Two methods for parameter estimation in granulation modelling

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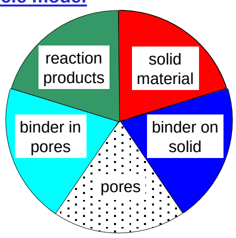
An integrated approach for estimating unknown parameters in a multidimensional population balance model for granulation is presented. Following a prescan of the parameter space, two different methods—the projection method and a Bayesian approach—are used to estimate five unknown rate constants. Similarities and differences between the methods are highlighted.

1. Background

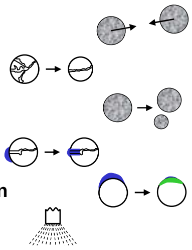
The evolution of the particles during a wet granulation process is described by a multidimensional framework with following particle model and incorporated transformations [1]:

Particle model



Transformations

- Coalescence
- Compaction
- Breakage
- Penetration
- Chemical reaction
- Addition of liquid



Unknown parameters

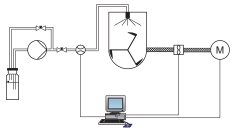
$\hat{K}_{coag} = ?$

$k_{comp} = ?$

$\hat{k}_{att} = ?$

$\hat{k}_{pen} = ?$

$k_{reac} = ?$



Experimental data

Fully parameterised model

2. Theory

2.1 Prescan of parameter space

- initial search in parameter space for best parameter combination
- spread points with quasi-random sequences (Halton sequences)

→ select best parameter combination for further treatment

2.2 Parameter estimation

Surrogate model

- construct surrogate model around chosen parameter combination
- here: 2nd order response surface
- represent unknown parameters as coded variables $\tilde{x}$

$$\eta(\tilde{x}) = \beta_0 + \sum_{k=1}^{K} \beta_k \tilde{x}_k + \sum_{k=1}^{K} \sum_{l \geq k}^{K} \beta_{kl} \tilde{x}_k \tilde{x}_l \qquad (1)$$

$\eta(\tilde{x})$ = model response
K = number of variables
$\beta_0, \beta_k, \beta_{kl}$ = parameters of surface

a) Projection method

- each parameter $\tilde{x}$ can be represented by

$$\tilde{x} = \tilde{x}_0 + c\xi \qquad \xi \sim N(0,1) \qquad (2)$$

- introduction of (2) in (1) yields a model response [2] whose *mean* is

$$\mu(\tilde{x}_0, c) = E[\eta(\tilde{x}_0, c, \xi)] = \eta(\tilde{x}_0) + \sum_{k=1}^{K} \beta_{kk} c_k^2$$

- apply *moment-matching objective function*

$$\Phi(\tilde{x}_0, c) = \sum_{i=1}^{N} \left(\left[\eta_{0,i}^{exp} - \mu_i(\tilde{x}_0, c)\right]^2 + \left[\sigma_i^{exp} - \sigma_i(\tilde{x}_0, c)\right]^2 \right)$$

i = experiment index ($i = 1, \ldots, N$)
μ, σ = model response
$\eta_0^{exp}, \sigma^{exp}$ = experimental values

to obtain *unknown parameters* $\tilde{x}_0^*$ and c^*

$$(\tilde{x}_0^*, c^*) = \underset{\tilde{x}_0, c}{\operatorname{argmin}} [\Phi(\tilde{x}_0, c)]$$

b) Bayesian approach

- models belief via probability distributions
- prior belief updated with data

prior belief —data→ posterior distribution

Bayes' theorem:
$$p(\tilde{x} \mid \eta^{exp}) = \frac{p(\eta^{exp} \mid \tilde{x}) p(\tilde{x})}{\int p(\eta^{exp} \mid \tilde{x}') p(\tilde{x}') d\tilde{x}'} \propto p(\eta^{exp} \mid \tilde{x}) p(\tilde{x})$$

$p(\tilde{x})$ = prior for parameters
$p(\tilde{x} \mid \eta^{exp})$ = posterior distribution
$p(\eta^{exp} \mid \tilde{x})$ = data distribution

- sampling from posterior parameter distribution with Metropolis-Hasting algorithm [3]

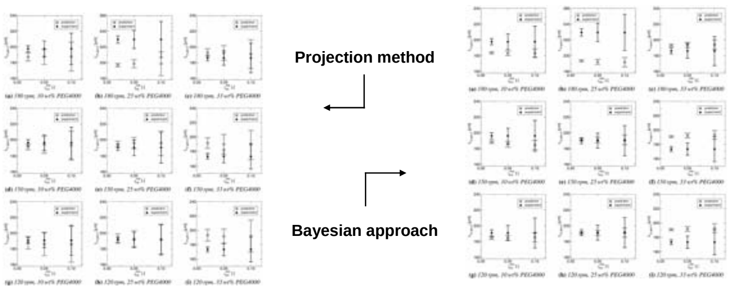
3. Results

Experimental system

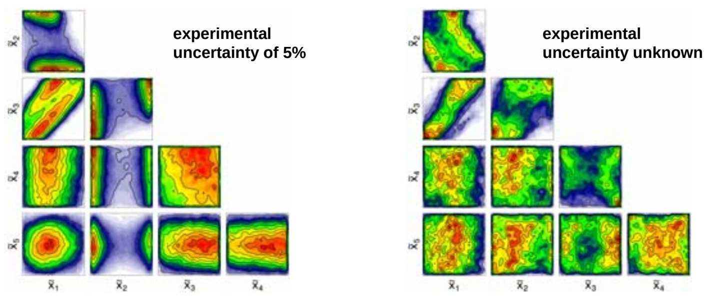
- wet granulation of sodium carbonate with aqueous PEG4000 solutions in bench scale mixer (Kemutec)
- process conditions: 3 impeller speeds and 3 binder compositions

Model predictions vs. experiments

- assumed experimental uncertainty of 2, 5 and 10 %



Posterior distributions (Bayesian approach)



- bimodal distribution for $\tilde{x}_2$ → question of identifiability
- correlation between $\tilde{x}_2$ and $\tilde{x}_3$

4. Conclusions

- model predictions and their uncertainties with parameter estimates obtained through projection method and Bayesian approach are similar
- bimodality in compaction rate constant $\tilde{x}_2$ raises question about identifiability
- correlation between 2 unknown parameters revealed with Bayesian approach
- experimental design might help to resolve current discrepancies

References

[1] A. Braumann, M. Kraft, W. Wagner, *J. Comput. Phys.* **2010**, 229*(20)*, 7672
[2] A. Braumann, P. L. W. Man, M. Kraft, *Ind. Eng. Chem. Res.* **2010**, *49(1)*, 428
[3] A. Braumann, P. L. W. Man, M. Kraft, *c4e-Preprint Series* **2010**, Technical Report 96

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