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DESIGN OF AN INTERVAL OBSERVER FOR EXOTHERMIC FED-BATCH PROCESSES

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Abstract: This paper proposes an interval observer for continuous or fed-batch chemical reactors with uncertainties related to both the heat exchange coefficient and the kinetics. The interval observer provides an upper and a lower bound of the state given bounds for the uncertain parameters.
The under- and over-estimators are state observers for the non-linear reactor model with two positive tuning parameters that guarantees that the estimated interval contains the state and that the interval width is limited.
The observer performance are illustrated with the production of a resin in an industrial fed-batch reactor.

Keywords: State observers, batch processes, interval observers

1. INTRODUCTION

Monitoring the components concentrations is a key question for productivity and safety in the chemical industry. However, in most cases the concentrations cannot be measured in real-time because suitable devices do not exist or are very expensive. The concentrations are then determined via off-line analyses. The use of state observers to estimate the concentration presents several benefits in this context. They provide a real-time value without requiring any specific device nor any human intervention. This can explain why the design and application of state observers in chemical and biochemical processes has been an active research area over the past decades (Dochain, 2003).

The industrial processes can be divided in two classes : the continuous processes and the batch / fed-batch processes. Although the continuous processes have several advantages due to their steady-state operating mode, fed-batch processes are an interesting option due to their flexibility to be used for the production of different products. This explain why fed-batch reactors are frequently encountered in the biochemical, pharmaceutical and chemical industry.

State estimation in fed-batch processes is subject to difficulties inherent to their operating mode. In particular the continuously changing operating point can become a limitation for the application of common state observation techniques based on the linearized model of the system (Agarwal and Bonvin, 1989), (Vallière and Bonvin, 1989). Furthermore the physical properties of the reacting fluid continuously evolve from those of the initial reactant to those of the final product. This badly known evolution can be at the origin of large uncertainties in the model parameters. For instance the viscosity of a reacting mixture can largely increase during a resin synthesis inmplying the decrease of the heat exchange coefficient. In this case the parameters values are uncertain but not unknown and in most cases the uncertain

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parameters values can be bounded so that they are guaranteed to lie within a given interval.

The interval observer concept has been introduced by (Kieffer *et al.*, 1998). It consists of providing a set estimate guaranteed to contain all the possible state values consistent with the observations given the uncertainties bounds and a set containing the initial state values. In practice, the interval observer can be seen as a couple of state estimators that respectively provides upper and lower bounds for the unmeasured state variables (Gouzé *et al.*, 2000).

Interval observers have been recently developed and applied to continuous wastewater treatment processes where the main uncertainties are related to the kinetics and the process inputs (Alcaraz-Gonzales *et al.*, 2002) and (Rapaport and Dochain, 2005). In both cases, the authors consider the change of coordinates at the origin of the asymptotic observer (Bastin and Dochain, 1990) to remove the dependence with respect to the kinetics. Then they compute two estimates that are guaranteed to bound the state taking advantage of cooperativity properties of the reconstruction error dynamics.

In this paper, we propose an interval observer for exothermic fed-batch reactors that deals with uncertainties related to both the kinetics and the heat exchange coefficient. The observer we propose gather two individual biased state observers based on the nonlinear process model and does not require any change of coordinates. Both provided estimates are guaranteed to bound the state by considering invariant sets and their convergence to a neighborhood of the state is showed using a Lyapunov-like function.

The paper is organized as follows. The first section introduces the reactor model and the interval observer design. Then the second section deals with the convergence analysis of the observer. Finally, the third section present application results of the interval observer to an industrial fed-batch reactor.

