Constrained multi-rate state estimator incorporating delayed measurements

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Abstract—Frequent and accurate concentration estimates are important for on-line control and optimization of chemical reaction systems. Such estimates can be obtained using state estimation methods that fuse frequent (fast) delay-free on-line measurements with infrequent (slow) delayed laboratory measurements. In this paper, we demonstrate how several recent advances made in state estimation can be combined in an on-line recursive state estimation framework by imposing knowledge-based and measurement-based constraints on the state estimates of multi-rate concentration measurements with time-varying time delays. This framework is illustrated using a simulated example for a bacterial batch fermentation of recombinant \textit{L. lactis}. It is shown that an extent-based formulation gives more accurate estimates than a conventional concentration-based formulation.

I. INTRODUCTION

Chemical and biochemical reactions are used in the chemical and pharmaceutical industries to convert feed materials into marketable products. Control and on-line optimization require frequent and accurate estimates of concentrations of various key analytes. State estimation techniques utilize measurements of different variables and generate accurate estimates of the process states by enforcing consistency with a known but inaccurate process model [1].

The dynamics of reaction systems are in general described by nonlinear differential or differential-algebraic equations. For these processes, on-line measurements such as temperature, pressure, and absorbance spectra of the reacting mixture are made frequently, and are available almost immediately. On the other hand, concentration measurements are obtained using laboratory instruments such as gas and liquid chromatographs, and are generally available infrequently and with a considerable delay after the samples are taken. The concentrations are all nonnegative and, depending on the type of reactions, they may also increase or decrease monotonically. It is very useful to take into account all these features in developing a state estimator for such reaction systems.

Several state estimators have been reported for nonlinear dynamic systems, among which the Extended Kalman Filter (EKF) is probably the most widely used because of its recursive nature and ease of implementation. Different approaches have been developed to include a variety of multi-rate sampling scenarios. Multi-rate Kalman filter for linear systems was documented by Raghvan \textit{et al.} [2] and Li \textit{et al.} [3]. Its application for a reactive distillation system has been shown by Valluru \textit{et al.} [4], Tatiraju \textit{et al.} [5] considered multi-rate state and parameter estimation for a nonlinear system. However, the EKF inability to include algebraic constraints often makes it unrealistic in practice. To incorporate constraints, one can use a receding-horizon nonlinear Kalman filter (RNK) [6], which is computational simple and includes a prediction and a correction steps as in Kalman filter.

Time delays that occur in the measurements have been handled in chemical systems either by providing a procedure to fuse slow measurements or through state augmentation. Gudi \textit{et al.} [7] applied multi-rate estimation techniques that include delayed measurements to a fermentation reactor. Gopalkrishnan \textit{et al.} [8] provided a detailed review of multi-rate estimation problem in the context of Extended Kalman filter (EKF).

The aforementioned advances in state estimation for nonlinear dynamical processes have been made as separate extensions to recursive estimators. The integration of all these features in a recursive state estimation strategy has not been explored or illustrated using an appropriate process. The objective of this paper is therefore to formulate a constrained multi-rate filtering and estimation problem incorporating delayed and infrequent measurements. Although, moving horizon estimators [9] can also be used to integrate these features, this work considers a recursive state estimation framework that leads to a computationally efficient option well suited to on-line applications.

Chemical reaction systems are often modeled from first principles through material and energy balance equations and are written as highly coupled ordinary differential equations (ODE) in terms of concentrations and temperatures. An alternative representation of the system in terms of vessel extents, proposed by Amrhein \textit{et al.} [10] and subsequently reformulated by Rodrigues \textit{et al.} [11],
can transform the equations into a form that can be easily analyzed. The extent-based formulation, which is shown to be more advantageous for imposing shape constraints, is chosen in this work.

This paper is organized as follows. Section 2 provides a generic description of reaction systems represented in terms of both numbers of moles and extents. In Section 3, the shape properties of the state variables are investigated. Section 4 discusses the types of measurements that are commonly encountered in chemical reaction systems. In Section 5, the RNK problem is reformulated to handle delayed measurements. Section 6 compares the performance of the new estimator via a case study, while Section 7 concludes the paper.

