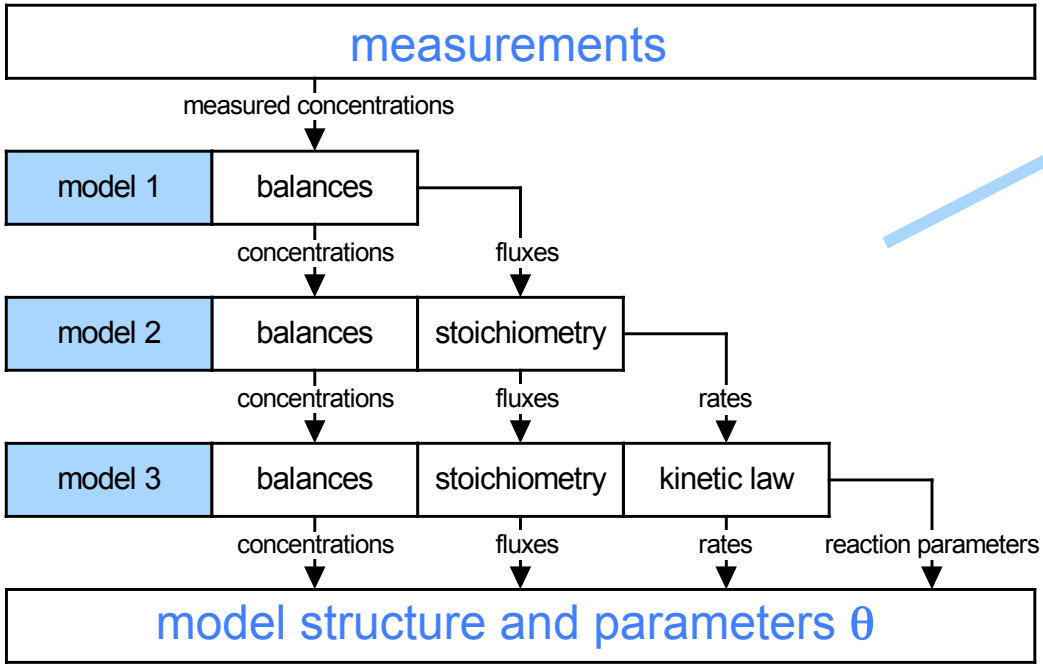
much faster identical

Why studying diffusion ?

- detrimental for product and process design
- very high experimental effort
- very few multi-component diffusion data available
- validity of diffusion models still a matter of debate

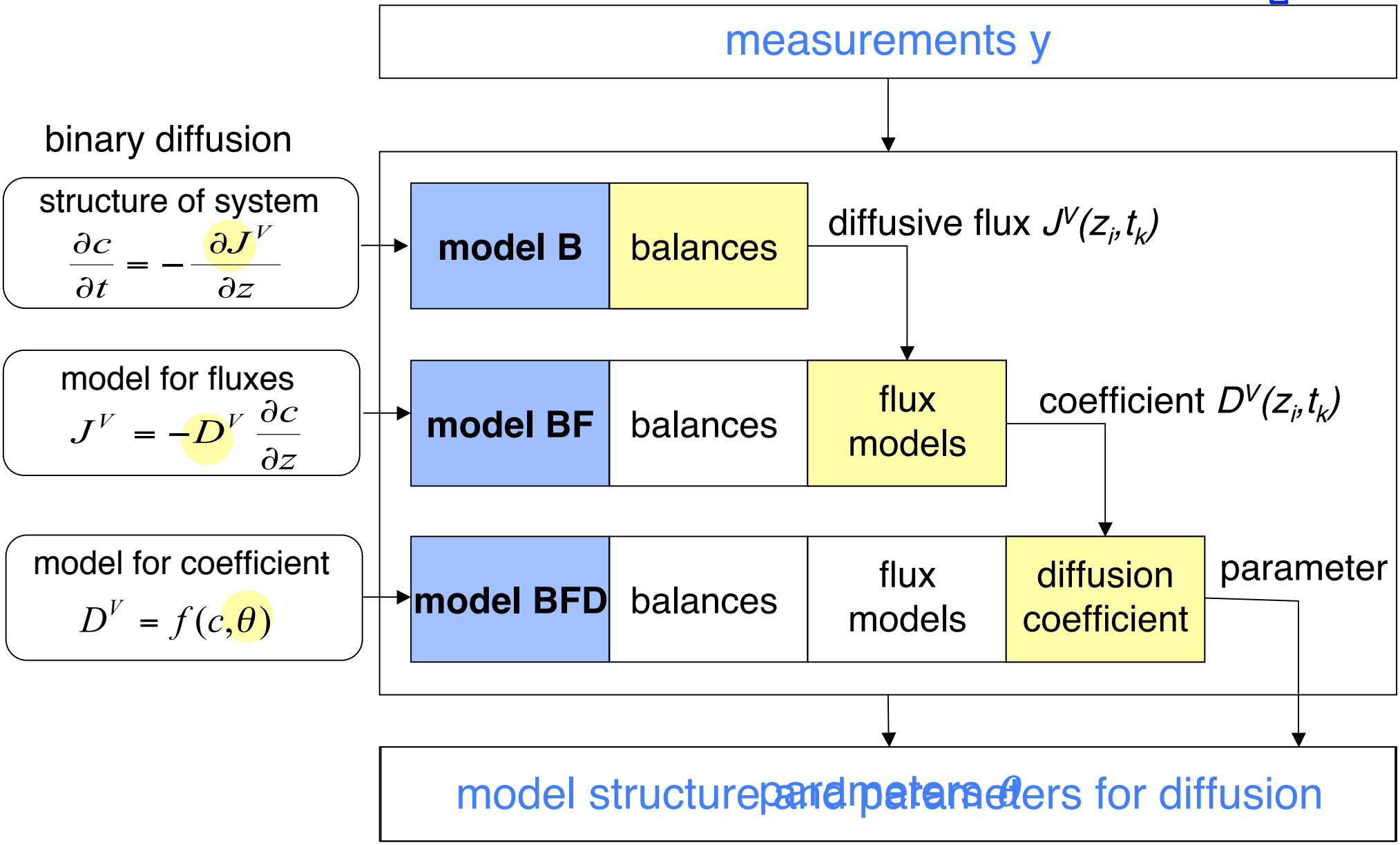


selectivity of heterogeneously catalyzed reactions (Pantelides & Urban, 2004)

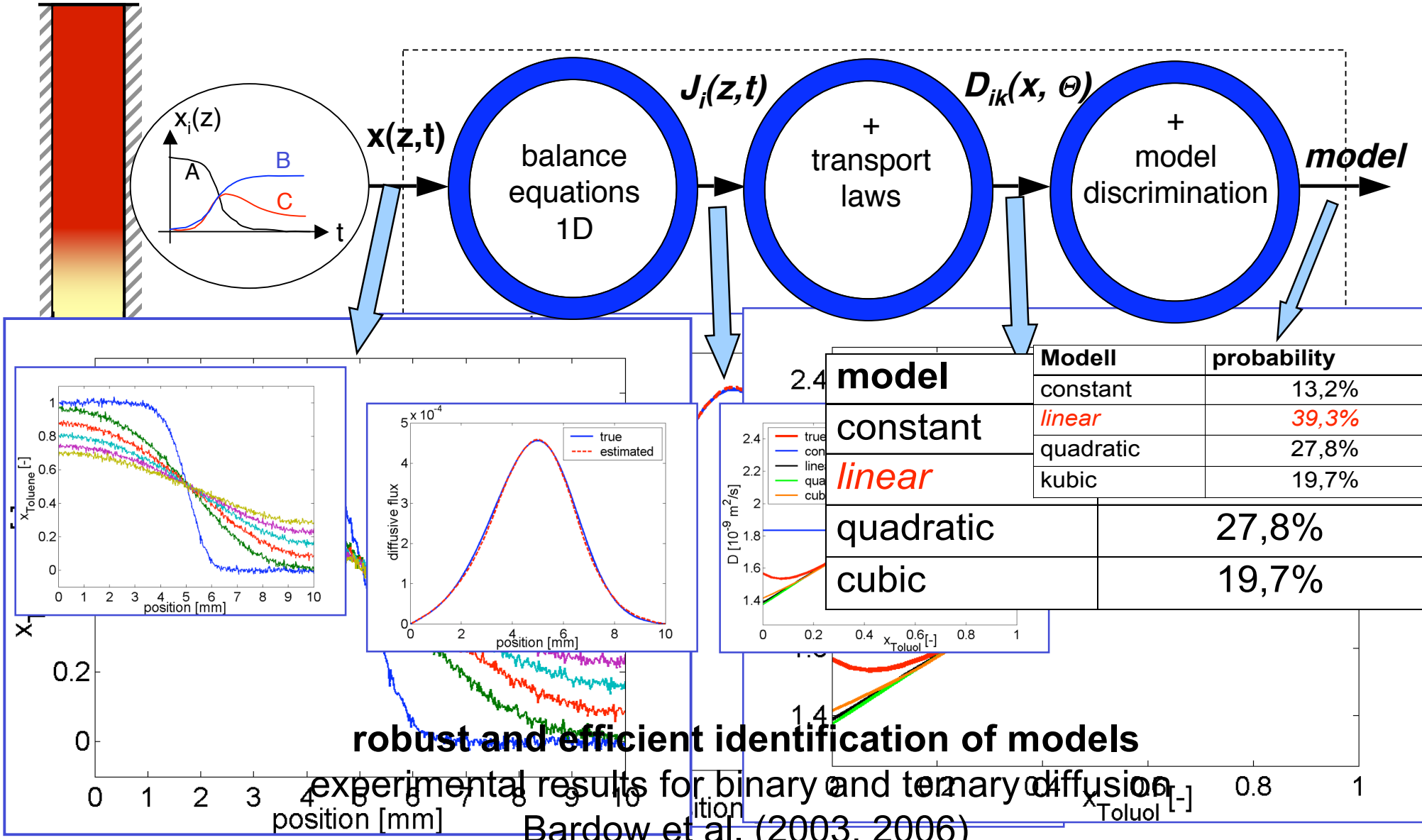


- a good model problem for the development of MEXA methodology

Incremental Model Identification



Incremental Model Identification



model	Modell	probability
constant	constant	13,2%
linear	*linear*	*39,3%*
quadratic	quadratic	27,8%
cubic	kubic	19,7%

robust and efficient identification of models

experimental results for binary and ternary diffusion

Bardow et al. (2003, 2006)