2. OBSERVER DESIGN

Let us consider an exothermic reactor whose dynamical model is obtained from mass and energy balances:

$$\dot{T} = \frac{Q}{V}(T_{in} - T) - \frac{\Delta H}{\rho c_p} r^* + \frac{UA}{\rho c_p V}(T_j - T) \quad (1)$$

$$\dot{C} = \frac{Q}{V}(C_{in} - C) - r^* \quad (2)$$

where T, C, Q, T_{in}, C_{in}, U, A, T_j, V, ρ, c_p, r^* and ΔH are the reactor temperature (K), the concentration of the limiting reactant (mol/m^3), the feed flowrate (m^3/s), the inlet temperature (K), the inlet concentration of the limiting reactant (mol/m^3), the overall heat exchange coefficient $(W/m^2/K)$, the heat exchange area (m^2), the cooling fluid temperature (K), the volume (m^3), the density (kg/m^3), the specific heat $(J/kg/K)$, the reaction rate $(mol/m^3/s)$ and the reaction heat (J/mol), respectively.

Let us also consider that the reactor temperature T is measured on-line and that we have to estimate the limiting reactant concentration C while the main sources of model uncertainties are related to the kinetics model and to the global heat exchange coefficient value.

The kinetics model r^* is commonly expressed as the product of a kinetics constant following the Arrhenius law by a function of the reactant concentrations r:

$$r^* = k_0 e^{-E/RT} r \quad (3)$$

where k_0, R and E are the kinetics constant, the gas constant $(J/mol/K)$ and the activation energy (J/mol), respectively. In practice, the kinetics model is identified from experimental results assuming a single global chemical reaction while the actual chemical reaction is the result of different consecutive and parallel elementary reactions between the different functional groups of the molecules. The kinetics law is therefore an approximation of the real reaction rate but it can be assumed that all uncertainties are gathered in the kinetics constant which is bounded as follows:

$$k_{0_{min}} < k_0 < k_{0_{max}} \quad (4)$$

The second source of uncertainties are related to the global heat exchange coefficient because this one can vary with time. These changes are quite limited for continuous processes where they can be due e.g. to the fouling of the heat exchanger tubes but they can become important in fed-batch processes due to e.g. the increase of the heat exchange area but also due to the evolution of the reacting fluid viscosity. Therefore the uncertainties are not only related to the heat exchange coefficient U but to the product UA which is bounded as follows:

$$UA_{min} < UA < UA_{max} \quad (5)$$

The interval observer presented in the following consists of two state observers that converge respectively toward an upper and a lower bound of the state. It is worth emphasizing that each observer incorporates the bounds of both parameters. The interval observer equations are :

- for the lower bound:

$$\dot{\hat{T}}^{\ominus} = \frac{Q}{V}\left(T_{in} - \hat{T}^{\ominus}\right) - \frac{\Delta H}{\rho c_p} k_{0_{min}} e^{-E/RT} r^{\ominus} + \frac{\underline{UA}}{\rho c_p V}\left(T_j - \hat{T}^{\ominus}\right) + \lambda_1^{\ominus}\left(T - \hat{T}^{\ominus}\right) \quad (6)$$

$$\dot{\hat{C}}^{\ominus} = \frac{Q}{V}\left(C_{A,in} - \hat{C}^{\ominus}\right) - k_{0_{max}} e^{-E/RT} r^{\ominus} + \lambda_2^{\ominus}\left(T - \hat{T}^{\ominus}\right) \quad (7)$$

- for the upper bound:

$$\dot{\hat{T}}^{\oplus} = \frac{Q}{V}\left(T_{in} - \hat{T}^{\oplus}\right) - \frac{\Delta H}{\rho c_p} k_{0_{max}} e^{-E/RT} r^{\oplus} + \frac{\overline{UA}}{\rho c_p V}\left(T_j - \hat{T}^{\oplus}\right) + \lambda_1^{\oplus}\left(T - \hat{T}^{\oplus}\right) \quad (8)$$

$$\dot{\hat{C}}^{\oplus} = \frac{Q}{V}\left(C_{A,in} - \hat{C}^{\oplus}\right) - k_{0_{min}} e^{-E/RT} r^{\oplus} + \lambda_2^{\oplus}\left(T - \hat{T}^{\oplus}\right) \quad (9)$$

