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Parametric uncertainty and disturbance attenuation in the suboptimal control of a non-linear electrochemical process

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SUMMARY

The optimal control of the hydrogen evolution reactions is attempted for the regulation and change of set-point problems, taking into account that model parameters are uncertain and I/O signals are corrupted by noise. Bilinear approximations are constructed, and their dimension eventually increased to meet accuracy requirements with respect to the trajectories of the original plant. The current approximate model is used to evaluate the optimal feedback through integration of the Hamiltonian equations. The initial value for the costate is found by solving a state-dependent algebraic Riccati equation, and the resulting control is then suboptimal for the electrochemical process. The bilinear model allows for an optimal Kalman–Bucy filter application to reduce external noise. The filtered output is reprocessed through a non-linear observer in order to obtain a state-estimation as independent as possible from the bilinear model. Uncertainties on parameters are attenuated through an adaptive control strategy that exploits sensitivity functions in a novel fashion. The whole approach to this control problem can be applied to a fairly general class of non-linear continuous systems subject to analogous stochastic perturbations. All calculations can be handled on-line by standard ordinary differential equations integration software. Copyright © 2007 John Wiley & Sons, Ltd.

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1. INTRODUCTION

The hydrogen evolution reactions (HER) system has received increasing attention in recent literature due to its basic role in modelling the dynamics of electrochemical devices. These reactions were widely used to study and experiment on water electrolysis and batteries' dynamics [1], but since the opposite directions of the kinetics can occur upon changing conditions, the same equations also model hydrogen consuming processes as in fuel-cells-type

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reactors [2]. Also H_2 -decontamination and corrosion prevention processes for heavy metals [3], and cold nuclear fusion [4, 5], include HER mechanisms in their dynamics.

The structure of the HER system is non-linear. Experiments show qualitative behaviours only possible for non-linear systems (see the extensive review by Hudson and Tsotsis [6]). So, usual linearizations around steady states are insufficient to reflect the characteristics of the flow for significative departures from a single equilibrium point, as those occurring in change of set-point operations. Moreover, for control systems it is well known that the same accuracy on trajectories for all admissible input functions cannot been guaranteed during a fixed period of time, however small [7].

In its simplest form, the HER system is a combination of several chemical reactions conducting to hydrogen deposition on the electrode of most electrochemical devices, and its ulterior desorption to reach molecular gaseous form. The model adopted here reduces to a series of three 'steps', each one known after the names of the distinguished chemists Volmer–Heyrovsky–Tafel (see [6, 8]), or globally as the VHT reaction mechanisms.

The control of fuel cells operation has begun to be studied recently, specially for non-isothermal proton exchange membrane prototypes (see [9, 10], and the references therein). In all cases dynamic non-linearities have been confirmed experimentally, which suggests that fuel cell technology would require non-linear control techniques for conducting changes in their operational conditions. Other aspects of control of electrochemical reactions attempt to avoid chaotic behaviour [11, 12], or to minimize the dissipation of electrical power during the process control [13].

Concern has also arisen with respect to the stochastic aspects of the problem, mainly the uncertainties on the parameters of the system and noise corruption of input/output signals, both approached in this paper. For the electrochemical system under study the output does not coincide with the state, and therefore an implicit stochastic tracking objective is involved, which in principle should be solved in the non-linear context. This problem has been addressed [14] through power series expansions of the Hamilton–Jacobi–Bellman equation, and by rearranging the resulting terms to construct two suboptimal (state and output) feedbacks. Unfortunately such an approach requires system stability in the whole range of the process, that in general cannot be guaranteed for changes of set-point requests. Also, it assumes conditions of invertibility or non-vanishing of coefficients on the matrices of the approximation, which neither can be assumed in the present case, above all when increasing the dimension of the model becomes necessary.

For these reasons, in this paper a novel approach to the combined problem of updating parameters and filtering noise is designed. A non-linear continuous updating algorithm through sensitivity matrices is devised along the course of non-linear least-squares strategies (see [15]), but using state instead of output deviations as the relevant data acquired on-line. Therefore, the design of a filter and an observer, both non-linear, becomes necessary. This approach is chosen given the highly non-linear control-dependent character of the observation function from one side, and the relatively simple initial-value dynamics governing the sensitivity matrices and the filtering problem for the bilinear model on the other.

Summarizing, the main contributions reside in (i) the development of new formulae for the continuous updating of coefficients in non-linear dynamics, based on sensitivity matrices; (ii) the algorithm to refine approximate realizations by adding only two new coefficients to the model matrices for each dimension increase; and (iii) the combination of these new results with non-linear optimal control, observers and filtering techniques to produce a control strategy robust

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with respect to signal noise and parameter uncertainty. This strategy is also suboptimal with respect to a classical quadratic cost and may be constructed completely on-line.

The organization of the paper is as follows: Section 2 describes the electrochemical system, its bilinear approximation near equilibrium points, and the optimal control objectives imposed, and also there are two subsections describing the Hamiltonian approach to the infinite-horizon suboptimal control solution, for the cases of regulation and changes of set-point. Section 3 shows how the sensitivity functions are employed in updating parameters of the bilinear approximations. Section 4 describes the method for increasing the dimension (but keeping the bilinear structure) of the approximating model before accuracy failures, and Section 5 the implementation of a Kalman–Bucy filter to attenuate disturbances in I/O signals, in parallel with a non-linear observer used to update the linear parameter of the filter. All steps are combined in an on-line calculation strategy, shown in a flowchart and explained in Section 6. Conclusions and final comments are included in Section 7.