II. SYSTEM DESCRIPTION

A. Mole balance equations

Considering a homogeneous chemical reaction system with S species, R reactions, p inlets and one outlet stream. The corresponding mole balance equations can be written as:

\[ \dot{n}(t) = N^T r_v(t) + W_{in} u_{in}(t) - \omega(t)n(t) \quad n(0) = n_0, \]  

(1)

where \( n \) is the S-dimensional vector of numbers of moles, \( r_v = Vr \) with \( V \) being the volume of the reactor and \( r \) the R-dimensional vector of reaction rates, \( N \) is the \( R \times S \) stoichiometric matrix, \( u_{in} \) is the \( p \)-dimensional vector of inlet mass flowrates, \( \omega = u_{out}/m(t) \) is the inverse residence time with the mass \( m \) and the outlet mass flowrate \( u_{out} \). \( W_{in} \) is the \( S \times p \) matrix of inlet compositions, and \( n_0 \) is the \( S \)-dimensional vector of initial conditions.

The mass \( m \) can be computed from either the knowledge of the numbers of moles \( n \) as \( m(t) = 1_S^T M_n n(t) \), where \( M_n \) is the \( S \)-dimensional diagonal matrix of molecular weights or by integrating the continuity equation \( \dot{n}(t) = 1_S^T u_{in}(t) - u_{out}(t) \) with \( n(0) = m_0 \).

The molar concentrations can be computed from the numbers of moles as \( c(t) = \frac{n(t)}{V_T} \), while the reaction rates \( r(t) \) are typically expressed as nonlinear functions of \( c(t) \) and the reactor temperature \( T(t) \).

The S mole balance equations are often redundant since the system variability is governed by the independent reactions and the inlet and outlet flows, with \( d := R + p + 1 \). Then, there exist \( q = S - d \) invariants that are equal to zero [11]:

\[ P^+ n(t) = 0_q, \]  

(2)

where the columns of the \( S \times q \) matrix \( P \) describe the left null space of the matrix \([N^T W_{in} n_0]\). The superscript + indicates pseudo-inversion.

Using (2), Eq. (1) can be written as:

\[ n_1(t) = N^T r_v(t) + W_{in,1} u_{in}(t) - \omega(t)n_1(t) \quad n_1(0) = n_{1,0} \]

\[ n_2(t) = -P_2 P^+_2 n_1(t), \]  

(3)

where \( n_1 \) is the \( d \)-dimensional vector of independent species, \( n_2 \) the \( q \)-dimensional vector of dependent species. \( N_1 \) the corresponding \( R \times d \) stoichiometric submatrix, \( W_{in,1} \) the \( d \times p \) submatrix of inlet compositions, \( n_{1,0} \) the \( d \)-dimensional vector of initial conditions, \( P_1 \) the \( d \times q \) submatrix of \( P \) corresponding to the independent species, and \( P_2 \) the \( q \times q \) submatrix of \( P \) corresponding to the dependent species. Note that the set of independent species are chosen such that the rank of the matrix \([N^T W_{in,1} n_{1,0}] = d \).

B. Alternative representation in terms of extents

The reaction system can alternatively be written in terms of extents through the linear transformation [11]:

\[ x(t) = T_1 n_1(t) = [N^T W_{in,1} n_{10}]^{-1} n_1(t) \]  

(5)

The resulting dynamic model in terms of extents reads:

\[ x_r(t) = r_v(t) - \omega(t)x_r(t), \quad x_r(0) = 0_R \]  

(6)

\[ x_{in}(t) = u_{in}(t) - \omega(t)x_{in}(t), \quad x_{in}(0) = 0_p \]  

(7)

\[ x_{ic} = -\omega(t)x_{ic}(t), \quad x_{ic}(0) = 1, \]  