where $\hat{T}^{\ominus}$, $\hat{C}^{\ominus}$, $\hat{T}^{\oplus}$ and $\hat{C}^{\oplus}$ are the under-estimate for the temperature (K), the under-estimate for the concentration (mol/m^3), the over-estimate for the temperature (K) and the over-estimate for the concentration (mol/m^3), respectively. And with

$$r^{\oplus} = r(C^{\oplus}) \quad (10)$$

$$r^{\ominus} = r(C^{\ominus}) \quad (11)$$

and

$$\overline{UA} = UA_{max} \text{ and } \underline{U} = UA_{min} \text{ if } T_j \geq T \quad (12)$$

$$\underline{UA} = UA_{min} \text{ and } \underline{U} = UA_{max} \text{ if } T_j \geq T \quad (13)$$

and where the observer gains $\lambda_1^{\oplus}$,$\lambda_2^{\oplus}$, $\lambda_1^{\ominus}$ and $\lambda_2^{\ominus}$ are computed as follows:

$$\lambda_1^{\ominus} = \omega^{\ominus} - \frac{Q}{V} - \frac{\underline{UA}}{\rho c_p V} - \frac{\Delta H}{c_p}\sigma^{\ominus} \quad (14)$$

$$\lambda_2^{\ominus} = -\sigma^{\ominus} \quad (15)$$

$$\lambda_1^{\oplus} = \omega^{\oplus} - \frac{Q}{V} - \frac{\overline{UA}}{\rho c_p V} - \frac{\Delta H}{c_p}\sigma^{\oplus} \quad (16)$$

$$\lambda_2^{\oplus} = -\sigma^{\oplus} \quad (17)$$

where $\omega^{\ominus}$, $\omega^{\oplus}$, $\sigma^{\ominus}$ and $\sigma^{\oplus}$ are positive tuning constants.

3. CONVERGENCE ANALYSIS

The convergence analysis of the interval observer is achieved via the convergence analysis of each of the over- and under-estimator. For each case we proceed in two steps. First we show that the under-estimator (resp., over-estimator) provides an estimate that remains lower (resp., greater) than the actual state. Secondly, we show that the estimate reaches a neighborhood of the state and remains inside this neighborhood indefinitely.

Theorem 3.1. Assuming r is an increasing function of C and $Q > 0$, the dynamical system described by equations (6) and (7) (resp. (8) and (9)) with initial conditions satisfying

$$T^{\ominus}(0) \leq T(0) \text{ (resp.} T^{\oplus}(0) \geq T(0)) \quad (18)$$

$$C^{\ominus}(0) \leq C(0) \text{ (resp.} C^{\oplus}(0) \geq C(0)) \quad (19)$$

is an under-estimator (resp. over-estimator) for the system described by equations (1) and (2)

Proof

Only the proof for the under-estimate is provided here since the proof for the over-estimator is similar. The first part of the proof shows that the state variables following the dynamics (6) and (7) remains lower than the state variables of the system given by (1) and (2). This goal is achieved by showing that the reconstruction errors remains positive.

Let us define the reconstruction error x associated to the under-estimator by

$$x_1 = T - \hat{T}^{\ominus} \quad (20)$$

$$x_2 = C - \hat{C}^{\ominus} \quad (21)$$

It follows from equations (1), (2), (6) and (7), that the reconstruction errors are governed by the following dynamical equations :

$$\dot{x}_1 = -\omega x_1 + \frac{\Delta H}{c_p}\sigma x_1 + \theta - \frac{\Delta H}{\rho c_p}\left(k_0 e^{-E/RT} r - k_{0_{min}} e^{-E/RT} r^{\ominus}\right) \quad (22)$$

$$\dot{x}_2 = \sigma x_1 - \frac{Q}{V}x_2 - \left(k_0 e^{-E/RT} r - k_{0_{max}} e^{-E/RT} r^{\ominus}\right) \quad (23)$$

where θ is defined as follows

$$\theta = \frac{UA - \underline{UA}}{\rho c_p V}\left(T_j - T\right) \quad (24)$$

and is non-negative by definition of $\underline{UA}$ (13).