Ternary Mixtures

model B	balances

$$\frac{\partial c_i}{\partial t} = -\frac{\partial J_i^V}{\partial z}; i = 1,2$$

→ problem decouples
→ solve binary problem twice

model BF	balances	flux models

$$J_1^V = -D_{11}^V \frac{\partial c_1}{\partial z} - D_{12}^V \frac{\partial c_2}{\partial z}$$

→ coefficients not identifiable

→ error-in-variables regression

model BFD	balances	flux models	diffusion coefficient

Example: simulated experiment as in Arnold & Toor (1967), noise level σ=0.01

Coefficient	D_{11}	D_{12}	D_{21}	D_{22}
Error [%]	-9.0	-33.4	-10.8	-10.4

estimation of constant diffusion coefficients from a single experiment with good precision

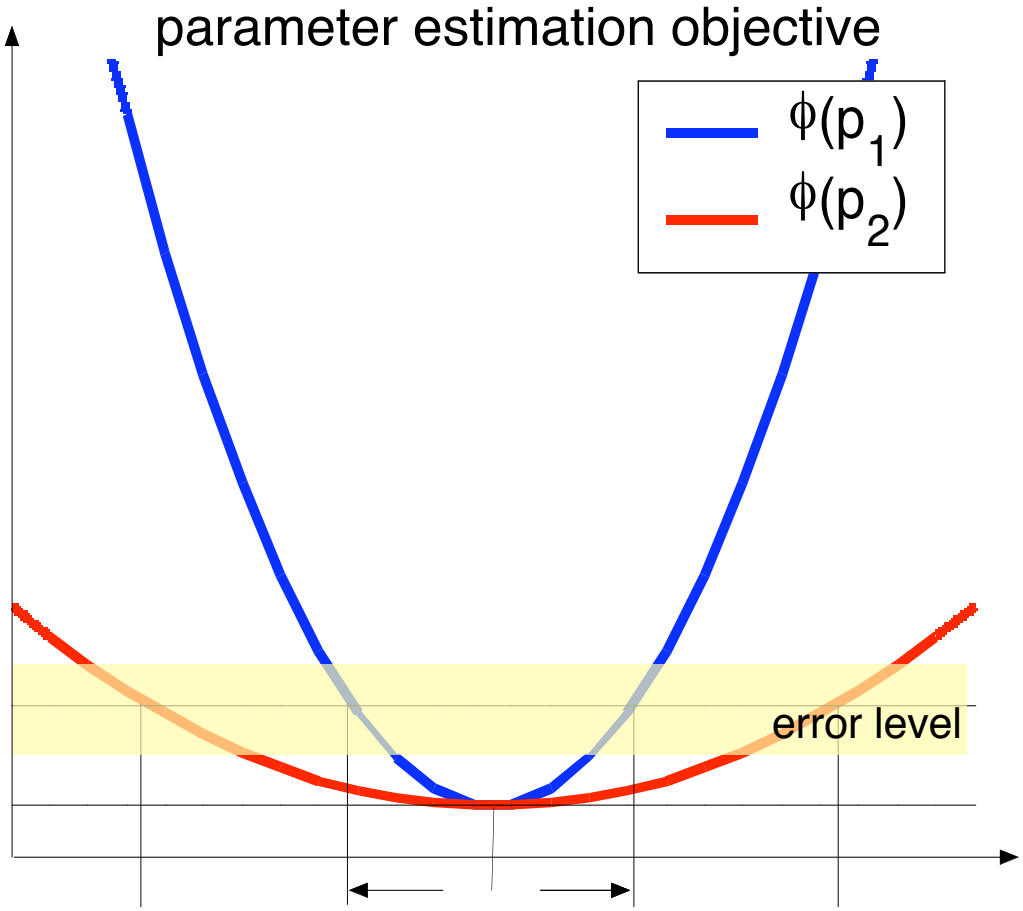
Optimal Experimental Design: Idea

set free variables ***p*** such that information on model parameters is maximized

=

maximize curvature of parameter estimation objective

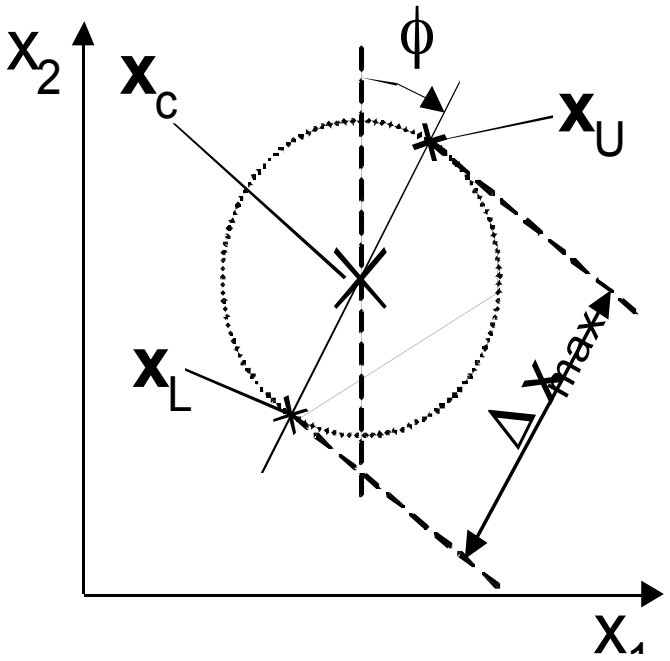
$$\max_{p} \frac{\partial^2 \phi}{\partial \theta^2}$$

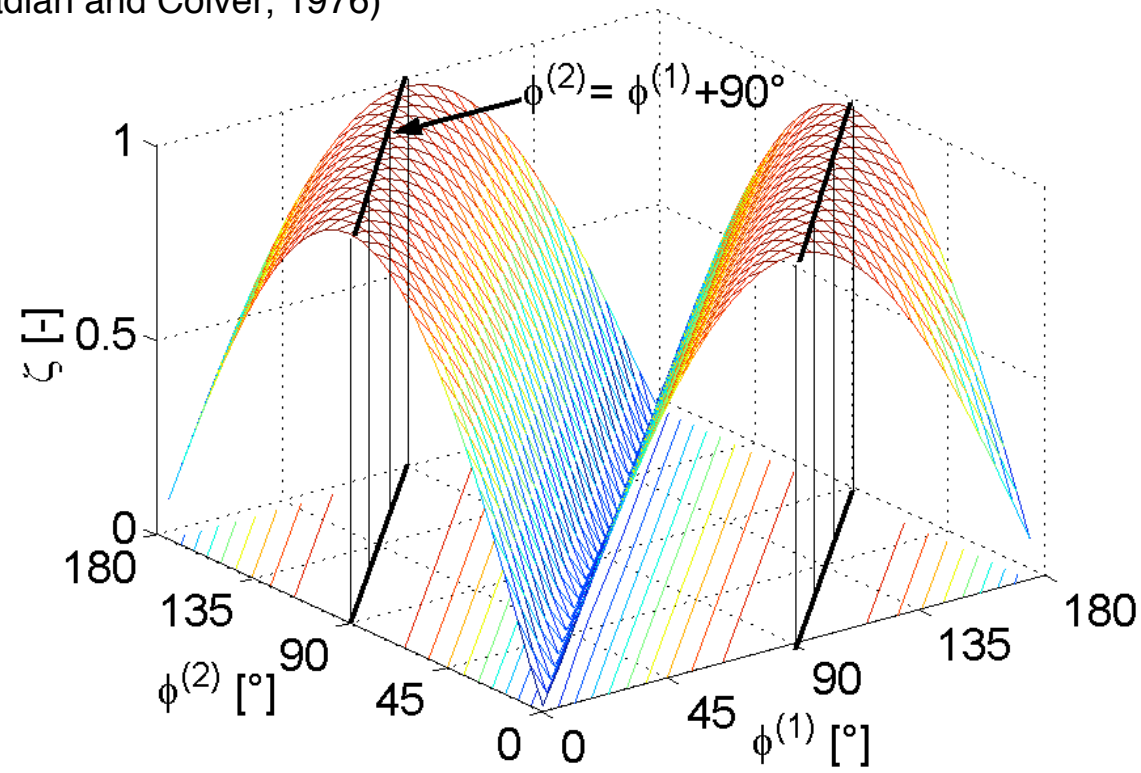


Optimal Ternary Mixture Composition

Example: acetone-benzene-methanol at $\mathbf{x}_c = [0.35; 0.302; 0.348]^T$,

(Alimadadian and Colver, 1976)