2. SYSTEM DYNAMICS AND APPROXIMATIONS. THE SUBOPTIMAL CONTROL PROBLEM

The VHT reactions and their kinetics are modelled as (see for instance [16])

$$\text{Volmer}: H_2O + e^- \rightleftharpoons H_{(ads)} + OH^-$$

$$v_V = v_V^e \left\{ \frac{1-\theta}{1-\theta_e} e^{-(1-\alpha) f\eta} - \frac{\theta}{\theta_e} e^{\alpha f \eta} \right\} \tag{1}$$

$$\text{Heyrovsky}: H_2O + H_{(ads)} + e^- \rightleftharpoons H_{2(g)} + OH^-$$

$$v_H = v_H^e \left\{ \frac{\theta}{\theta_e} e^{-(1-\alpha) f\eta} - \frac{1-\theta}{1-\theta_e} e^{\alpha f \eta} \right\} \tag{2}$$

$$\text{Tafel}: H_{(ads)} + H_{(ads)} \rightleftharpoons H_{2(g)}$$

$$v_T = v_T^e \left\{ \left(\frac{\theta}{\theta_e}\right)^2 - \left(\frac{1-\theta}{1-\theta_e}\right)^2 \right\} \tag{3}$$

By taking all three routes into account, and assuming that the electrode's surface coverage is proportional to the number of atoms of $H_{(ads)}$, then the HER stoichiometric balance translates into a θ accumulation rate equation

$$\dot{\theta} = \frac{F}{\sigma}(v_V - v_H - 2v_T), \quad (\triangleq g(\theta, \eta)) \tag{4}$$

where the main variables involved are: θ is the surface coverage (the fraction of the electrode surface covered by adsorbed atomic hydrogen $H_{(ads)}$); $\dot{\theta}$ denotes the time (t) derivative of θ (t in seconds); and η, the overpotential imposed on the system to run the reaction. The variable η will be identified with the control input of the system. Other symbols and parameters used mean:

$H_{2(g)}$, gaseous (desorbed) molecular hydrogen; θ_e, specific equilibrium surface coverage ($\theta_e = 0.1$ in numerical calculations and graphics of this paper); α, adsorption symmetric factor

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(= 0.5 in calculations); **R**, gas constant = $8.3145\,\mathrm{J\,mol^{-1}\,K^{-1}}$; F, Faraday constant = 96484.6 $\mathrm{C\,mol^{-1}}$; T, absolute temperature, here taken equal to 303.15 K; $f = F/\mathbf{R}T = 38.2795\,\mathrm{C\,J^{-1}}$; and σ is the experimentally measured surface density of electric charge needed to complete a monolayer coverage of adsorbed hydrogen atoms. In the numerical results of this paper a value of $\sigma = 2.21 \times 10^{-4}\,\mathrm{C\,cm^{-2}}$ will be adopted, corresponding to a standard Pt electrode (see [8]), and

v_V^e, v_H^e, v_T^e: equilibrium reaction rates of each step. The nominal values for these parameters were chosen as to produce a dynamic response in the order of seconds, and at the same time to maintain both the Volmer–Heyrovsky and the Volber–Tafel reaction routes active (see [17])

$$v_T^e = 10^{-12}\,\mathrm{mol\,cm^{-2}\,s^{-1}}$$

$$v_H^e = 10^{-13}\,\mathrm{mol\,cm^{-2}\,s^{-1}}$$

$$v_V^e = 10^{-10}\,\mathrm{mol\,cm^{-2}\,s^{-1}}$$

Equation (4) (after the expressions of the three velocities are inserted) reflects the non-linearity of the problem. The generating function g takes the form of a second order polynomial on the state variable θ when controls are kept constant. This quadratic dependence may result in undesirable high velocities when the Tafel step dominates, and theoretically an explosive behaviour is possible in finite time, which never happens in linear systems. Also, the θ^2 terms are responsible for the hysteresis loops appearing for sawtooth-like forcings applied to the overpotential η (see [13, 14]), that cannot be handled with linear approximations. The exponential dependence of g on η shows that all powers of the control values will affect the dynamics to some extent. No simplifying change of variables of the type $\eta \to \mu = e^{\kappa\eta}$ or similar has been found to render Equation (4) into an affine (linear plus a constant term) expression on such artificial control μ. Therefore, in this paper a non-linear system point of view will be pursued, and the control effort will be present in the objectives to optimize.

In general, optimal control problems do not have close solutions for non-linear systems Σ and arbitrary cost objectives $\mathscr{J}$. The subclass of bilinear systems $\tilde{\Sigma}$ may be used as universal approximations to non-linear systems in the following sense: if the admissible values for the control variable $u(.)$ are bounded during a fixed finite period of time $T_a = [0, t_a]$, then a bilinear system $\tilde{\Sigma}$ can be found such that its state trajectories differ from the trajectories of the original Σ (corresponding to the same control functions applied during the period T_a) in less than an arbitrary tolerance tol > 0, fixed *a priori* (see [18, 19]). It is well-known that a similar result cannot be obtained by using linear approximations, and since it is crucial to count with a period T_a where the quality of the approximation is guaranteed, in this paper bilinear models will be used. 'Suboptimal control' will then mean in this context that the optimal control strategies constructed for a bilinear approximation $\tilde{\Sigma}$ and a given cost objective $\mathscr{J}$, will only approximate the solution to the optimal control problem for $(\Sigma, \mathscr{J})$. But the main advantage of using this type of approximations comes from the fact that many control situations have been clarified and solved for the class of bilinear systems, as it is the case for the optimal bilinear-quadratic control and the Kalman–Bucy filtering problems, both discussed below.