(8)

where \( x_r \) are the \( R \) reaction extents, \( x_{in} \) the \( p \) extents of inlet, and \( x_{ic} \) the extent of initial conditions. The numbers of moles can be reconstructed as:

\[ n(t) = N^T x_r(t) + W_{in} x_{in}(t) + n_0 x_{ic} \]  

(9)

III. MEASUREMENTS

The measurements can be categorized as fast and slow depending on the associated sampling frequency. Let \( h \) represent the smallest sampling interval. The fast measurements, \( y_f \in \mathbb{R}^r \), are available at the sampling instants \( \{t_k = k h; \ k = 0, 1, 2, \ldots\} \), whereas the slow measurements, \( y_s \in \mathbb{R}^s \), are available with the constant delay \( \theta \) at the sampling instants \( \{t_k = bh + \theta; \ b \in \mathbb{N}^+\} \).

The sampling scheme is illustrated in Figure 1.

![Fig. 1. Fast and slow sampling with corresponding measurement availability.](image)

The available measurements are as follows:

- At the **fast sampling** instants, \( t_k = kh \):

\[ y_f(t_k) = C_f x(t_k) + \nu_f(t_k) \]  

(10)

- At the **slow sampling** instants, \( t_k = bh + \theta \):

\[ y_s(t_k) = \begin{bmatrix} C_f & 0_{r \times d} & C_s \end{bmatrix} \begin{bmatrix} x(t_k) \\ \nu_f(t_k) \\ y(t_k - \theta) \end{bmatrix}, \]  

(11)
where $C_f$ and $C_s$ are the fast and slow measurement matrices, respectively. We assume that, at the slow sampling instants, the fast and slow measurements are available simultaneously.

The measurement noises are modeled as zero-mean white-noise processes with Gaussian distribution, that is, $\nu_f(t_k) \sim \mathcal{N}(0, R_f)$ and $\nu_s(t_k - \theta) \sim \mathcal{N}(0, R_s)$. For the sake of simplicity, the approach is presented here for the case of regular sampling and constant measurement delay. However, the approach can easily be modified to apply to irregularly sampled systems with time-varying delays.

IV. State Constraints

The constraints for the state estimation problem are either known a priori or inferred from measurements. These constraints are discussed next.

A. Knowledge-based constraints

Knowledge-based constraints express physical considerations and are derived from first principles [12]:

- All numbers of moles are nonnegative. Furthermore, the invariant relationships call for $P^+ n(t_k) = 0_n$.
- The number of moles of a non-added non-produced species that is only involved in irreversible reactions is nonincreasing. Similarly, the number of moles of a non-removed non-consumed species that is only involved in irreversible reactions is nondecreasing.
- In a batch or semi-batch reactor, the extents of irreversible reactions are (i) nonnegative nondecreasing functions, and (ii) concave (convex) functions if the corresponding rate expressions for nonnegative nonincreasing (nondecreasing).

B. Measurement-based constraints

Even for cases where knowledge-based shape constraints cannot be formulated a priori, certain shape constraints exist and can be derived from measurements [12]. The computational procedure is as follows:

1. Evaluate analytically the expressions of the first and second derivatives for the extents (6)–(8), that is, $\dot{x}_j(x)$ and $\ddot{x}_j(x)$, $j = 1, \ldots, d$, using the kinetic rate expressions $r(x)$.
2. Compute the experimental extents $\bar{x} = T_x \bar{n}$ using the measured numbers of moles $\bar{n}$.
3. Using the analytical expressions obtained in Step (1), compute the numerical values of the first and second derivatives for each extent.
4. For the window $\mathcal{X}$ of size $m$, impose convexity/concavity constraints based on the estimated second derivatives (using tolerance $\delta_2 > 0$):
   - if $\ddot{x}_j > \delta_2$ in $\mathcal{X}$, then $x_j$ is convex in $\mathcal{X}$,
   - if $\ddot{x}_j < -\delta_2$ in $\mathcal{X}$, then $x_j$ is concave in $\mathcal{X}$,
   - otherwise, no second-order shape constraints can be imposed in $\mathcal{X}$; the presence of first-order shape constraints can be investigated based on the estimated first derivatives (using tolerance $\delta_1 > 0$):
     - if $\dot{x}_j > \delta_1$ in $\mathcal{X}$, then $x_j$ is monotonically increasing in $\mathcal{X}$,
     - if $\dot{x}_j < -\delta_1$ in $\mathcal{X}$, then $x_j$ is monotonically decreasing in $\mathcal{X}$,
     - otherwise, no shape constraints can be imposed.