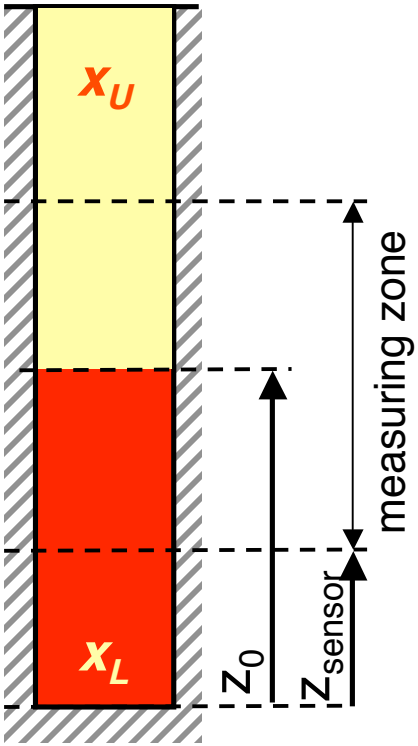


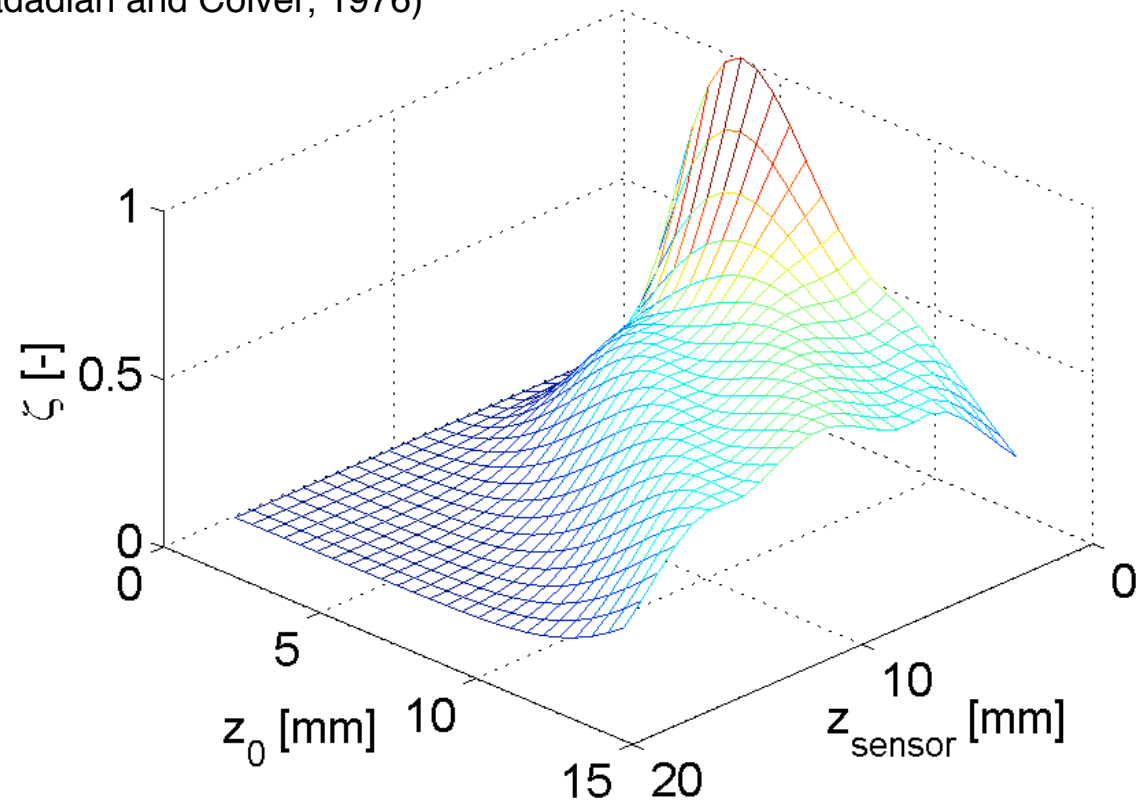
- scaled objective ζ-efficiency measures information per parameter
- one Raman experiment suffices to determine ternary Fick matrix
- two different experiments result in substantial improvement
- experiments should be as distinct as possible ($\phi^{(2)}=\phi^{(1)}+90°$)

Optimal Initial Conditions

Example: acetone-benzene-methanol at $\mathbf{x}_c = [0.35; 0.302; 0.348]^T$,

(Alimadadian and Colver, 1976)

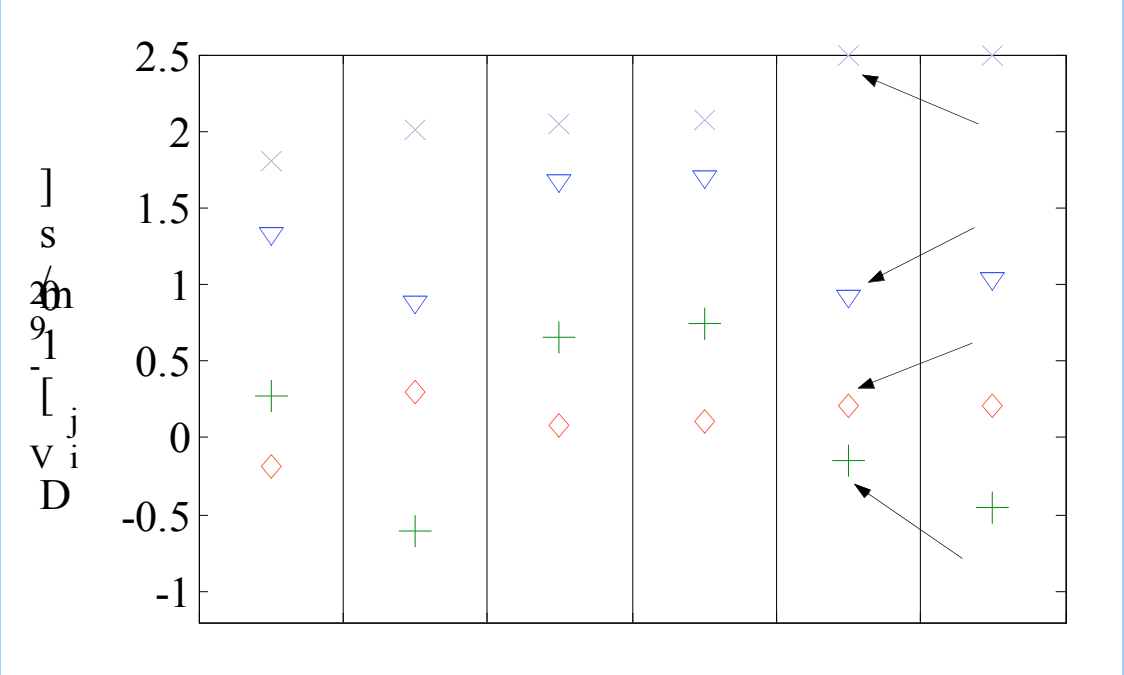




- measurements at the wall, i.e. restricted diffusion experiments
- unequal volume of both phases, almost independent of concrete mixture
- short experiments are beneficial

System: 1-Propanol - 1-Chlorobutane - n-Heptane

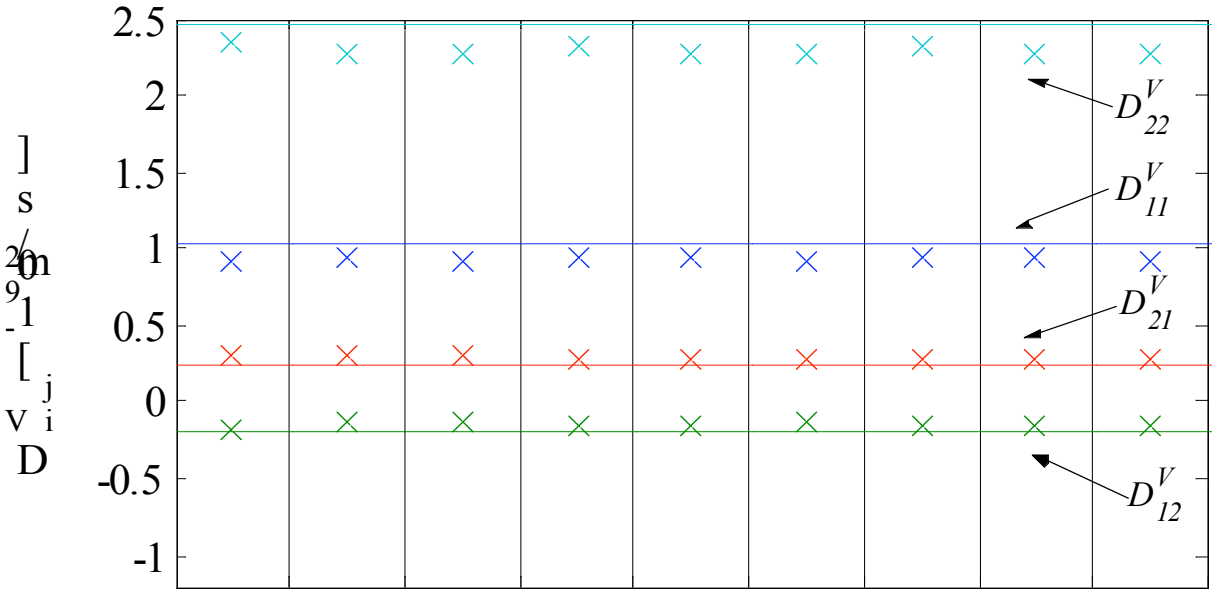
Diffusivities from a single Raman experiment