The original system is one-dimensional, but since the dimension of the approximating model can increase, the notation is kept in matrix form for generality. The simplest one-dimensional bilinear approximation to the VHT system may be constructed as follows. Around an

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equilibrium (a state/control pair (θ_0, η_0) such that $g(\theta_0, \eta_0) = 0$), the dynamics admits a Taylor expansion

$$\begin{aligned} g(\theta, \eta) = g(\theta_0, \eta_0) &+ \left[\frac{\partial g}{\partial \theta}(\theta_0, \eta_0)\right](\theta - \theta_0) + \left[\frac{\partial g}{\partial \eta}(\theta_0, \eta_0)\right](\eta - \eta_0) \\ &+ \frac{1}{2!}\left\{\left[\frac{\partial^2 g}{\partial \theta^2}(\theta_0, \eta_0)\right](\theta - \theta_0)^2 + \left[\frac{\partial^2 g}{\partial \eta^2}(\theta_0, \eta_0)\right](\eta - \eta_0)^2 \right. \\ &\left. + 2\left[\frac{\partial^2 g}{\partial \theta \partial \eta}(\theta_0, \eta_0)\right](\theta - \theta_0)(\eta - \eta_0)\right\} + \frac{1}{3!}\left\{\left[\frac{\partial^3 g}{\partial \theta^3}(\theta_0, \eta_0)\right](\theta - \theta_0)^3 + \cdots \right. \end{aligned} \tag{5}$$

After calling

$$A \triangleq \frac{\partial g}{\partial \theta}(\theta_0, \eta_0), \quad B \triangleq \frac{\partial g}{\partial \eta}(\theta_0, \eta_0), \quad N \triangleq \frac{\partial^2 g}{\partial \theta \partial \eta}(\theta_0, \eta_0) \tag{6}$$

$$x \triangleq \theta - \theta_0, \quad u \triangleq \eta - \eta_0 \tag{7}$$

then the bilinear system

$$\dot{x} = Ax + Bu + Nxu = \tilde{g}(x, u) \tag{8}$$

is clearly an approximation in the sense that $\tilde{g}(x, u) \approx g(\theta, \eta)$ near the selected equilibrium point.

Figure 1 shows the responses to step controls of different sizes and signs near the equilibrium

$$(\theta_0, \eta_0) = (0.38545805, -0.05)$$

when applied to the non-linear VHT dynamics and to its first bilinear approximation with coefficients

$$A = -363.7724, \quad B = -2714.1, \quad N = -770.1951 \tag{9}$$

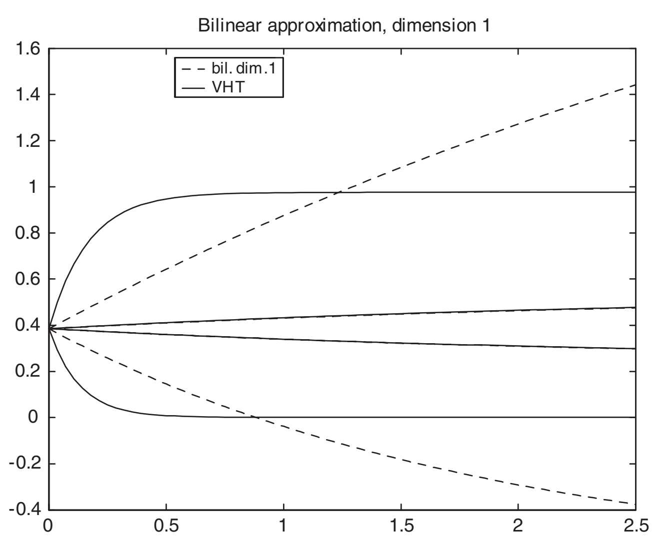

Figure 1. Step responses of the non-linear system and its one-dimensional bilinear approximation, for $\eta = \pm 0.02, \pm 0.095$.

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The resulting trajectories show that for small-size input steps (± 0.02) the one-dimensional bilinear approximation is good enough (the dashed line coincides with the solid line). But for bigger changes (± 0.095) the two state trajectories depart significantly, after a certain time the bilinear response going even beyond the operational range ($\theta \in [0, 1]$). The non-linearity of the system reflects in the sizes of the asymptotic changes for state θ due to changes in η of opposite sign (for $\eta = 0.095$ the change is around 50% bigger than for $\eta = -0.095$).

The VHT system admits a continuous locus of equilibrium points (see [13] for an explicit formula), all of them with negative coefficients A but with sign-changing coefficients N.

2.1. Regulation

The optimal control of bilinear systems under a quadratic criterion and an infinite horizon

$$\mathscr{J}(u) = \int_0^\infty (Qx^2 + Ru^2)\,\mathrm{d}t \tag{10}$$

will be posed and solved in what follows. Clearly the desired state is the origin, so the problem consists in abating perturbations $x_0 = \theta(0) - \theta_0 \neq 0$ with optimal cost. The usual assumptions $Q \geqslant 0, R > 0$, are kept. The control will remain unidimensional, but higher dimensions for the state will be allowed to take into account an eventual increase on the dimension of the matrices for better accuracy (discussed in Section 4 below).

See [20] for the foundations of the initial-value Hamiltonian equations for this problem, namely

$$\dot{x} = \frac{\partial \mathscr{H}^0}{\partial \lambda} = Ax - \frac{1}{2R}(B + Nx)(B + Nx)'\lambda, \quad x(0) = x_0 \tag{11}$$

$$\dot{\lambda} = -\frac{\partial \mathscr{H}^0}{\partial x} = -2Qx - A'\lambda + \frac{1}{2R}N'\lambda(B + Nx)'\lambda, \quad \lambda(0) = 2\pi(x_0)x_0 \tag{12}$$

where $\mathscr{H}^0(x, \lambda)$ is the Hamiltonian of the system along the optimal trajectory, and λ is the adjoint or costate variable, that in the bilinear case verifies

$$\lambda = 2\pi(x)x \tag{13}$$

with $\pi(x)$ denoting the unique positive-definite solution to the state-dependent Riccati equation

$$Q + \pi(x)A + A'\pi(x) - \pi(x)\frac{(B + Nx)(B + Nx)'}{R}\pi(x) = 0 \tag{14}$$

Since the Hamiltonian equations can be solved on-line in this case, then the optimal control can be evaluated in a state/costate feedback fashion

$$u = -\frac{1}{2R}(B + Nx)'\lambda, \quad \eta = \eta_0 + u \tag{15}$$

Figure 2 illustrates the non-linear and bilinear state evolutions corresponding to the optimal control calculated through Equation (15). Calculations were performed with the following values of the cost-weight parameters: $Q = 1$; $R = 5$. Initial state perturbation: 0.05 from initial coverage $\theta_0 = 0.38545805$. Both trajectories reach θ_0 asymptotically, as desired.