V. State estimation

The state estimation problem is formulated as a RNK filter for which constraints on the state variables can easily be imposed. The uncertainties in the kinetic models and measurement equations are modeled as additive Gaussian random variables.

The system representation in terms of $n_1$ reads:

$$\dot{n}_1(t) = N_f^T r_c(t) + W_{in,1} u_{in}(t) - \omega(t) n_1(t) + w_{n1}(t)$$

where $w_{n1} \sim \mathcal{N}(0_d, Q_{n1})$ is a zero-mean Gaussian random variable with covariance matrix $Q_{n1}$.

The system representation in terms of extents becomes:

$$\dot{x}_c(t) = r_c(t) - \omega(t)x_c(t) + w_c(t), \quad x_c(0) = 0_d$$

$$x_{in}(t) = u_{in}(t) - \omega(t)x_{in}(t) + w_{in}(t), \quad x_{in}(0) = 0_e$$

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

with $w_c(t) \sim \mathcal{N}(0_d, Q_c)$, $w_{in}(t) \sim \mathcal{N}(0_d, Q_{in})$ and $w_{ic}(t) \sim \mathcal{N}(0_d, Q_{ic})$.

Receding-horizon nonlinear Kalman filter

The RNK filter implementation is briefly discussed next. For the sake of brevity, the estimator is only formulated in terms of extents.

A. RNK with only fast measurements

The following shorthand notations are introduced. The right-hand side of Eqs. (13)–(15) are denoted as $f_r(\cdot), f_{in}(\cdot)$ and $f_{ic}(\cdot)$ and are assembled in the vector $f$. The block-diagonal matrix $Q$ represents the error covariance matrix of the modeled extents:

$$x = \begin{bmatrix} x_r \\ x_{in} \\ x_{ic} \end{bmatrix}, \quad f = \begin{bmatrix} f_r \\ f_{in} \\ f_{ic} \end{bmatrix}, \quad w = \begin{bmatrix} w_r \\ w_{in} \\ w_{ic} \end{bmatrix}, \quad Q = \begin{bmatrix} Q_r & 0 & 0 \\ 0 & Q_{in} & 0 \\ 0 & 0 & Q_{ic} \end{bmatrix}$$

We discuss next the prediction and update steps.
1) Prediction step: At time instant $t_k$ we define a fixed lag augmented state vector $x_{T_k} = [x_{T_k}^T \ldots x_{T_k}^{T-m+2}]^T$ for the prediction window $T_k$ of size $m$. Given the smoothed estimates $\hat{x}_{T_k}$ and the corresponding covariance $P_{T_k|m-1}$, the open loop estimates of the $x_{T_k}$ is obtained by appropriately integrating the nonlinear state evolution equations (13)-(15). These are assembled in the $md\times (m+1)d$ matrix

$$A_{T_k} = \begin{bmatrix} A_{T_k,m} & 1 \ldots & 0 \\ A_{T_k,m+1} & A_{T_k,m} & \ldots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ \prod_{i=k-m}^{k-1} A_{T_k} & \prod_{i=k-m+1}^{k-1} A_{T_k} & \ldots & I_d \end{bmatrix}$$