→ one Raman experiment gives full diffusion matrix
→ currently scatter in data is still significant

Ternary Results

Diffusivities from two optimal Raman experiments



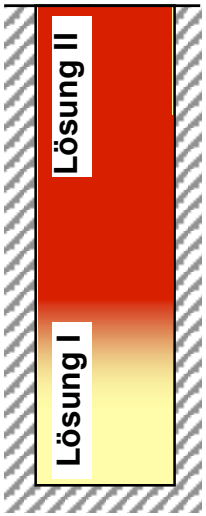
with reference data from Weingärtner et al. (1994)

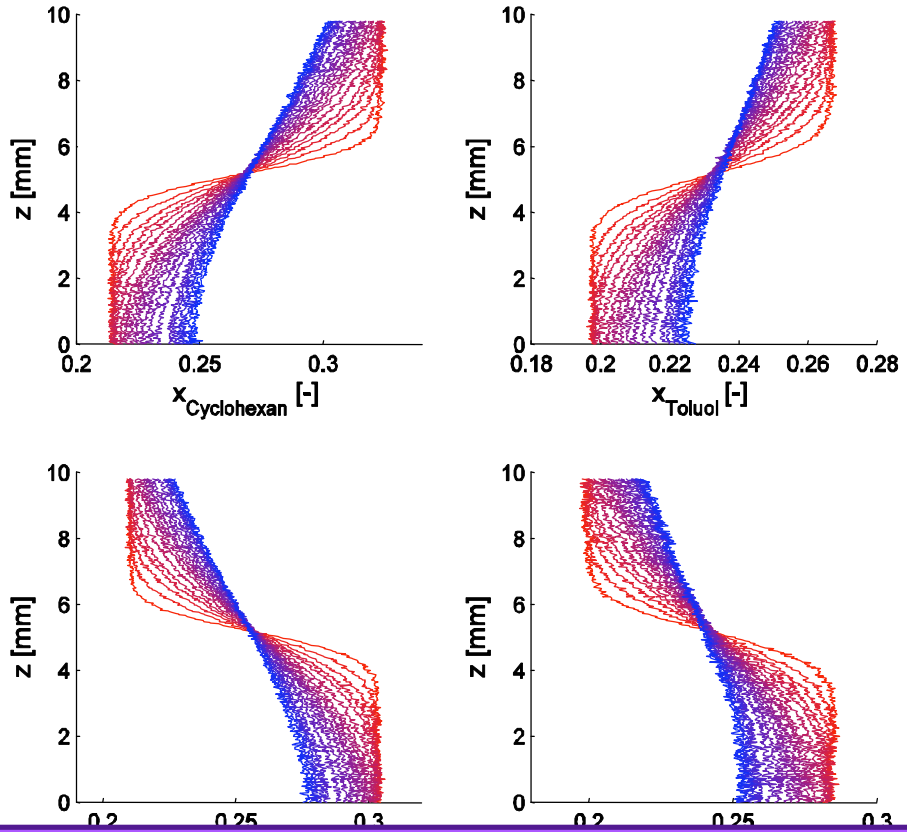
→ one Raman experiment gives full diffusion matrix
→ good precision from 2 optimized runs
→ robust & efficient measurement
→ quantitative validation of design predictions

Quaternary Diffusion

concentration measurements with Raman spectroscopy and model-based design and evaluation of diffusion experiments

(cyclohexane – toluene – dioxane – chlorobutane)





matrix of Fick's diffusion coefficients

$$D_{ij} \left[10^{-9}\, m^2/s\right]$$

$$\begin{bmatrix} 1{,}708 & 0{,}086 & 0{,}244 \\ 0{,}019 & 2{,}066 & 0{,}028 \\ 0{,}296 & 0{,}111 & 1{,}74 \end{bmatrix}$$

thermodynamical factor – accuracy improvements – electrolytes

Benefits of MEXA for Diffusion Problem

high resolution measurements

model-based methods

reduces experimental effort

- ✓ fewer experiments
- ✓ maximum precision
- ✓ simple design and preparation

- experimentally validated for binary, ternary, quaternary and quinternary mixtures
- concentration dependence of diffusion coefficients

diffusion experiments for

- ✓ - multi-component mixtures
- - reactive systems
- - electrolytes

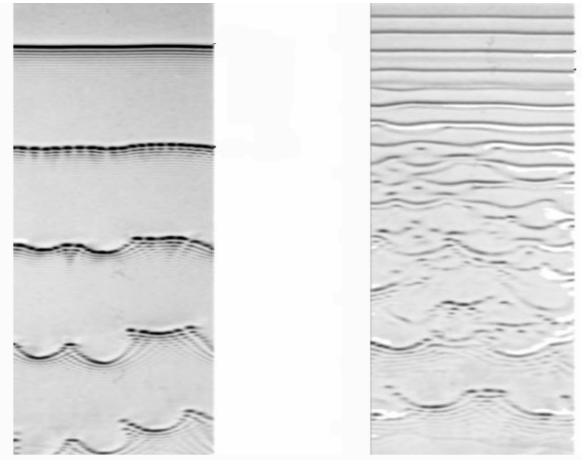
- towards reactive and electrolytes mixtures
- further improvement of method
- diffusion modeling

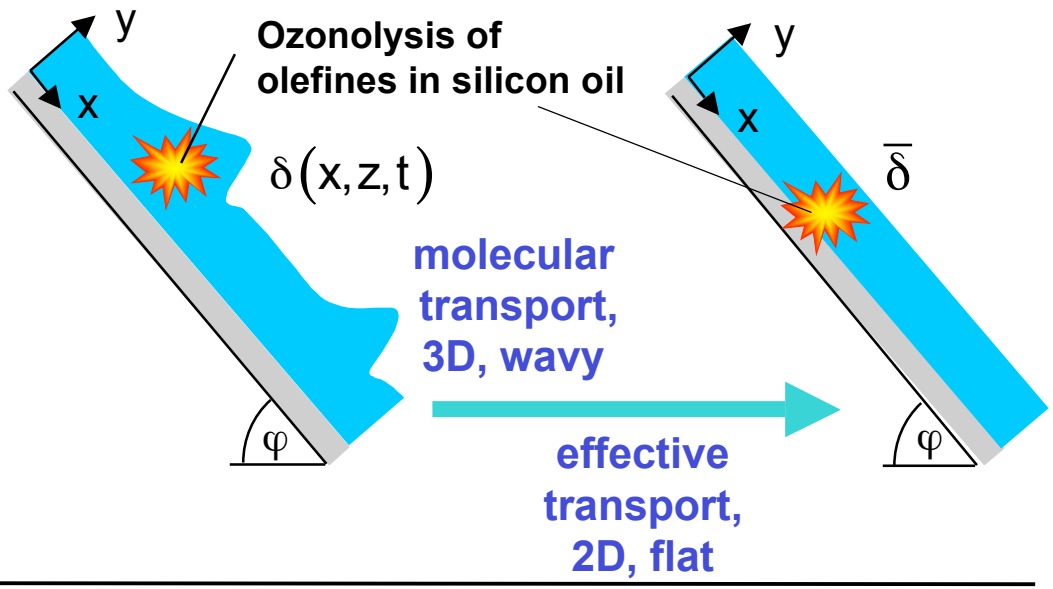
Kinetic Phenomena in Falling Film Reactors