Even for small perturbations, the non-linear approach produces control strategies that depart from linear controllers. An illustration of this point is depicted in Figure 3. The 'Linear' horizontal reference represents the proportional gains that would be obtained from the dynamic

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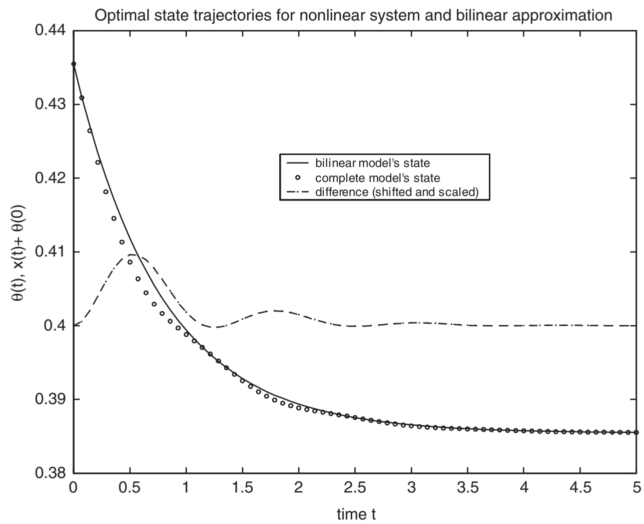

Figure 2. Suboptimal state trajectory for the non-linear system (optimal for the bilinear approximation). The difference between trajectories is shifted to 0.4 and magnified threes times to show in the same picture. Initial perturbation: 0.05.

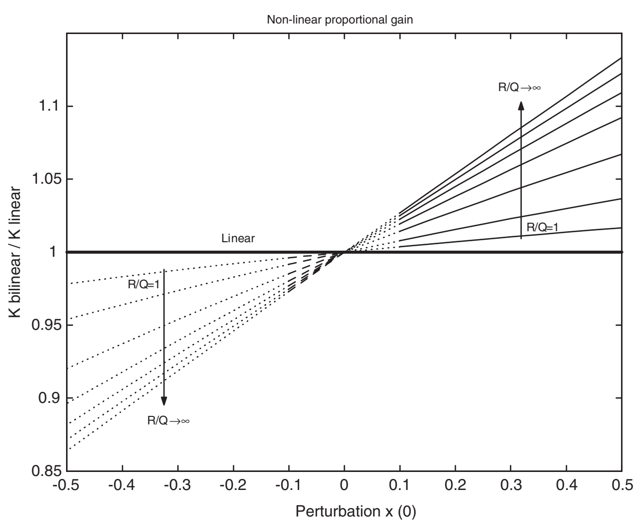

Figure 3. Ratio between the bilinear (apparent) proportional gain and the solution to the linear-quadratic problem (strictly proportional), depending on initial perturbation. Curves for some R/Q values from 1 to 100 are shown.

parameters A, B of the system, different control effort *versus* state accuracy (R/Q) design choices, and the solution to the algebraic Riccati equation corresponding to the approximate linear-quadratic problem. The optimal solution is always a proportional control, with a gain that is independent from the initial perturbation. For the bilinear approximation an 'apparent'

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gain is calculated using the same parameters plus the coefficient N of the non-linear term, and the value of the initial perturbation $x(0)$. For a fixed R/Q value, the ratio between the non-linear and the linear controller gains depends on the initial perturbation and is shown in one of the non-horizontal curves. The procedure is run for increasing R/Q values. Results show that, as the control effort (weighted by R) has greater incidence on the total cost (i.e. as R/Q grows), then the optimal gain for the bilinear approximation (K_{bilinear}) increasingly differs from its linear counterpart (K_{linear}). Therefore, in order to obtain optimal results for this system, non-linear departs from linear control results as the cost of supplying overpotential η grows. Notice that minimizing the overpotential is roughly equivalent to minimizing power expenses when the output current density is kept nearly constant (see [13]).

2.2. Set-point changes

Original equilibrium (θ_0, η_0), with $g(\theta_0, \eta_0) = 0$. Target set-point: $(\bar{x}, \bar{u}) = (\theta_1 - \theta_0, \eta_1 - \eta_0)$ with $g(\theta_1, \eta_1) = 0$.

The problem consists in steering the system from the original equilibrium towards the target set-point while minimizing the deviation quadratic cost

$$\mathscr{J}(u) = \int_0^\infty [Q(x-\bar{x})^2 + R(u-\bar{u})^2]\,\mathrm{d}t \tag{16}$$

The corresponding Hamiltonian differential equations, Riccati algebraic equation and optimal feedback for this problem have also been discussed in [20] and here take the form

$$\dot{x} = Ax + (B+Nx)\bar{u} - \frac{(B+Nx)(B+Nx)'\lambda}{2R}, \quad x(0) = x_0(=0) \tag{17}$$

$$\dot{\lambda} = -2Q(x-\bar{x}) - (A+\bar{u}N)'\lambda + \frac{N'\lambda(B+Nx)'\lambda}{2R}, \quad \lambda(0) = 2\bar{\pi}(x_0)(x_0 - \bar{x}) \tag{18}$$

In this case the costate is related to the state in the form

$$\lambda = 2\bar{\pi}(x)(x-\bar{x}) \tag{19}$$

and $\bar{\pi}(.)$ is the state-dependent solution to

$$Q + \bar{\pi}(x)(A+\bar{u}N) + (A+\bar{u}N)'\bar{\pi}(x) - \bar{\pi}(x)\frac{(B+Nx)(B+Nx)'}{R}\bar{\pi}(x) = 0 \tag{20}$$

