On the use of shape constraints for state estimation in reaction systems

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June, 2016

Outline

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- System representation
- Shape constraints
  1. using concentrations
  2. using extents
- State estimation via RNK filter
- Simulated case study
- Conclusion

Motivation

Problem definition

- Measurements are usually corrupted with both systematic and random errors
- Models of the reaction system also contain some uncertainity

Problem definition

- Given a process model and measurements up to time t_h, what are the best estimates of the state variables at t_h?

- The estimated variables can then be used for process monitoring and control

System representation

Material balance equations

- Consider a reaction system with S species, R reactions, p inlets and one outlet stream
- System representation in terms of numbers of moles:

Material balance equations - All species and invariants

$$
\begin{array}{lll}
\text{(Species)} & \dot{\mathbf{n}}(t) = \mathbf{N}^{\mathrm{T}}\,\mathbf{r}_v(t) + \mathbf{W}_{in}\,\mathbf{u}_{in}(t) - \omega(t)\mathbf{n}(t) & \mathbf{n}(0) = \mathbf{n}_0 \\
\text{(Invariants)} & \mathbf{P}^{+}\mathbf{n}(t) = \mathbf{0}_q & \mathbf{P}^{+}\left[\mathbf{N}^{\mathrm{T}}\,\mathbf{W}_{in}\,\mathbf{n}_0\right] = \mathbf{0}_q
\end{array}
$$

- where $\omega(t) := \frac{u_{out}(t)}{m(t)}$ is the inverse residence time
- $d = R + p + 1$ is the number of variant states and $q = S - d$ is the number of invariants
- *Note:* $d = R + p$ for semi-batch and $d = R$ for batch reactor

- Consider a reaction system with S species, R reactions, p inlets and one outlet stream
- System representation in terms of numbers of moles:

Material balance equations - Independent and dependent species

$$\begin{aligned}
&\text{(Independent)} \qquad \dot{\mathbf{n}}_1(t) = \mathbf{N}_1^{\mathrm{T}}\mathbf{r}_v(t) + \mathbf{W}_{in,1}\mathbf{u}_{in}(t) - \omega(t)\mathbf{n}_1(t) \quad \mathbf{n}_1(0) = \mathbf{n}_{01} \\
&\text{(Dependent)} \qquad \mathbf{n}_2(t) = -(\mathbf{P}_2)\,\mathbf{P}_1^{+}\,\mathbf{n}_1(t)
\end{aligned}$$

- d differential equations and q algebraic equations

System representation

Vessel extents equations

- An alternative representation is based on the concept of extents[1]
- For a chemical reactor with S species, R reactions, p inlets and one outlet stream: there are d variant states called extents and q invariant states

Vessel extents equations

$$\dot{\mathbf{x}}_r(t) = \mathbf{r}_v(t) - \omega(t)\,\mathbf{x}_r(t) \qquad \mathbf{x}_r(0) = \mathbf{0}_R$$

$$\dot{\mathbf{x}}_{in}(t) = \mathbf{u}_{in}(t) - \omega(t)\,\mathbf{x}_{in}(t) \qquad \mathbf{x}_{in}(0) = \mathbf{0}_p$$

$$\dot{x}_{ic}(t) = -\omega(t)\,x_{ic}(t) \qquad x_{ic}(0) = 1$$

$$\mathbf{x}_{iv}(t) = \mathbf{0}_q$$

$$\mathbf{n}(t) = \mathbf{N}^{\mathrm{T}}\,\mathbf{x}_r(t) + \mathbf{W}_{in}\mathbf{x}_{in}(t) + \mathbf{n}_0 x_{ic}(t)$$

[1] Rodrigues et al., *Variant and Invariant States for Chemical Reaction Systems*, Comp & Chem Eng. 73, p. 23-33, 2015

Example

Semi-batch reactor

- Consider the following two-reaction system:

Reaction system

$$R1: \quad A + B \rightarrow C \qquad\qquad r_1 = k_1\, c_A\, c_B$$
$$R2: \quad A + C \rightarrow D \qquad\qquad r_2 = k_2\, c_A\, c_C$$

- The reaction system is operated in a semi-batch reactor with an inlet stream of B
- The number of independent species is equal to $d = R + p = 3$
- Species A, B and D are chosen as the independent species