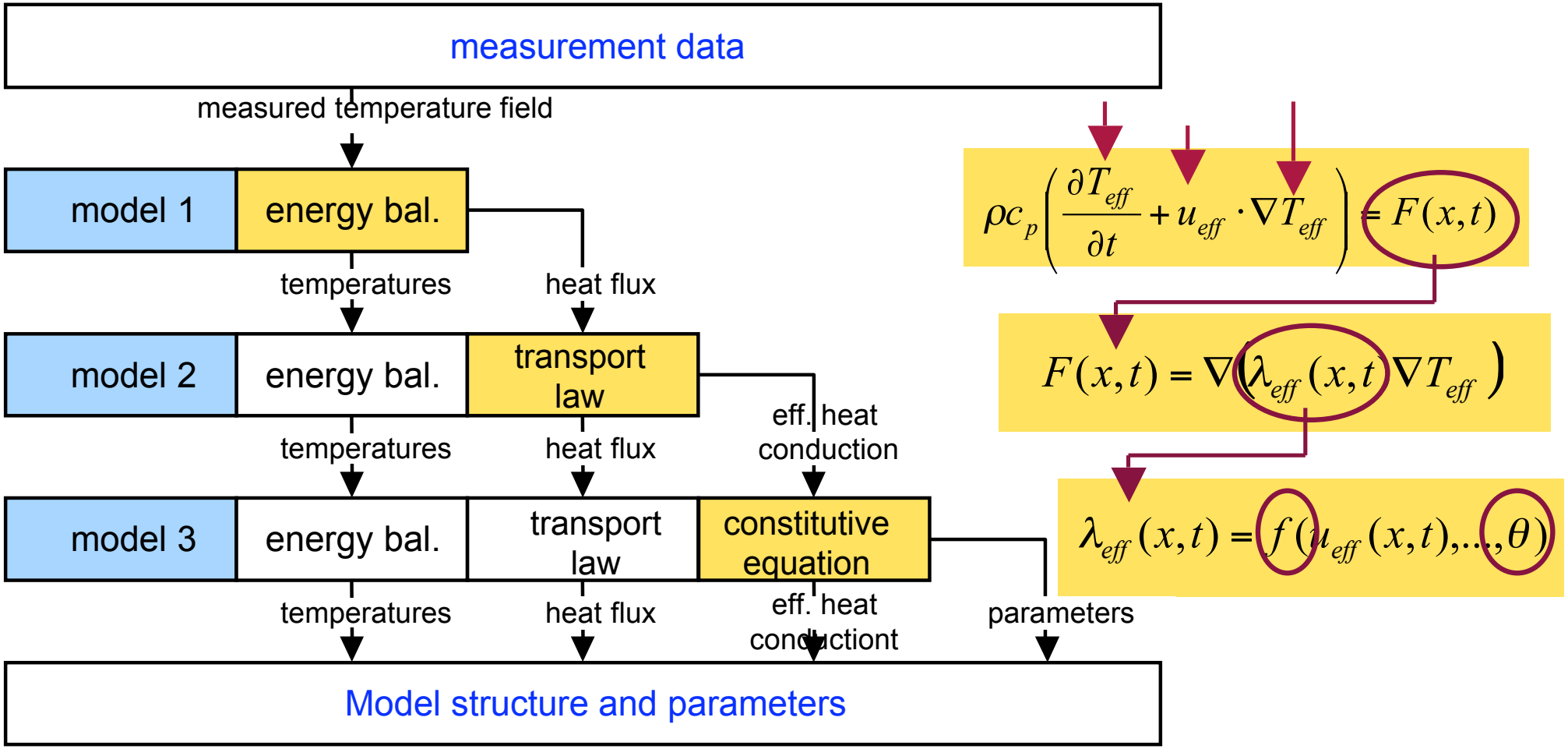
- falling films are all around:
  - falling film cooler
  - falling film evaporator
  - falling film absorber
  - falling film reactors

- transport phenomena are hardly understood, interaction between
  - fluid dynamics with free surface
  - heat and mass transfer
  - chemical reaction

- first milestone: modelling of heat transfer with effective transport coefficients

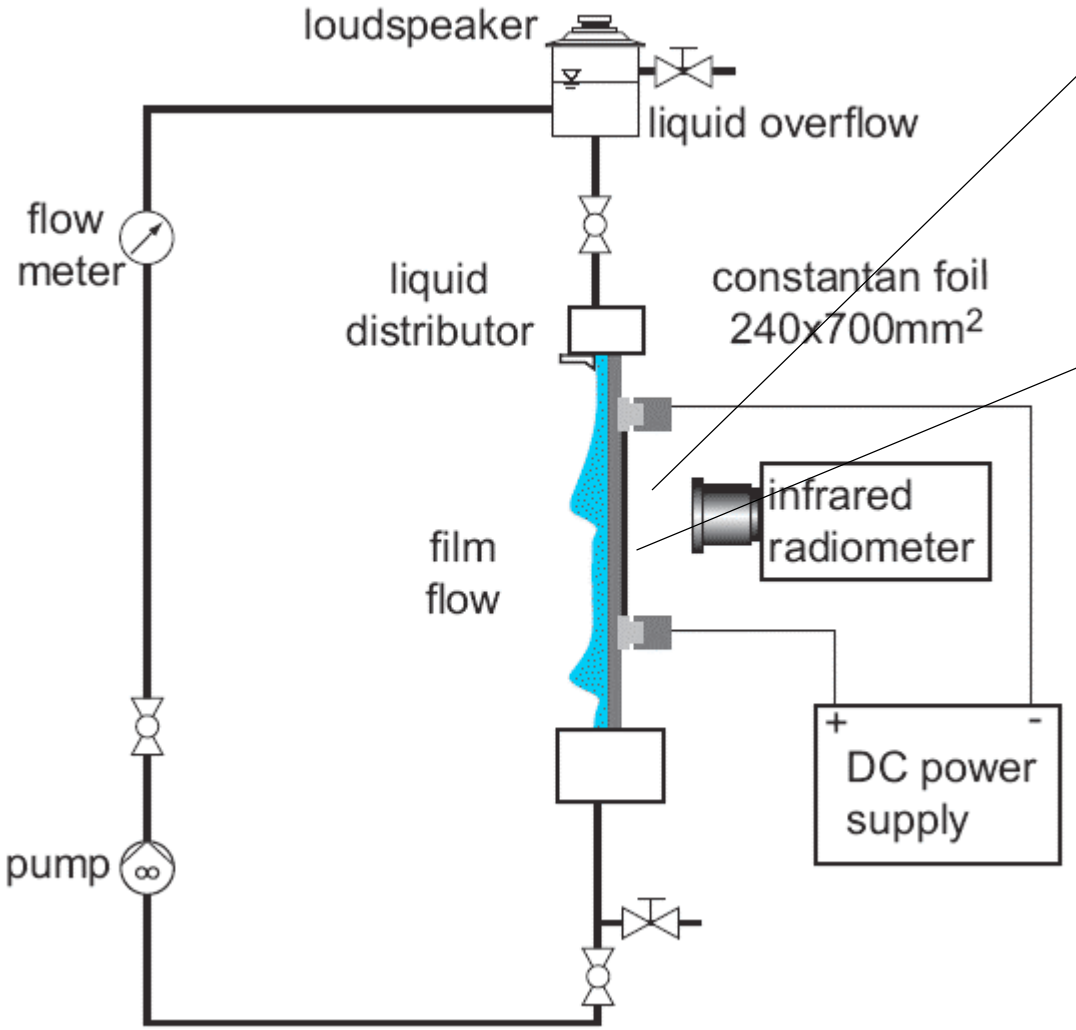


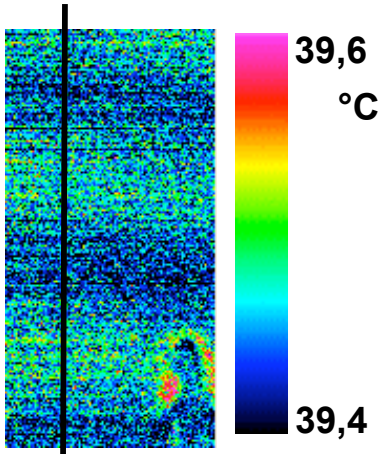



measurement data
measured temperature field
model 1
energy bal.
temperatures
heat flux
model 2
energy bal.
transport law
temperatures
heat flux
eff. heat conduction
model 3
energy bal.
transport law
constitutive equation
temperatures
heat flux
eff. heat conductiont
parameters
Model structure and parameters
$\rho c_p \left(\frac{\partial T_{eff}}{\partial t} + u_{eff} \cdot \nabla T_{eff} \right) = F(x,t)$
$F(x,t) = \nabla \left(\lambda_{eff}(x,t) \nabla T_{eff} \right)$
$\lambda_{eff}(x,t) = f(u_{eff}(x,t),..,\theta)$