The optimal feedback results then

$$u = \bar{u} - \frac{1}{2R}(B+Nx)'\lambda, \quad \eta = \eta_0 + u \tag{21}$$

A numerical example of a set-point change for the bilinear approximation, from $\theta_{\text{equil}}(\eta_0) = \theta_{\text{equil}}(-0.05) = 0.38545805$ towards $\theta_1 \triangleq \theta_{\text{equil}}(\eta_1) = \theta_{\text{equil}}(-0.06) = 0.46030979$ is shown in Figure 4. The value of θ_1 is closely approached by the bilinear system, for which the corresponding equilibrium control value is around $\eta_{1\text{b}} = -0.055$, the asymptotic limit for $u(t)$ in the figure. But the original dynamics has an equilibrium state $\theta_{1\text{b}} = \theta_{\text{equil}}(\eta_{1\text{b}}) = 0.4229$, the asymptotic value for $\theta(t)$ in the figure, different from the desired θ_1. The inaccuracies in the final values ($\theta_{1\text{b}}$ instead of θ_1, and $\eta_{1\text{b}}$ instead of η_1) show the need for parameters' updating, at least for set-point change operations.

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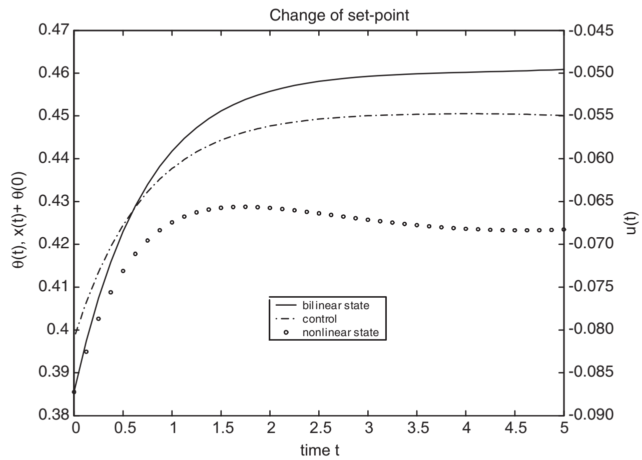

Figure 4. States' trajectories ($\theta(t)$ for the non-linear system and $x(t)+x_0$ for its bilinear approximation), corresponding to a suboptimal control trajectory $u(t)$ for a change of set-point problem. The scale for states' values is on the left y-axis, and for control values in the right y-axis.

3. UPDATING PARAMETERS *VIA* SENSITIVITY FUNCTIONS

Since the analytical expressions of the original and the approximate dynamics (Equations (1)–(4) and (5)–(8), respectively) are smooth, then the existence of smooth local flows (ϕ and $\tilde{\phi}:\mathscr{T}\times\mathscr{X}\times\mathscr{P}\times\mathscr{U}\rightarrow\mathscr{X}$, respectively) are guaranteed (here $\mathscr{T},\mathscr{X},\mathscr{P},\mathscr{U}$ denote the admissible time, state, parameter and control value sets). Uncertainties on the original parameters $v_{\mathrm{V}}^{\mathrm{e}}, v_{\mathrm{H}}^{\mathrm{e}}, v_{\mathrm{T}}^{\mathrm{e}}, \theta_{\mathrm{e}}, \alpha, T$ translate into uncertainties on the parameters p of the bilinear approximation, i.e. on A, B, N. The sensitivities with respect to these parameters p in the context of this section will be defined as

$$S_p \triangleq \frac{\partial\tilde{\phi}}{\partial p} \tag{22}$$

It is well known (see for instance [21, 22]) that the sensitivities are governed by the 'equation on variations'

$$\dot{S}_p = \frac{\partial\tilde{g}}{\partial\theta}S_p + \frac{\partial\tilde{g}}{\partial p}, \quad S_p(0)=0 \tag{23}$$

The use of sensitivities in modelling (see [22]) has been extended to control matters, but specifically in adapting parameter values we propose a generalization of the continuous-case recursive least-squares approach given in [15]. The objective is to minimize the deviation-square

$$\chi^2(t,p) \triangleq \int_0^t \mathrm{e}^{-a(t-\tau)}[\theta(\tau)-\tilde{\theta}(\tau)]^2\,\mathrm{d}\tau \tag{24}$$

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with respect to the parameters p, where a is adequately called 'the forgetting factor'. By applying Leibnitz's rule

$$\frac{\partial \chi^2}{\partial p}(t,p) = -2\int_0^t e^{-a(t-\tau)}[\theta(\tau) - \tilde{\theta}(\tau)]S_p(\tau)\,d\tau \tag{25}$$

and by approximating

$$\tilde{\theta}(\tau) = \tilde{\theta}(\tau,p) \approx \tilde{\theta}(\tau,\bar{p}) + S_p'(\tau)(p-\bar{p}) \tag{26}$$

where $\bar{p}$ is the nominal value of the parameter vector, then the critical value of p (i.e. $\arg(\partial\chi^2/\partial p)(t,p) = 0$) for the time-period $[0,t]$ would be

$$p(t) \triangleq \arg\min \chi^2(t,p) = \bar{p} + P(t)Z(t) \tag{27}$$

where the variables P, Z are notations for

$$P(t) \triangleq \left[\int_0^t e^{-a(t-\tau)} S_p^2(\tau)\,d\tau\right]^{-1} \quad \forall t > 0 \tag{28}$$

$$Z(t) \triangleq \int_0^t e^{-a(t-\tau)}[\theta(\tau) - \tilde{\theta}(\tau,\bar{p})]S_p(\tau)\,d\tau \tag{29}$$

If continuous (on-line) updating of the parameter is desired, then Equation (27) can be differentiated to obtain

$$\dot{p} = PS_p\,\varepsilon \tag{30}$$

where

$$\varepsilon(\tau) \triangleq \theta(\tau) - \tilde{\theta}(\tau, p(\tau)) \tag{31}$$

that should be integrated along with the differential equation satisfied by P, namely

$$\dot{P} = aP - PS_pS_p'P \tag{32}$$

Notice that the initial 'value' of P would be ∞, so Equations (30) and (32) are only used after some small (resetting) time, say $t = \delta$, in order to get a good estimation of $P(\delta) \approx \bar{P}$ from Equation (28). If δ is small, then $p(\delta) = \bar{p}$ and $P(\delta) = \bar{P}$ may be safely taken as initial conditions for the corresponding ordinary differential equations (ODEs).