Let us define the subset U of R^2 as the set of the non-negative reconstruction errors:

$$U = \left\{(x_1, x_2) \in \mathrm{R}^2 : x_1 \geq 0, x_2 \geq 0\right\} \quad (25)$$

and show that this set is an invariant set for the system described by (22) and (23). The set U has a boundary ∂U which can be splitted into two subsets defined as follows:

$$\partial U_1 = \left\{(0, x_2) \in \mathrm{R}^2 : x_2 \geq 0\right\} \quad (26)$$

$$\partial U_2 = \left\{(x_1, 0) \in \mathrm{R}^2 : x_1 \geq 0\right\} \quad (27)$$

such that

$$\partial U = \partial U_1 \cup \partial U_2 \tag{28}$$

Now let us choose an arbitrary point P of the boundary of U, then we necessarily have either

$$P \in \partial U_1 \tag{29}$$

or

$$P \in \partial U_2 \tag{30}$$

Let us assume that $P \in \partial U_1$, then $x_1 = 0$ and Equation (22) becomes

$$\dot{x}_1 = \theta - \frac{\Delta H}{\rho c_p} e^{-E/RT} \left(k_0 r - k_{0_{min}} r^{\ominus} \right) \tag{31}$$

As $P \in U$, we have $\hat{C} \geq C$ so that

$$k_0 e^{-E/RT} r - k_{0_{min}} e^{-E/RT} r^{\ominus} \geq 0 \tag{32}$$

because r is an increasing function of C. This shows that the second term of Equation (31) is positive because the reaction is exothermic ($\Delta H < 0$). As by definition (24) θ is non-negative, it can be stated that for any point $P \in \partial U_1$ we have:

$$\dot{x}_1 > 0 \tag{33}$$

so that the boundary ∂U_1 is never crossed by the error dynamical system (22) and (23).

Similarly, let us assume that $P \in \partial U_2$, then $x_2 = 0$ and $r = r^{\ominus}$ and Equation (23) becomes

$$\dot{x}_2 = \sigma x_1 \tag{34}$$

so that we have for any point $P \in \partial U_2$:

$$\dot{x}_2 \geq 0 \tag{35}$$

showing that the boundary ∂U_2 is never crossed.

It can be concluded from the above reasoning that the subset U is an invariant set for the dynamical system described by (22) and (23). As by assumptions (18) (19), the initial reconstruction errors lies in U

$$x_1(0) \geq 0 \tag{36}$$
$$x_2(0) \geq 0 \tag{37}$$

it can be concluded that the reconstruction error remains positive.

The second part of the proof shows that the reconstruction error converges toward a neighborhood of the origin and remains indefinitely in this neighborhood.

As we have shown that the reconstruction error x remains positive, we can limit the domain of our analysis to the set U. Let us define the following function

$$V(\epsilon) = x_1 - \frac{\Delta H}{\rho c_p} x_2 \tag{38}$$

The above function is an increasing function that is positive everywhere on U except on the origin :

$$V(0) = 0 \tag{39}$$
$$V(x) > 0 \ \forall x \in U \backslash 0 \tag{40}$$
$$\frac{\partial V}{\partial x_1} = 1 \ \forall x \in U \tag{41}$$
$$\frac{\partial V}{\partial x_2} = -\frac{\Delta H}{\rho c_p} \ \forall x \in U \tag{42}$$

The time derivative of V can be computed using (22) and (23):

$$\dot{V} = \theta - \omega x_1 + \frac{\Delta H}{\rho c_p} \frac{Q}{V} x_2 - \frac{\Delta H}{c_p} \left(k_{0_{max}} - k_{0_{min}} \right) e^{-E/RT} r^{\ominus} \tag{43}$$