- For the reaction system in a semi-batch reactor

$$R1: \quad A + B \rightarrow C \qquad\qquad r_1 = k_1\, c_A\, c_B$$
$$R2: \quad A + C \rightarrow D \qquad\qquad r_2 = k_2\, c_A\, c_C$$

Material balance equations

$$\dot{n}_A(t) = -V(t)\, r_1(t) - V(t)\, r_2(t) \qquad n_A(0) = n_{A0}$$
$$\dot{n}_B(t) = -V(t)\, r_1(t) + w_{in,B}\, u_{in}(t) \qquad n_B(0) = n_{B0}$$
$$\dot{n}_D(t) = V(t)\, r_2(t) \qquad n_C(0) = n_{C0}$$
$$n_C(t) = n_{A0} + n_{C0} + 2\, n_{D0} - n_A(t) - 2\, n_D(t)$$

Example

System representation

- For the reaction system in a semi-batch reactor

$$R1: \quad A + B \rightarrow C \qquad\qquad r_1 = k_1\, c_A\, c_B$$
$$R2: \quad A + C \rightarrow D \qquad\qquad r_2 = k_2\, c_A\, c_C$$

Vessel extent equations

$$\dot{x}_{r,1}(t) = V(t)\, r_1(t) \qquad\qquad x_{r,1}(0) = 0$$
$$\dot{x}_{r,2}(t) = V(t)\, r_2(t) \qquad\qquad x_{r,2}(0) = 0$$
$$\dot{x}_{in}(t) = u_{in}(t) \qquad\qquad x_{in}(0) = 0$$
$$\mathbf{n}(t) = \mathbf{N}^{\mathrm{T}}\, \mathbf{x}_r(t) + \mathbf{W}_{in}\mathbf{x}_{in}(t) + \mathbf{n}_0$$

Shape constraints

Numbers of moles - Generally valid constraints

- Numbers of moles are affected by various rate processes - Hard to impose shape constraints

Batch reactor

- If a species appears only as reactant (product) in an irreversible reaction, then the corresponding number of moles is monotonically decreasing (increasing)

Semi-batch reactor

- If a species appears only as reactant (product) in an irreversible reaction and is not added via an inlet stream, then the corresponding number of moles is monotonically decreasing (increasing)

Shape constraints

Vessel extents - Generally valid constraints (batch and semi-batch reactor)

- Each vessel extent is affected by a single rate process - Easier to impose shape constraints

Vessel extents of inlet

- Nonnegative monotonically increasing functions
- Convex (concave) if the corresponding inlet flowrates are monotonically increasing (decreasing)

Vessel extents of reactions

- Nonnegative monotonically increasing functions,
- Concave (convex) if the corresponding reaction rates are monotonically decreasing (increasing).

Shape constraints

Vessel extents - generally valid constraints (reactors with outlet)

- Each vessel extent is affected by a single rate process and also by the outlet flow rate - There are very few generally valid constraints

Vessel extents of initial conditions

- The extent of initial conditions is a nonnegative monotonically decreasing function

- Constraints on other extents need to be inferred from measurements

Shape constraints

Constraints from measurements

Shape constraints based on measurements

- Select a time window $\mathcal{T}$ of size N
- Compute the extents $\tilde{\mathbf{x}}(t_h) = \mathbf{T}\,\tilde{\mathbf{n}}(t_h)$ in the time window $\mathcal{T}$ from the *measured* numbers of moles $\tilde{\mathbf{n}}(t_h)$
- Calculate the first and second derivatives of each extent using the analytical expressions of the kinetic models
- Monotonicity constraints based on the sign of the estimated first derivatives: increasing (+) / decreasing (-)
- Design shape constraints based on the sign of the estimated second derivatives: convex (+) / concave (-)

- Note that measurement-based constraints can also be applied to numbers of moles

State estimator

Receding-horizon nonlinear Kalman filter (RNK)

- The RNK filter is a nonlinear filter based on the prediction and update steps of a Kalman filter
- The system representation with process and measurement noises can be written as:

System representation - Vessel extents

$$
\begin{aligned}
\dot{\mathbf{x}}_r(t) &= \mathbf{f}_r = \mathbf{r}_v(t) - \omega(t)\,\mathbf{x}_r(t) + \mathbf{w}_r(t) & \mathbf{x}_r(0) &= \mathbf{0}_R \\
\dot{\mathbf{x}}_{in}(t) &= \mathbf{f}_{in} = \mathbf{u}_{in}(t) - \omega(t)\,\mathbf{x}_{in}(t) + \mathbf{w}_{in}(t) & \mathbf{x}_{in}(0) &= \mathbf{0}_p \\
\dot{x}_{ic}(t) &= f_{ic} = -\omega(t)\,x_{ic}(t) + w_{ic}(t) & x_{ic}(0) &= 1 \\
\mathbf{y}(t) &= \mathbf{f}_y = \mathbf{N}^{\mathrm{T}}\,\mathbf{x}_r(t) + \mathbf{W}_{in}\mathbf{x}_{in}(t) + \mathbf{n}_0 x_{ic}(t) + \mathbf{v}_y(t) & &
\end{aligned}
$$

- where $\mathbf{w}_r$, $\mathbf{w}_{in}$, w_{ic}, $\mathbf{v}_y$ are Gaussian random variables with zero-mean and constant variance-covariances $\mathbf{Q}_r$, $\mathbf{Q}_{in}$, q_{ic} and $\mathbf{R}_y$

State estimator

RNK - Prediction step

- Given the state vector $\mathbf{x}(t_h|t_h)$, compute the *a priori* estimate $\mathbf{x}_{\mathcal{T}|t_h} = [\mathbf{x}(t_{h+1}|t_h), \ldots, \mathbf{x}(t_{h+N}|t_h)]$ for the time window $\mathcal{T}$
- The elements of the covariance matrix $\mathbf{P}_{\mathcal{T}|t_h}$ are estimated from $\mathbf{P}(t_h|t_h)$ using the following iterative relationships

A priori covariance estimation

$$\mathbf{P}_{t_{h+N}|t_h} = \mathbf{A}^{\mathrm{T}}_{t_{h+N-1}} \mathbf{P}_{t_{h+N-1}|t_h} \mathbf{A}_{t_{h+N-1}} + \mathbf{Q}_x$$

$$\mathbf{P}_{(t_{h+N-1})(t_{h+N})|t_h} = \mathbf{P}_{(t_{h+N-1})(t_{h+N-1})|t_h} \mathbf{A}^{\mathrm{T}}_{t_{h+N-1}}$$

- where $\mathbf{Q}_x = \begin{bmatrix} \mathbf{Q}_r & 0 & 0 \\ 0 & \mathbf{Q}_{in} & 0 \\ 0 & 0 & q_{ic} \end{bmatrix}$ and $\mathbf{A}_{t_h} := \exp\{ \frac{\partial \mathbf{f_x}}{\partial \mathbf{x}} |_{\mathbf{x}(t_h|t_h)} \}$

State estimator

RNK - Update step

- Given the N measured outputs $\mathbf{y}_{\mathcal{T}} := \left[\mathbf{y}(t_{h+1})^{\mathrm{T}}, \ldots, \mathbf{y}(t_{h+N})^{\mathrm{T}}\right]^{\mathrm{T}}$, the update step is formulated as an optimization problem

Update step

$$
\begin{aligned}
\min_{\mathbf{x}_{\mathcal{T}|t_{h+N}}} \quad & \boldsymbol{\alpha}^{\mathrm{T}} \mathbf{P}_{\mathcal{T}|t_h}^{-1} \boldsymbol{\alpha} + \boldsymbol{\beta}^{\mathrm{T}} \mathbf{R}_y^{-1} \boldsymbol{\beta} \\
\text{s.t.} \quad & \boldsymbol{\alpha} := \mathbf{x}_{\mathcal{T}|t_{h+N}} - \mathbf{x}_{\mathcal{T}|t_h} \\
& \boldsymbol{\beta} := \mathbf{y}_{\mathcal{T}} - \mathbf{f}_y\left(\mathbf{x}_{\mathcal{T}|t_h}\right) \\
& \mathbf{h}(\mathbf{x}_{\mathcal{T}|t_{h+N}}) \leq \mathbf{0}_m \\
& \mathbf{x}_{\mathcal{T}|t_{h+N}} \geq \mathbf{0}
\end{aligned}
$$

- where $\mathbf{h}(\cdot)$ denotes the m applicable shape constraints

State estimator

RNK - Update step

- The a posteriori estimate of the covariance matrix is computed as:

A posteriori covariance estimation

$$\begin{aligned}\mathbf{K}_{\mathcal{T}|t_{h+N}} &= \mathbf{P}_{\mathcal{T}|t_h}\mathbf{C}_{\mathcal{T}|t_h}(\mathbf{C}_{\mathcal{T}|t_h}\mathbf{P}_{\mathcal{T}|t_h}\mathbf{C}^{\mathrm{T}}_{\mathcal{T}|t_h} + \mathbf{R}_y)^{-1} \\ \mathbf{P}_{\mathcal{T}|t_{h+N}} &= (\mathbf{I} - \mathbf{K}_{\mathcal{T}|t_{h+N}}\mathbf{C}_{\mathcal{T}|t_h})\mathbf{P}_{\mathcal{T}|t_h}\end{aligned}$$

- where $\mathbf{C}_{\mathcal{T}|t_h}$ is the linearized measurement equation obtained at $\mathbf{x}_{\mathcal{T}|t_h}$

Example

Semi-batch reactor

Reaction system

$$R1: \quad A + B \rightarrow C \qquad r_1 = 0.5\, c_A\, c_B$$
$$R2: \quad A + C \rightarrow D \qquad r_2 = 0.3\, c_A\, c_C$$

- The reaction system is simulated in a semi-batch reactor with $V = 1$ L, $n_{A0} = 5$ mol, and $n_{B0} = n_{C0} = 0$ mol
- Species B is fed to the reactor with the mass flow rate 5 g min^{-1}
- The estimator is initialised with (incorrect) parameter values $\hat{k}_1 = 0.75$ and $\hat{k}_2 = 0.5$ for a window size $N = 10$
- The measurement and process noise matrices are assumed to be known

Example

Semi-batch reactor - Generally valid constraints

- The following constraints are known from prior knowledge

Numbers of moles

- $n_A(t)$ is monotonically decreasing,
- $n_D(t)$ is monotonically increasing.

Vessel extents

- $x_{r,1}(t)$ is concave,
- $x_{r,2}(t)$ is monotonically increasing,
- $x_{in}(t)$ is monotonically increasing.

Example

Semi-batch reactor - Generally valid constraints

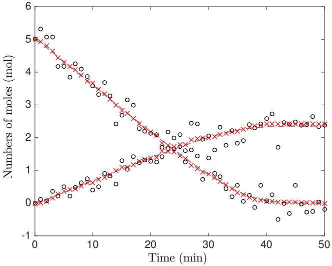

Figure : True (- -), measured ($\circ$) and estimated ($\times$) number of moles for species A and D

Species	Unconstrained	RNK estimation	
	via **n**	via **n**	via **x**
A	0.96	0.44	**0.10**
B	0.19	0.13	**0.06**
C	1.98	0.63	**0.27**
D	0.52	0.21	**0.12**

Table : Sum of squared errors for the measured and estimated numbers of moles

Example

Semi-batch reactor - Measurement-based constraints

- Measurement-based constraints are added to the generally valid constraints

Numbers of moles

- Concave and convex constraints are obtained from measurements for all species

Vessel extents

- Concave and convex constraints on $x_{r,2}(t)$ and $\mathbf{x}_{in}(t)$ are obtained from measurements

- Measurement-based constraints are added to the generally valid constraints

Species	Unconstrained	Generally valid constraints		Measurement-based constraints	
	via **n**	via **n**	via **x**	via **n**	via **x**
A	0.96	0.44	0.10	0.27	**0.06**
B	0.19	0.13	0.06	0.07	**0.04**
C	1.98	0.63	0.27	0.37	**0.26**
D	0.52	0.21	0.12	0.13	**0.10**

Table : Sum of squared errors for the measured and estimated numbers of moles

Conclusion

- The addition of shape constraints improves the accuracy of the estimated state variables
- Shape constraints are easier to define in terms of vessel extents than in terms of numbers of moles
- Measurement-based constraints can also be estimated and improve the estimation
- Extensions: Extending the state estimation problem to simultaneous state and parameter estimations, and also on generating generally valid constraints for reactors with outlet.

Conclusion

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- Measurement-based constraints can also be estimated and improve the estimation
- Extensions: Extending the state estimation problem to simultaneous state and parameter estimations, and also on generating generally valid constraints for reactors with outlet.

Thank you!