Figures 5 and 6 illustrate an intent to update parameters during the same change of set-points $x_0 \to x_1$ used in previous sections. The three parameters A, B, N change widely when calculated from bilinearizing the original dynamics around (x_0, u_0) and around (x_1, u_1). The parameter N reflects the non-linearity of the approximation, and its values suffer the biggest change when calculated around both equilibria (it even changes sign). For these reasons an intent of adapting only N was performed, and the result was already satisfactory. Nevertheless, another run was simulated by adapting A, B, N simultaneously, without obtaining any significant improvement in the state behaviour. The values of N adapted under both conditions are plotted in Figure 6.

4. INCREASING THE DIMENSION OF THE APPROXIMATION

If continuous parameter updating results are insufficient to meet the accuracy requirements over the trajectories of the bilinear system, then a better approximation to the dynamics may be

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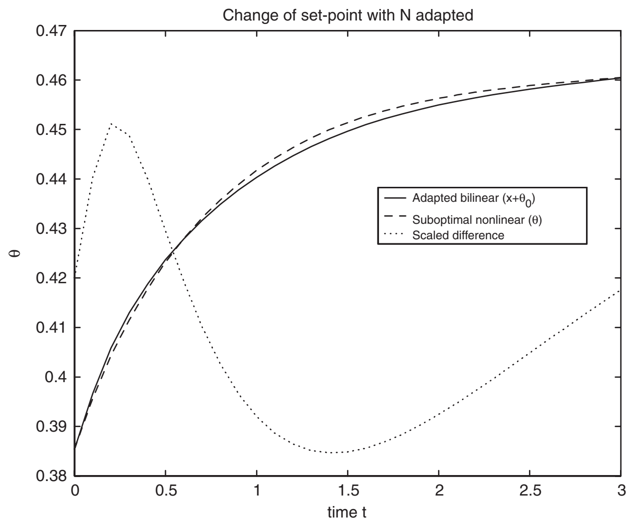

Figure 5. States for a change of set-point operation, resulting from the suboptimal control applied to the original plant, and to its bilinear approximation (with the parameter N adapted). The difference in states' trajectories is shifted to 0.42, and magnified 20 times.

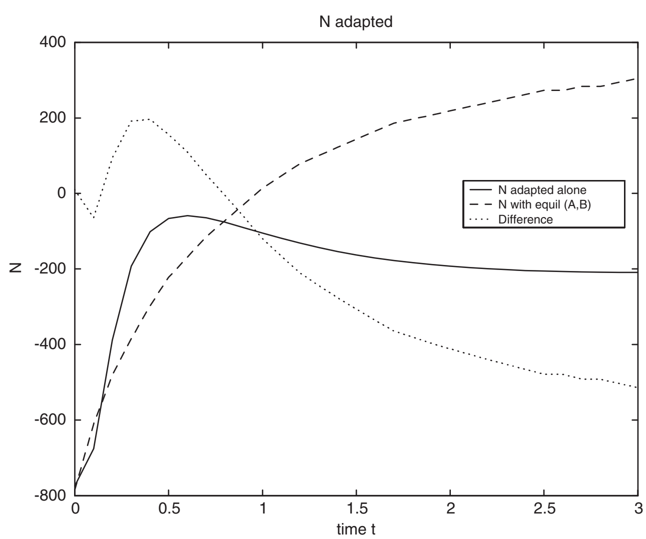

Figure 6. Parameter N adapted *via* sensitivity functions during the change of set-points $x_0 \to x_1$. In the solid line, parameters A, B are maintained at their nominal values (calculated around x_0), and only N is modified. The dashed line shows the values of N when the three parameters (A, B, N) are adapted simultaneously. The dotted line is a plot of the difference between the two updating strategies.

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needed. The natural step in this direction is to refine the approximation intended in Section 2 by adding higher order derivatives in the Taylor expansion (Equation (5)) of the original differential equation. By defining

$$x_1 \triangleq x$$
$$x_2 \triangleq x^2 \tag{33}$$

then several new terms from the series may be included in the dynamics

$$\dot{x}_1 = \dot{x} \approx A_1 x_1 + B_1 u + N_1 x_1 u + A_2 x_2 + N_2 x_2 u$$
$$\dot{x}_2 = 2x\dot{x} \approx 2A_1 x_2 + 2B_1 x_1 u + 2N_1 x_2 u \tag{34}$$

where the new coefficients are

$$A_1 \triangleq A, \quad B_1 \triangleq B, \quad N_1 \triangleq N, \quad A_2 \triangleq \frac{1}{2!}\frac{\partial^2 g}{\partial x^2}(\theta_0, \eta_0), \quad N_2 \triangleq \frac{1}{2!}\frac{\partial^3 g}{\partial x^2 \partial u}(\theta_0, \eta_0) \tag{35}$$