Since the noise in the states is uncorrelated with the state predictions, the error covariance matrix estimates $\hat{\xi}$ can be computed as the solution to the following constrained linear least-squares problem:

$$\begin{align*}
\hat{\xi}_{T_k} &= \arg \min_{\xi_{T_k}} \xi_{T_k}^T P_{T_k|m-1}^{-1} \xi_{T_k} + \alpha \xi_{T_k}^T R_{f,T_k}^{-1} \xi_{T_k}, \\
\text{s.t.} \quad &C(x_{T_k|m-1}) \leq 0, \quad \forall i = 1, \ldots, m
\end{align*}$$

where $R_{f,T_k}$ is the $md \times md$ measurement noise covariance matrix. The function $C(.)$ represents the shape constraints applicable to the corresponding time window. The a posteriori covariance matrix $P_{T_k|m}$ is computed as follows [6]:

$$K_{T_k} = P_{T_k|m} C_{f,T_k}^T (C_{f,T_k} P_{T_k|m} C_{f,T_k}^T + R_{f,T_k})^{-1}$$

$$P_{T_k|m} = (I_m - K_{T_k} C_{f,T_k}) P_{T_k|m-1}$$

Although the update step for $P_{T_k|m}$ does not strictly hold for constrained estimates, this step is retained because it is indicative of the quality of the estimates [6]. The prediction-update steps are repeated for the next time window from $t_k+m+1$ to $t_{k+1}$ as illustrated in Figure 2. $x_{T_k}|m+1$ is required for the next window is obtained from the first $d$ elements of $x_{T_k}|m$, while $P_{T_k|m+1}$ is obtained from the first $d \times d$ block of $P_{T_k|m}$.

B. RNK with fast and slow measurements

The sample augmentation method [8] is used to include the delayed slow measurements into the RNK filter formulation. The $md\times (m+1)d$-dimensional state vector over the window $T_k$ is extended to also include a $d\times d$-dimensional state $x_{sf}$, where $\tau$ is the time instant at which the slow measurement sample is taken. This new state can be interpreted as an ephemeral state with no dynamics and initial condition equal to its filtered estimate at $\tau$, that is, $dx_{sf}/dt \big|_{t=\tau} = 0$, $x_{sf}(0) = x_{sf}|\tau$. For each subsequent window, the open loop estimate of $x_{sf}$ would be given by the smoothed estimate from the previous instance. The $(m+1)d \times d$-dimensional extended augmented fixed lag state vector is defined as

$$x_{T_k}^a = [x_{T_k}^T \, x_{sf}^T]^T,$$

which can also be similarly expressed in terms of $x_{T_k}$ and the noise terms $w_i, i = t_k-m+1, t_k-m+2, \ldots, t_k$ as:

$$x_{T_k}^a = A_{T_k}^a [x_{T_k}^T \, w_{T_k|m+1} \ldots w_{T_k}^T \, x_{sf}^T]^T$$

with the $(m+1)d\times (m+1)d$ matrix

$$A_{T_k}^a = \begin{bmatrix} A_{T_k,m} & I_d \ldots & 0 \\ A_{T_k,m+1} & A_{T_k,m} & \ldots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ \prod_{i=k-m}^{k-1} A_{T_k} & \prod_{i=k-m+1}^{k-1} A_{T_k} & \ldots & I_d \end{bmatrix}$$

The error covariance matrix for $x_{T_k}^a$ at the slow sampling instant $t_k = \tau$ is initialized as $P_{T_k|m} = P_{T_k|m}$ and
The latter representing the cross covariance between the estimates at \( \tau \) and \( t_k - m \).

The error covariance matrix for the augmented state at any time \( t_k \) before measurement arrival is computed as in Eq. (16):

\[
P_{\tau,t_k-m|t_k} = P_{t_k,t_k-m|t_k}.
\]

The state augmentation ceases as soon as all the measurements pertaining to the augmented states has been received. The update step follows as in RNK with only fast measurement with modified model matrices as described in section III.