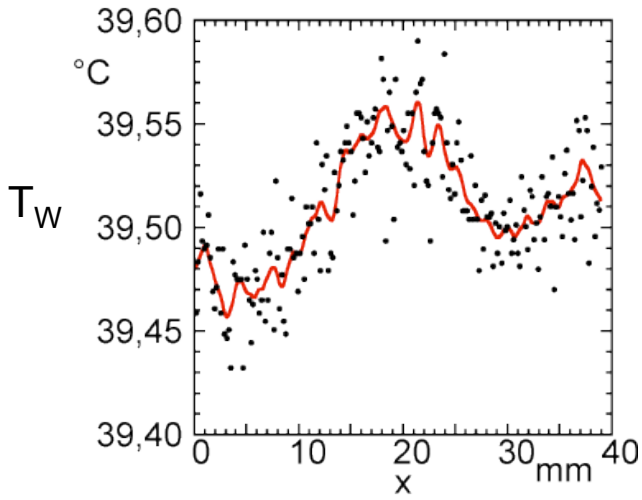
Experimental Set-up

eperiments:
F. Al-Sibai, A. Leefken, R. Kneer, U. Renz, SFB 540





wall temperature measurements



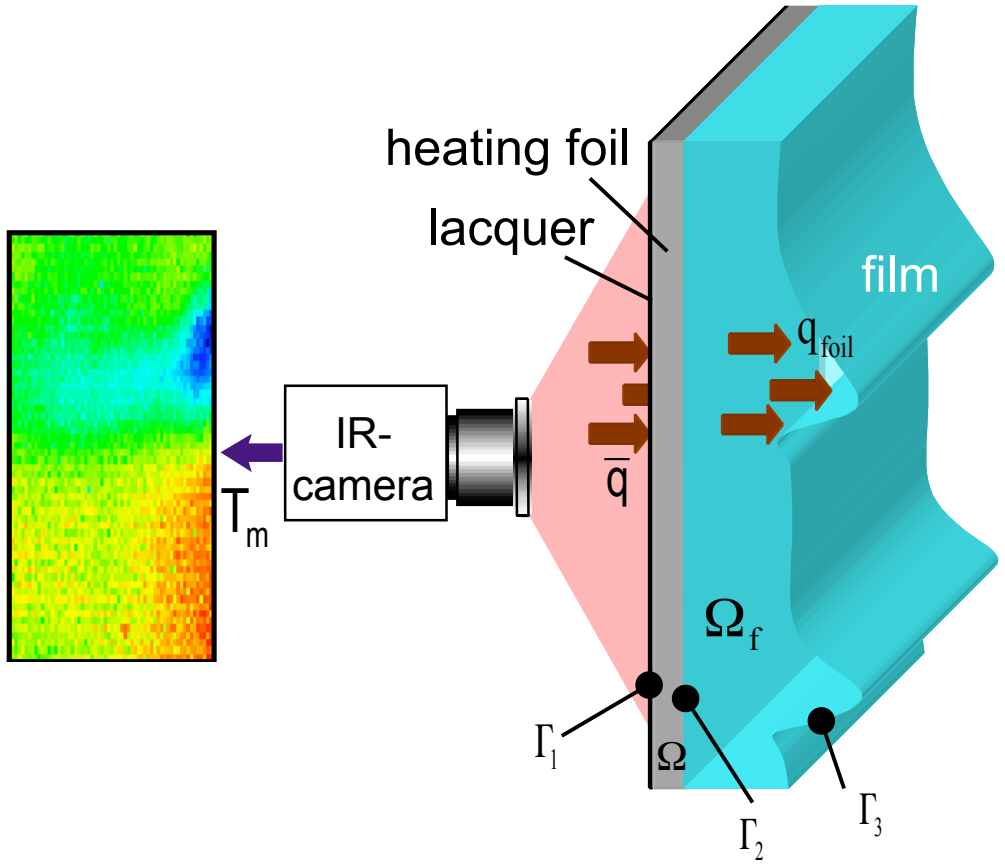
Mathematical Problem Formulation

Inverse heat conduction problem as a minimization problem:

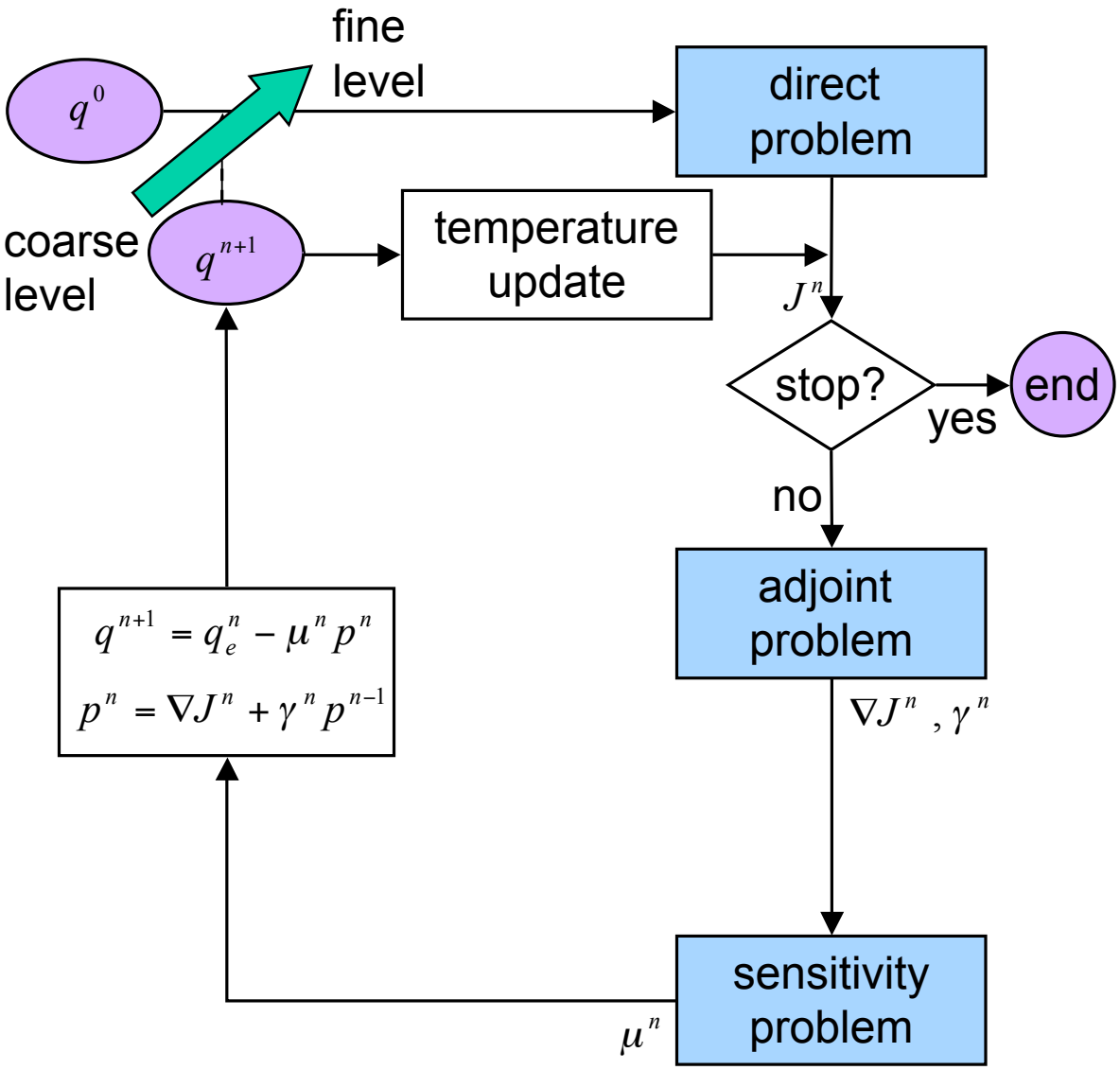
$$J(q_{foil}) := \int_0^{t_f} \int_{\Gamma_1} \left[T(q_{foil}, x, t) - T_m(x, t)\right]^2 dx\, dt + R(q_{foil}) \;\rightarrow \min$$

s. t. T satisfies the **direct heat problem**:

$$\begin{aligned}
\frac{\partial T}{\partial t}(x,t) &= a\Delta T(x,t) \\
T(x,0) &= T_0(x) \\
-\lambda\frac{\partial T}{\partial n}(x,t) &= \bar{q}(x,t) \quad \text{on } \Gamma_1 \\
-\lambda\frac{\partial T}{\partial n}(x,t) &= q_{foil}(x,t) \quad \text{on } \Gamma_2
\end{aligned}$$

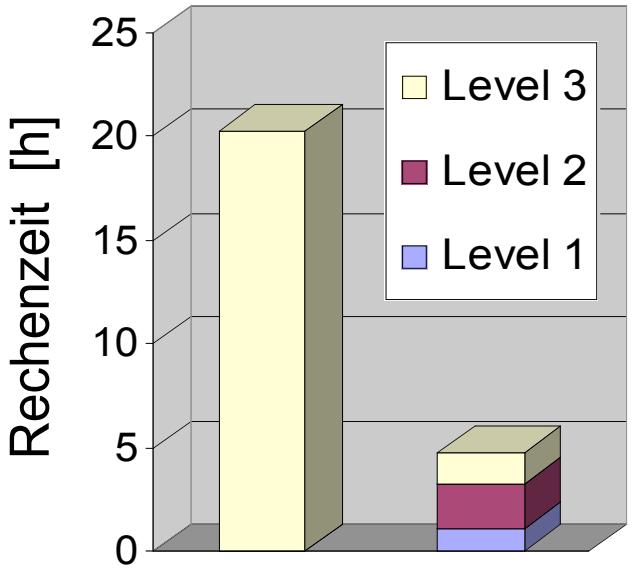


Optimization Algorithm (CG)



DROPS: adaptive FEM, 3D, anisotropic grids:
Soemers, Groß, Reichelt, Reusken, CRC 540