Then, after augmenting the state through $x \leftarrow [x_1 \;\; x_2]'$, a new bilinear approximation is generated and simulated (Figure 7). Now

$$\dot{x} \approx \check{A}x + \check{B}u + \check{N}xu, \quad x \in \mathbb{R}^2 \tag{36}$$

where

$$\check{A} \triangleq \begin{pmatrix} A_1 & A_2 \\ 0 & 2A_1 \end{pmatrix}, \quad \check{B} \triangleq \begin{pmatrix} B_1 \\ 0 \end{pmatrix}, \quad \check{N} \triangleq \begin{pmatrix} N_1 & N_2 \\ 2B_1 & 2N_1 \end{pmatrix} \tag{37}$$

In the present case, the nominal values of the new parameters result in

$$A_2 = -86.2384, \quad N_2 = 0 \tag{38}$$

This technique due to Carleman (see [23, 24]) is clearly extensible to other powers of the state, and allows to write increasingly higher order Taylor approximations of the original dynamics as higher order bilinear systems. For instance, the augmented matrices for a third-order bilinear approximation would read

$$\check{A} = \begin{pmatrix} A_1 & A_2 & A_3 \\ 0 & 2A_1 & 2A_2 \\ 0 & 0 & 3A_1 \end{pmatrix}, \quad \check{B} = \begin{pmatrix} B_1 \\ 0 \\ 0 \end{pmatrix}, \quad \check{N} = \begin{pmatrix} N_1 & N_2 & N_3 \\ 2B_1 & 2N_1 & 2N_2 \\ 0 & 3B_1 & 3N_1 \end{pmatrix} \tag{39}$$

where the new parameters are only those in the two upper right corners of the square matrices, namely

$$A_3 \triangleq \frac{1}{3!}\frac{\partial^3 g}{\partial x^3}(\theta_0, \eta_0), \quad N_3 \triangleq \frac{1}{3!}\frac{\partial^4 g}{\partial x^3 \partial u}(\theta_0, \eta_0) \tag{40}$$

No physical meaning of the state is lost (the original x always remains in x_1, and the other components represent just powers of the original state deviations). It should be noticed that only two new parameters would need to be estimated after increasing the dimension by one. Therefore, a bilinear approximation of dimension n will 'essentially' have $2n+1$ coefficients, while its matrices $\check{A}, \check{B}, \check{N}$ will have $2n^2 + n = n(2n+1)$ degrees of freedom, i.e. an order of magnitude less. This computationally convenient aspect seems not to have been taken into account in previous literature.

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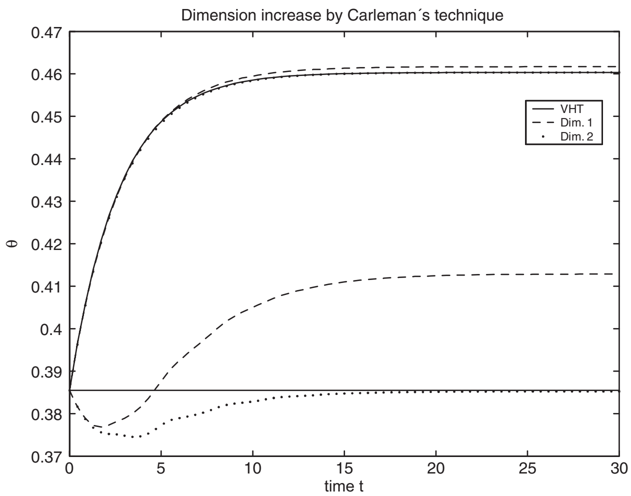

Figure 7. Step response for the original plant, the bilinear approximations of dimensions 1 and 2. The differences of the approximations with respect to the non-linear plant are correspondingly plotted in the lower part of the figure, magnified 20 times and shifted to θ_0.

Reduction of order may at some point be conducing, especially after the dimension has become so high as to endanger on-line computations. A trivial approach to order reduction may be just to return to the previous (smaller) dimension when strict and continuous accuracy has been detected during some agreed period of time. Here this aspect will not be discussed; for alternative approaches see [25].

5. NOISE IN I/O SIGNALS

Concerning disturbances, often called signal noise, the bilinear approximation constructed above is also useful (no general filter theory is available for non-linear systems). The stochastically perturbed bilinear system can be rewritten as

$$\dot{x} = [A + u(t)N]x + Bu(t) + \xi_1 \tag{41}$$

$$y = Cx + \xi_2 \tag{42}$$

where ξ_1 and ξ_2 are stochastic differentials of Brownian motions with coefficients σ_1 and σ_2, respectively, and y denotes the perturbed output signal, whose relation to physical variables is discussed below, together with a procedure for estimating and updating the observation parameter C.

The treatment that follows is referred to the optimal regulation problem (the extension to the servo problem for set-point changes being straightforward), so the underlying control (in feedback form) will be

$$u = -\frac{1}{R}(B + Nx)'\pi(x)x \tag{43}$$

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where $\pi(x)$ is the solution to the state-dependent Riccati equation (14). In what follows $u(t)$ will denote the time value of the suboptimal control (in practice $u(.)$ can be constructed on-line without knowing $\pi(x)$, since $x(.)$ and $\lambda(.)$ are available as the solutions to the initial-value Hamiltonian equations (11) and (12)).

In this context, the Kalman–Bucy filter for the approximated model can be implemented through (see [26, 27])

$$\dot{X} = [A + u(t)N]X + Bu(t) + G(t)[y - CX], \quad X(0) = \mathbb{E}(x_0) \tag{44}$$

where $G(t) \triangleq \Pi(t)C'\sigma_2^{-1}$ and $\Pi(.)$ is the solution to another Riccati-type ODE, integrable as an initial-value problem

$$\dot{\Pi} = [A + u(t)N]\Pi + \Pi[A + u(t)N]' - \Pi C' C_2^{-1} C\Pi + C_1, \quad \Pi(0) = \mathrm{Cov}(x_0) \tag{45}$$

and $C_1 \triangleq \sigma_1\sigma_1'$, $C_2 \triangleq \sigma_2^2$. Then, given the initial-value structure of the coupled ODEs' problem (44)–(45), this filter can be integrated on-line once the output variable $y(.)$ is clearly defined. The natural observable of the system is the current density J (see [13]) generated by the deposition of charges in the electrodes (e^- in left-hand-sides of Equations (1) and (2)), namely