VI. SIMULATED CASE STUDY

The application of the RNK framework is illustrated on a simulated batch fermentation reactor [13]. There are two reactions, corresponding to the biomass growth and the death phase, and four species, namely glucose \((G)\), biomass \((X)\), hyaluronic acid \((HA)\), and lactic acid \((LA)\). The temperature and pH are controlled at constant values, and the initial glucose and biomass concentrations are 40 g/L and 0.05 g/L, respectively. The dynamic model reads:

\[
\frac{dX}{dt} = (\mu - m_s)X
\]

\[
\frac{dG}{dt} = -\frac{1}{Y_{XG}}\mu X
\]

\[
\frac{dLA}{dt} = (\alpha_{LA}\mu + \beta_{LA})X
\]

\[
\frac{dHA}{dt} = (\alpha_{HA}\mu + \beta_{HA})X
\]

The specific growth rate \( \mu \) is a function of the concentrations of all the species of interest

\[
\mu = \frac{\mu_{max}G}{(K_s + G + \frac{G}{K_H})(1 + \frac{LA}{K_L})(1 + \frac{HA}{K_H})},
\]

where all the concentrations are in g/L. The model has 11 parameters, \( \mu_{max}, m_s, Y_{XG}, \alpha_{LA}, \beta_{LA}, \alpha_{HA}, \beta_{HA}, K_s, K_L, K_{LA}, \) and \( K_{HA} \). The parameters used for this study are based on Badle [13].

The system of reactions can be reformulated in terms of the two extents of reaction \( \dot{x}_r = [x_1^r, x_2^r]^T \) as

\[
\frac{dx_1^r}{dt} = \mu X
\]

\[
\frac{dx_2^r}{dt} = X
\]

The process noise covariance matrix \( Q \), which is of dimension \( 2 \times 2 \) as there are only two states in the extent formulation, is given as:

\[
Q = \begin{bmatrix} 0.0001 & 0 \\ 0 & 0.05 \end{bmatrix}
\]

For numbers of moles

\[
\text{Nonnegativity of } x_i(t_k), \quad \alpha_{LA}, \beta_{LA}, \alpha_{HA}, \beta_{HA}, K_s, K_L, K_{LA}, K_{HA}
\]

The shape constraints listed in Table II are imposed. A window size of 30 min \((m = 5)\) is chosen, which corresponds to the largest measurement arrival delay. \( R_s \) is assumed to be diagonal, with the diagonal elements indicating the error variances of the measured species.

\[
R_s = \begin{bmatrix} 0.0001 & 0 & 0 & 0 \\ 0 & 0.04 & 0 & 0 \\ 0 & 0 & 0.04 & 0 \\ 0 & 0 & 0 & 0.0001 \end{bmatrix}
\]

The process noise covariance matrix \( Q \), which is of dimension \( 2 \times 2 \) as there are only two states in the extent formulation, is given as:

\[
Q = \begin{bmatrix} 0.0001 & 0 \\ 0 & 0.05 \end{bmatrix}
\]
A. Results and discussions
The performance of the estimators designed in both the number of mole and extent domains is compared. To obtain a dimensionless evaluation criterion, the ratio of the sum of squared errors to the square of the concentration range is computed for each species. Tables IV and III list the estimation performance in terms of numbers of moles and extents, respectively.

A significant improvement is observed when shape constraints are applied. This is more pronounced in the extent-based formulation thanks to the additional constraints that are present in that domain. Performance is further increased by including slow measurements and working in the extent domain.

### TABLE III

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### TABLE IV

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VII. Conclusion
The paper presents the formulation of a multi-rate constrained state estimator that incorporates infrequent and delayed measurements. The estimator is designed using state-space representations in terms of both numbers of moles and extents of reactions. The formulation can be extended to cases where the delay is not fixed but varies with time. A comparison is made between the estimates obtained using stand-alone fast measurements and those obtained through a combination of both fast and slow measurements. The constrained multi-rate state estimator outperforms the unconstrained one in both the number of moles and extent domains. The extent-based formulation provides better estimates due to the additional constraints that can be enforced.

REFERENCES