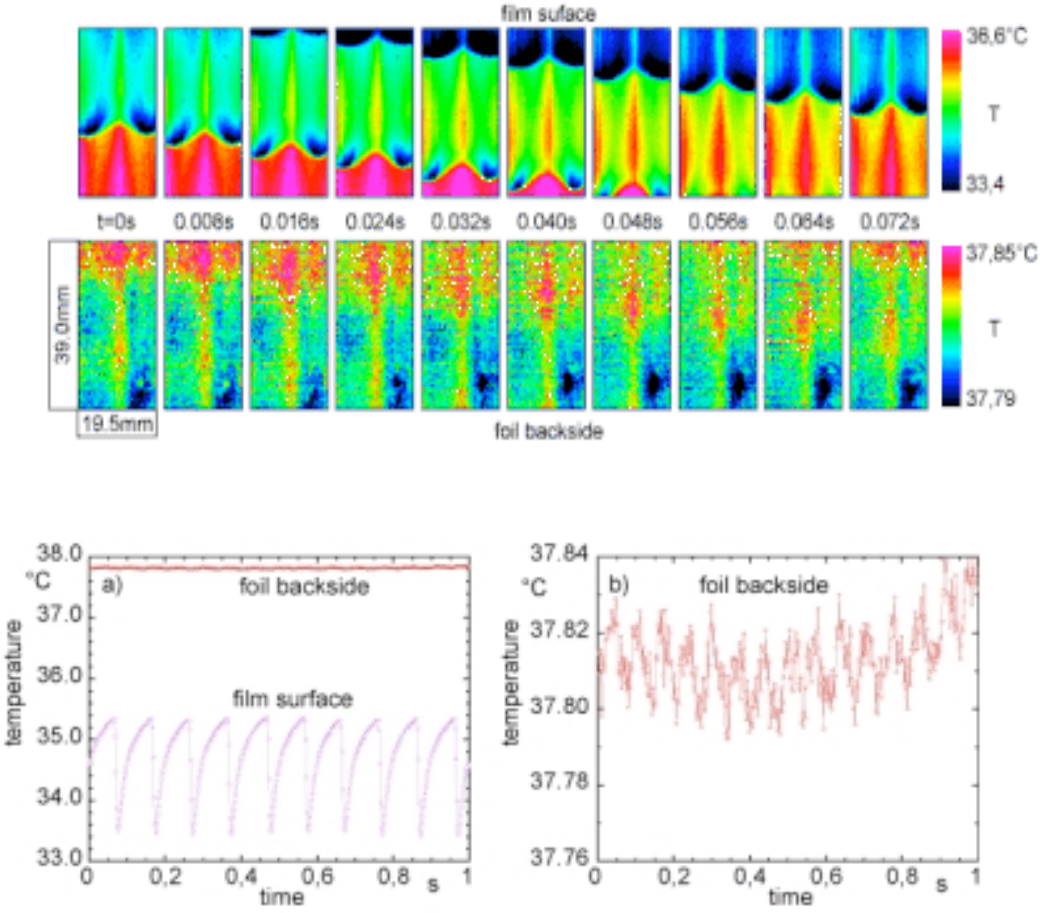
nested iteration



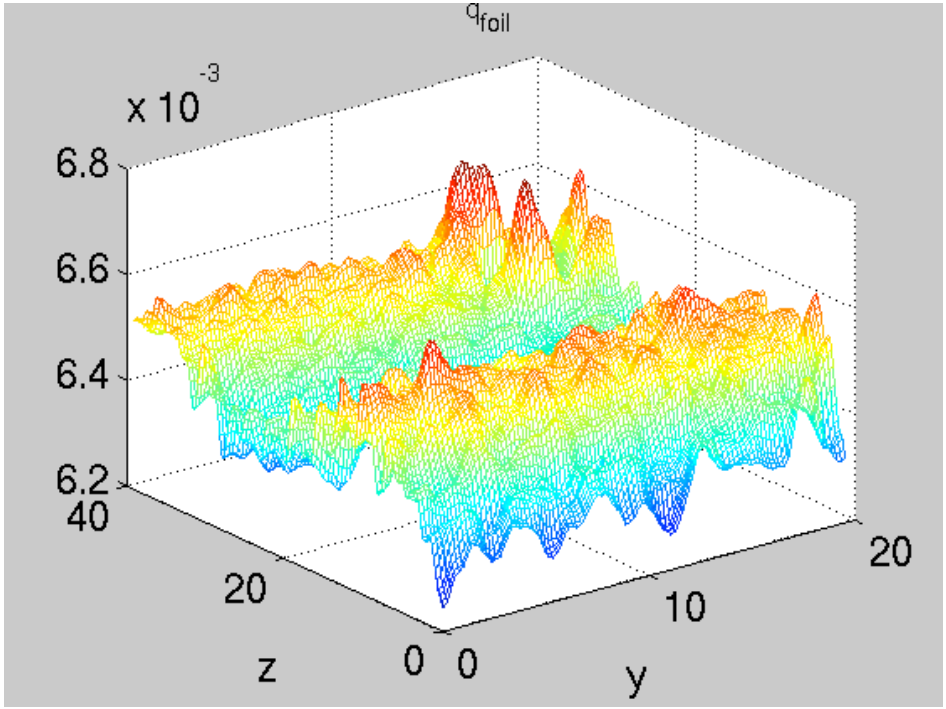
- multi-level strategy: exploit grid hierarchy
- good initial guesses from coarse grid results