Assuming that θ, Q, $r^{\ominus}$, T and V are bounded as follows :

$$\theta(t) < \theta_{max} \tag{44}$$
$$V(t) < V_{max} \tag{45}$$
$$r^{\ominus} < r^{\ominus}_{max} \tag{46}$$
$$Q(t) > Q_{min} \tag{47}$$
$$T(t) > T_{min} \tag{48}$$

so that we consider a function W such that

$$\dot{W} = \theta_{max} - \omega x_1 + \frac{\Delta H}{\rho c_p} \frac{Q_{min}}{V_{max}} x_2 - \frac{\Delta H}{c_p} \left(k_{0_{max}} - k_{0_{min}} \right) e^{-E/RT} r^{\ominus} \tag{49}$$

Then, for any $x \in U$ we have:

$$\dot{V}(x) \leq \dot{W}(x) \tag{50}$$

Let us now define the set S by:

$$S = \left\{ x \in U : \dot{W}(x) \geq 0 \right\} \tag{51}$$

which is a non-empty closed set containing at least the origin. This allows to define two complementary sets :

$$P = x \in U : V(x) \geq \nu \tag{52}$$
$$P^{\perp} = x \in U : V(x) > \nu \tag{53}$$

where

$$\nu = \max_{x \in S} V(x) \tag{54}$$

As we have for any point of $P^{\perp}$

$$\forall x \in P^{\perp} : \dot{V}(x) \leq \dot{W}(x) < 0 \tag{55}$$

this shows that any trajectory starting in $P^{\perp}$ finally converges toward P. Furthermore, we have by definition :

$$U = P \cup P^{\perp} \tag{56}$$

Therefore any trajectory governed by (22) and (23) and starting in U converges to the closed set P and remains inside this set indefinitely.

□

4. APPLICATION

Let us consider the industrial production of a resin P from two reactants A and B following the exothermic chemical reaction

$$A + bB \rightarrow P \tag{57}$$

where $b > 0$ is a stoichiometric coefficient and for which the reaction rate is given by

$$r^* = k_0 e^{-E/RT} C_A^{p_1} C_B^{p_2} \tag{58}$$

with p_1 and p_2 two non-negative constants such that $p_1 + p_2 \neq 0$ and where C_A, C_B, T, k_0, E and R stand for the concentration in reactant A (mol/m^3), the concentration in reactant B (mol/m^3), the temperature (K), the kinetics constant$(mol^{1-p_1-p_2}\ m^{-3(1-p_1-p_2)}\ s^{-1})$, the activation energy (J/mol) and the gas constant $(J/mol/K)$, respectively.

This synthesis is carried out in the fed-batch operating mode as follows : the reactant A is progressively added to the reactor that initially contains the reactant B. This operating mode allows to control the temperature rise by limiting the amount of reactant A available in the reacting fluid, therefore the reactant A is also called the limiting reactant. Such a reactor can be modeled by the following set of four differential equations :

$$\dot{T} = \frac{Q}{V}(T_{in} - T) - \frac{\Delta H}{\rho c_p} r^* + \frac{UA}{\rho c_p V}(T_j - T) \tag{59}$$

$$\dot{C}_A = \frac{Q}{V}(C_{A,in} - C_A) - r^* \tag{60}$$

$$\dot{C}_B = -\frac{Q}{V} C_B - b r^* \tag{61}$$

$$\dot{V} = Q \tag{62}$$

where $Q, T_{in}, C_{A,in}, U, A, T_j, V, \rho, c_p$ and ΔH are the feed flowrate (m^3/s), the inlet temperature (K), the inlet concentration of A (mol/m^3), the overall heat exchange coefficient $(W/m^2/K)$, the heat exchange area (m^2), the cooling fluid temperature (K), the volume (m^3), the density (kg/m^3), the specific heat $(J/kg/K)$ and the reaction heat (J/mol), respectively.

