Data Reconciliation in Open Reaction Systems using the Concept of Extents

Sriniketh Srinivasan[1], Julien Billeter[1], Shankar Narasimhan[2] and Dominique Bonvin[1]

[1] Laboratoire d'Automatique, Ecole Polytechnique Fédérale de Lausanne. Switzerland
[2] Department of Chemical Engineering, Indian Institute of Technology, Madras

Kinetic models of chemical reaction systems are typically represented in terms of state variables, such as concentrations, temperature and partial pressures [1]. These state variables in turn depend on the underlying reactions, transfer phenomena, and transport due to the inlet and outlet flows. Kinetic models are derived from first principles – material and energy balances – and are expressed as a set of differential and algebraic equations (DAE), with the differential equations describing the evolution over time and the algebraic equations describing relationships that have to be satisfied at each time instant [2]. The identification of kinetic models along with the estimation of their parameters is carried out using measurements obtained from experiments performed under well-chosen and often ideal experimental conditions [3]. Kinetic models can then be adapted to non-ideal process operations using real-time measurements for the purpose of monitoring, control and optimization [4-5].

Measurements are inherently corrupted with noise. The quality of measurements made during the identification experiment affects kinetic identification, while the quality of measurements made during process operation affects its efficiency. Data reconciliation techniques rely on balance equations (here algebraic constraints) to improve the accuracy of these measurements, with more relationships leading to better reconciliation [6]. Hence, data reconciliation can be formulated as a constrained optimization problem in terms of measured and reconciled variables. However, since these variables are typically involved in more than one rate process, it is difficult to add additional shape constraints involving for example monotonicity.

Several alternative representations of reaction systems in terms of variants and invariants have been proposed in the literature. Asbjørnsen and co-workers [7], for example, introduced a two-way decomposition into reaction variants and reaction invariants. Unfortunately, the transformed variables are also flow variant. Srinivasan et al. [8] introduced a nonlinear decomposition into reaction variants, flow variants, and reaction and flow invariants. Note that all these representations use abstract variables that do not carry any physical meaning. Recently, a representation of reaction systems using a linear transformation has been proposed. The transformed states, called *vessel extents*, have a clear physical meaning, and, in addition, each vessel extent is associated with a single rate process [9].

Srinivasan et al. [10] have recently shown that the transformation to vessel extents allows a general formulation of process constraints under all common operating conditions, namely, batch, semi-batch and continuous mode. In addition, the extents are monotonically increasing in the absence of an outlet stream (batch and semi-batch mode). The same authors have also shown that the addition of monotonicity constraints to the data-reconciliation problem improves the accuracy of the reconciled estimates. Unfortunately, the monotonicity of extents cannot be guaranteed in the presence of an outlet stream.

In this contribution, piecewise monotonicity constraints on extents are applied for the purpose of data reconciliation in the presence of an outlet stream, that is, in continuous operation mode. A general procedure is presented to identify regions where these state variables are monotonically increasing or decreasing. The strength of these piecewise shape constraints for the task of data reconciliation will be illustrated via simulated examples.

References

[1] O. Levenspiel. *Chemical Reaction Engineering*, Wiley, 1972.

[2] C. G. Hill and W. R. Thatcher. *Introduction to Chemical Engineering Kinetics and Reactor Design*, John Wiley & Sons, 2014.

[3] A. Bardow and W. Marquardt. Incremental and simultaneous identification of reaction kinetics: Methods and comparison, Chem. Eng. Sci., 59, 2673-2684, 2004.

[4] A. Marchetti, B. Chachuat and D. Bonvin. Modifier-adaptation methodology for real-time optimization, Ind. Eng. Chem. Res., 48(13), 6022-6033, 2009.

[5] S. Srinivasan, J. Billeter and D. Bonvin. On the use of extents for process monitoring and fault diagnosis, 14th Conf. on Chemometrics in Analytical Chemistry, Richmond (USA), 2014.

[6] S. Narasimhan and C. Jordache. *Data Reconciliation and Gross Error Detection*, Elsevier, 1999.

[7] O. A. Asbjørnsen. Reaction invariants in the control of continuous chemical reactors, Chem. Eng. Sci., 27, 709-717, 1972.

[8] B. Srinivasan, M. Amrhein, and D. Bonvin. Reaction and flow variants/invariants in chemical reaction systems with inlet and outlet streams, AIChE J., 44(8), 1858-1867, 1998.

[9] D. Rodrigues, S. Srinivasan, J. Billeter and D. Bonvin. Variant and invariant states for reaction systems, Comput. and Chem. Eng., 73, 23-33, 2015.

[10] S. Srinivasan, J. Billeter, S. Narasimhan and D. Bonvin. Data reconciliation in reacting systems using the concept of extents, 25th European Symp. on Computer Aided Process Engineering, Copenhagen (Denmark), 2015.