Heat Flux Estimates from Experimental Data

measured temperature distributions

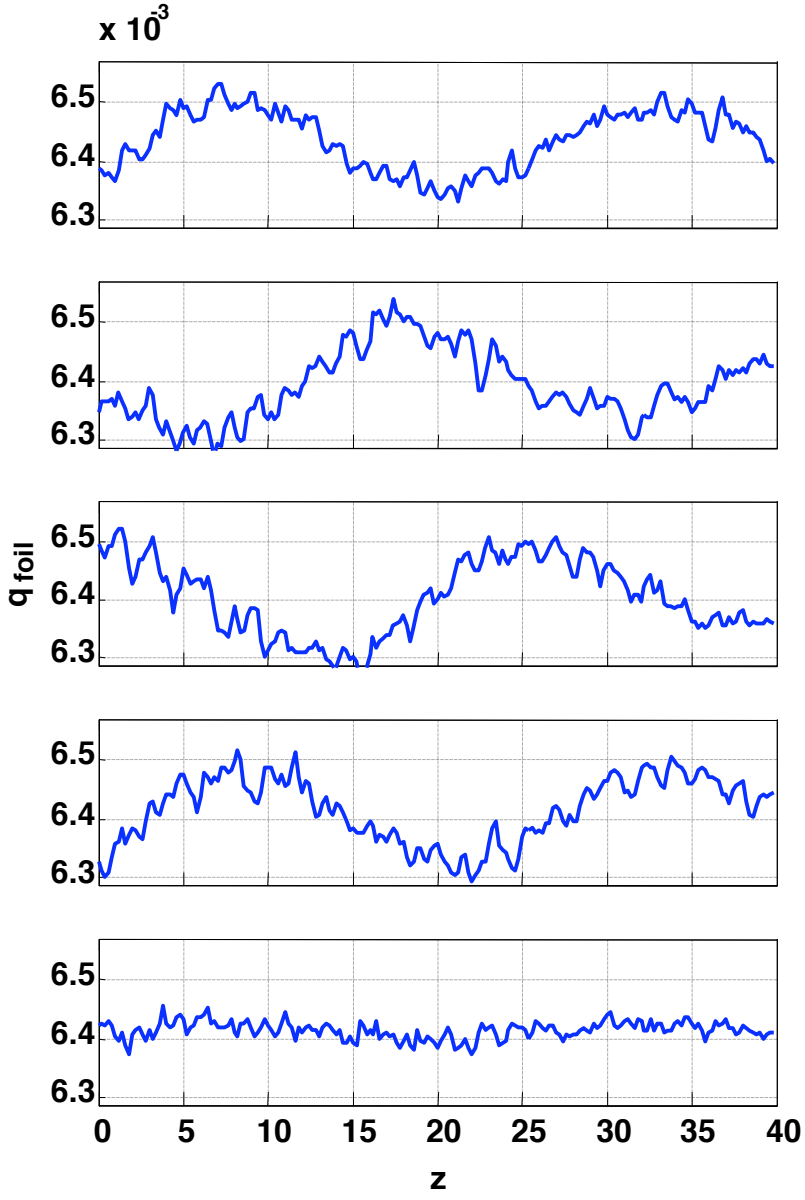


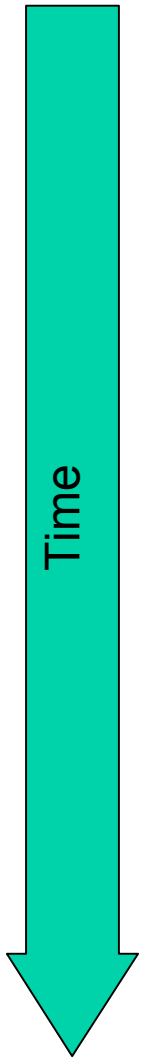
estimated heat flux distribution

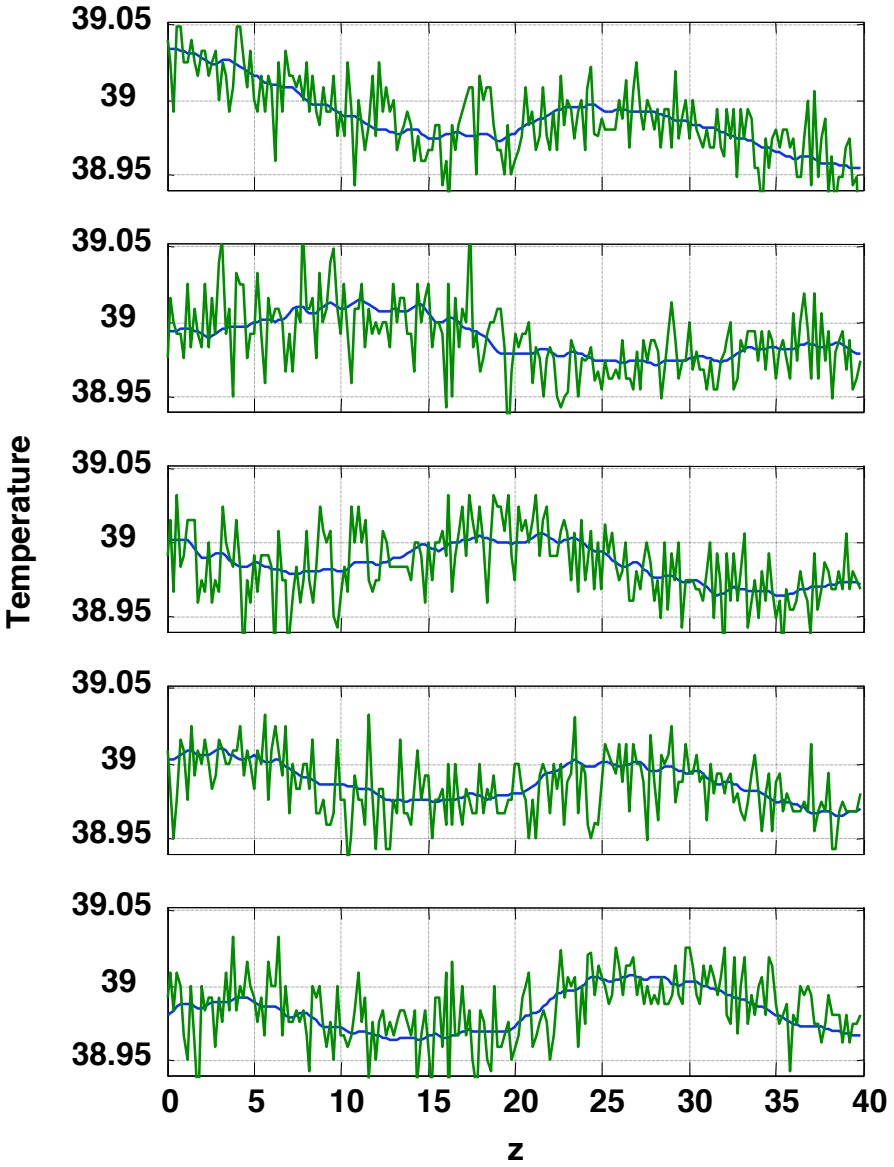


(Groß, Soemers, Mhamdi, Al-Sibai, Reusken, Marquardt, Renz, 2005)

Heat Flux Estimates from Experimental Data







Towards Inverse Transport Problems

CG method for the solution of inverse problem embeds DROPS (Reusken u.a., SFB 540) ein.

DROPS employs

adaptive multi grid methods,

finite element discretization,

levelset method

and facilitates the numerical simulation of multi-phase flow problems in 3D

at high resolution

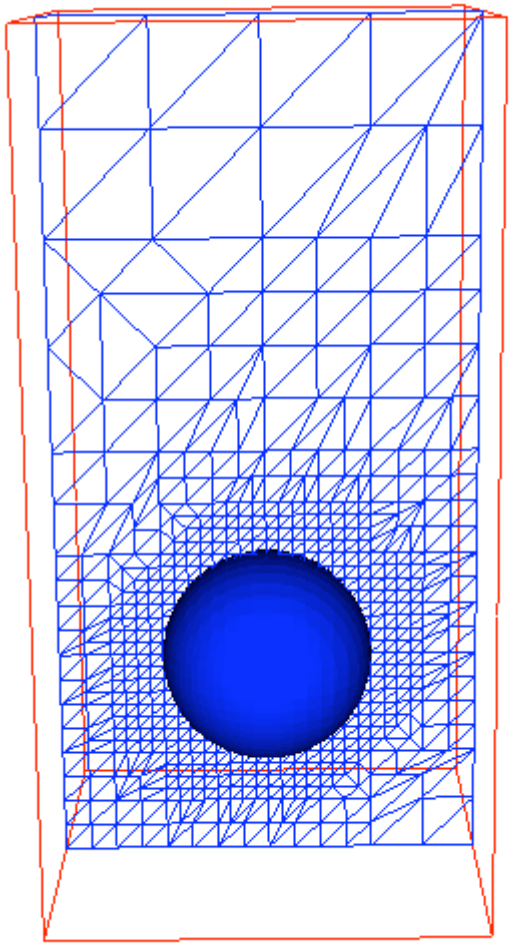
of the phenomena at the phase interface,

efficient and error-controlled

with

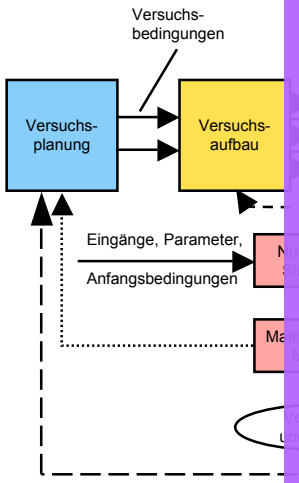
appropriate flexibility

for model extensions.

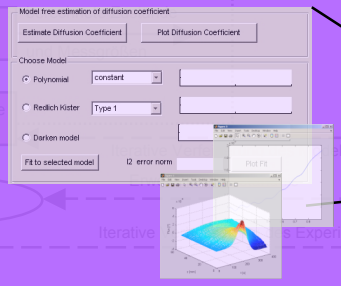


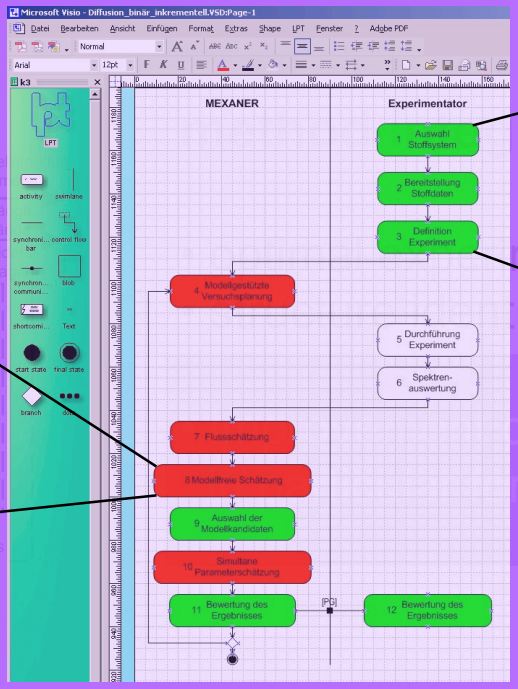
a droplet rising in a stagnant liquid
Pfennig, Reusken u.a., CRC 540

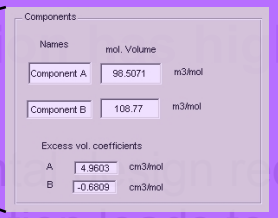
MEXA Business Process – Evaluation



our vision:



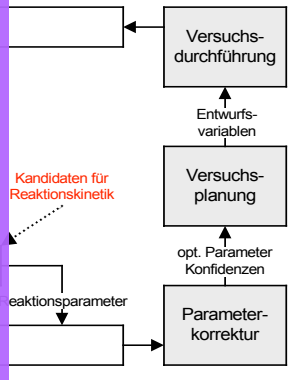




development of an integrated MEXA tool set to transfer systems methods to experimentators

- ⇨ ***planning and coordination…***
- ⇨ ***documentation…***
- ⇨ ***guidance …***
- ⇨ ***process reengineering …***

…of MEXA processes



Lessons Learned

- **accept interactions** between kinetic phenomena **in experiments**,
  but
  isolate them **during identification** by a suitable decomposition strategy

- **high precision calibration** of high-resolution measurements
  (PIV, LIC, LCSM, NMR imaging, Raman / IR spectroscopy etc.)
  often is a difficult modeling problem in itself

- **statistics of measurement errors** need to be included in the analysis

- **flux estimation** is the key to reliable identification

- **tremendous improvements** are possible by systematic
  cross-disciplinary linking of **process systems** and **experimental skills**

Some Challenges for Future Work

- **refinement of MEXA** work process
  - flux estimation (estimation quality, numerical efficiency)
  - exploit error statistics
  - integrated calibration and kinetics identification
  - tailor optimal experimental design methods to incremental identification
  - assessment of identifiability
  - adjust model parameterization to information content of experiments

- **roll-out MEXA** strategy from meso- to micro- and macro-scale
  - hybrid modeling on macro-scale
  - model structure generation from molecular simulation results

- **application and benchmarking of MEXA** work process
  - complicated reaction and transport problems
  - population systems (crystallization, ...)
  - biological systems (metabolic pathways, ...)

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