Let us consider that the reactor volume and the initial concentration of B are known. Then the reactor model can be reduced to a second order model using a material balance between the initial time t_0 and the present time t. Considering the stoichiometry of reaction (57), it can be computed that for any time t :

$$C_B(t) = b C_A(t) + \frac{1}{V(t)} \cdot \left(V(t_0) C_B(t_0) - b \int_{t_0}^{t} Q(\tau) C_{A,in} d\tau \right) \tag{63}$$

Introducing the above results into Equation (58) leads to an expression with the following structure

$$r^* = k_0 e^{-E/RT} r(C_A, V, Q) \tag{64}$$

where r is a non-negative increasing function of C_A

$$r \geq 0 \tag{65}$$

$$\frac{\partial r}{\partial C_A} \geq 0 \tag{66}$$

The reactor model can finally be written as follows:

$$\dot{T} = \frac{Q}{V}(T_{in} - T) - \frac{\Delta H}{\rho c_p} r^* + \frac{UA}{\rho c_p V}(T_j - T)$$

$$\dot{C}_A = \frac{Q}{V}(C_{A,in} - C_A) - r^*$$

so that the interval observer described by equations (6) to (9) can be used to estimate the concentration of the limiting reactant.

This interval observer has been applied to data coming from an industrial resin synthesis carried out in a fed-batch reactor of several tons in which the chemical heat generation is about 25 W/kg. An interval of $\pm 20\%$ around a nominal value has been imposed for the global heat exchange coefficient and an interval of $\pm 10\%$ around a nominal value has been imposed for the kinetics constant. The tuning parameters for both under- and over-estimators have been respectively chosen identical.

$$\omega = \omega^{\ominus} = \omega^{\oplus} \tag{67}$$

$$\sigma = \sigma^{\ominus} = \sigma^{\oplus} \tag{68}$$

If σ is set to zero, the concentration estimation ((7) and (9)) is totally decoupled from the energy balance ((6) and (8)) and no information

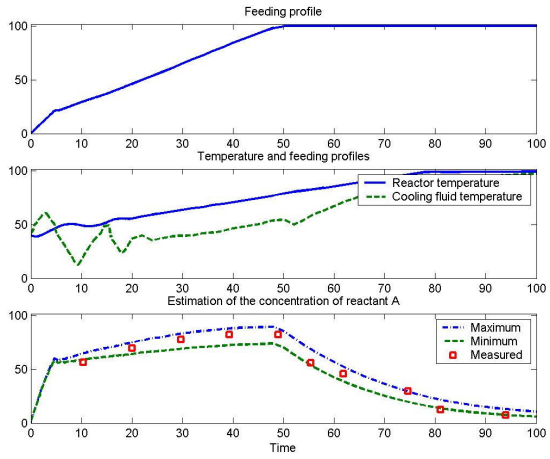

Fig. 1. Application of the interval observer to an industrial exothermic reactor

are retrieved from the temperature evolution. In this case, the interval width only depends on the kinetics constant uncertainties. Increasing the value of σ gives more influence to the energy balance and makes the estimation robuster with respect to the kinetics model, however the interval width increases and depends on the heat exchange uncertainty. The parameters σ and ω have been tuned by simuylations as follows:

$$\sigma = 0.2 \tag{69}$$

$$\omega = 50 \tag{70}$$

The results are shown on Figure 1 where it can be seen that the experimental data lie inside the estimated interval which has a narrow width.

5. CONCLUSION

In this paper we have designed an interval observer to estimate the limiting component concentration in continuous or fed-batch exothermic chemical reactors with uncertainties concerning both the kinetics and the heat exchange coefficient. This interval observer consists in two parallel state observers providing two estimates that are guaranteed to be respectively lower and greater than the state assuming the reaction rate is an increasing function of the concentration.

The performance of the interval observer have been illustrated with an application example related to the production of a resin in an industrial fed-batch reactor.

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