$$J = F(v_V + v_H) \ (\triangleq h(\theta, \eta)) \tag{46}$$

Here an approximation to the deviation of J with respect to its (in general non-zero) initial value $J(0)$

$$y \triangleq J - h(\theta_0, \eta_0) \tag{47}$$

will be taken as the output for the model. Consistently with the Kalman–Bucy formulation, the following approximation is adopted:

$$y \approx \left[\frac{\partial h}{\partial \theta}(\theta_0, \eta_0) + \frac{\partial h}{\partial \eta}(\theta_0, \eta_0)\frac{\mathrm{d}k}{\mathrm{d}x}(0)\right] x = Cx \tag{48}$$

where the notation $k(x)$ refers to the feedback expression of the suboptimal control, namely

$$k(x) \triangleq -\frac{1}{R}(B + Nx)'\pi(x)x \tag{49}$$

and then the nominal (initial) value $\bar{C}$ of the observation parameter C can be calculated from

$$\bar{C} \triangleq \frac{\partial h}{\partial \theta}(\theta_0, \eta_0) - \frac{\partial h}{\partial \eta}(\theta_0, \eta_0)\frac{B'\pi(0)}{R} \tag{50}$$

It may be assumed at this point that a filtered state X is on-line available, and therefore a measure of the deviation from the bilinear model would be

$$\varepsilon \triangleq X - x \approx (\theta - \theta_0) - (\tilde{\theta} - \theta_0) \tag{51}$$

This error may be used to update the parameters of the bilinear approximation through Equations (30), (32) and (23) instead of the theoretical definition (31), since θ is not measured on-line. But, since theoretically the output J has a non-linear structure, then the parameter $\bar{C}$ would also need updating from the nominal value defined above, along similar lines to those of Section 3, for which another appropriate measure of the error between current densities is needed.

Assuming some updating for C is performed (see below), then an approximate filtered current density $\hat{J}$ may be calculated on-line from

$$\hat{J} \triangleq CX + h(\theta_0, \eta_0) \tag{52}$$

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This regularized variable should be a good approximation to the theoretical J defined in Equation (46), which is unavailable since it is corrupted by noise ξ_2. But taking into account the expected functionality of the current density, a deterministic non-linear observer can be constructed. An efficient scheme is discussed in [28]. The following intermediate definitions illustrate the structure of such a device:

$$\bar{E} \triangleq e^{f\eta}, \quad \hat{E} \triangleq e^{\alpha f\eta}, \quad \check{E} \triangleq e^{(\alpha-1)f\eta}$$

$$v_V^0 \triangleq \frac{\check{E}}{1-\theta_e}, \quad v_V^1 \triangleq v_V^0 + \frac{\check{E}}{\theta_e}, \quad v_H^0 \triangleq \frac{\hat{E}}{1-\theta_e}, \quad v_H^1 \triangleq v_H^0 + \frac{\check{E}}{\theta_e}$$

$$\check{A}(\eta) \triangleq -\frac{F}{\sigma}\left[v_V^e v_V^1 + v_H^e v_H^1 + \frac{4v_T^e}{(1-\theta_e)^2}\right], \quad \check{B} \triangleq -\frac{2Fv_T^e}{\sigma}\left[\frac{1}{\theta_e^2} - \frac{1}{(1-\theta_e)^2}\right]$$

$$\check{D}(\eta) \triangleq \frac{F}{\sigma}\left[v_V^e v_V^0 + v_H^e v_H^0 + \frac{2v_T^e}{(1-\theta_e)^2}\right], \quad \check{C}(\eta) \triangleq F[v_H^e v_H^1 - v_V^e v_V^1], \quad \check{E}(\eta) \triangleq F[v_V^e v_V^0 - v_H^e v_H^0] \tag{53}$$

The original dynamics given by Equations (4) and (46) take the following form:

$$\dot{\theta} = \check{A}(\eta)\theta + \check{B}\theta^2 + \check{D}(\eta) \tag{54}$$

$$J = \check{C}(\eta)\theta + \check{E}(\eta) \tag{55}$$

The non-linear observer adapted to this system generates $\Theta(.)$, a new approximation to the unmeasured physical variable $\theta(.)$, through the coupled dynamics

$$\dot{\Theta} = \tilde{A}(\eta)\Theta + \tilde{B}\Theta^2 + \tilde{D}(\eta) + \frac{\tilde{C}(\eta)}{\zeta}\{\hat{J} - \tilde{C}(\eta)\Theta - \tilde{E}(\eta)\}, \quad \Theta(0) = \theta_0 \tag{56}$$

$$\dot{\zeta} = -\mu\zeta - 2\tilde{A}(\eta)\zeta + 2[\tilde{C}(\eta)]^2, \quad \zeta(0) = 1 \tag{57}$$

where the ODE for the correction factor ζ is found from the requirement that

$$W(t, e_{obs}) \triangleq \frac{1}{2}\zeta(t)(e_{obs})^2 \tag{58}$$

be a time-dependent Lyapunov function, i.e. a smooth positive-definite function of the theoretical observation error

$$e_{obs}(.) \triangleq \Theta(.) - \theta(.) \tag{59}$$

with negative derivative along e_{obs}-trajectories (see [28]).

The updating, filtering and observing strategies were applied simultaneously to the same change of set-point treated before. The results are depicted in Figure 8, and since the initial value θ_0 for the observer is exactly known, the trajectory $\Theta(.)$ keeps too close to the filtered state as to be noticed in the figure.

Figure 9 illustrates only the performance of the observer after a simulated perturbation of 0.02 in the (unknown) state, a value of $\mu = 1$ (in this case, $\check{B} < 0$, so any positive μ will do) and

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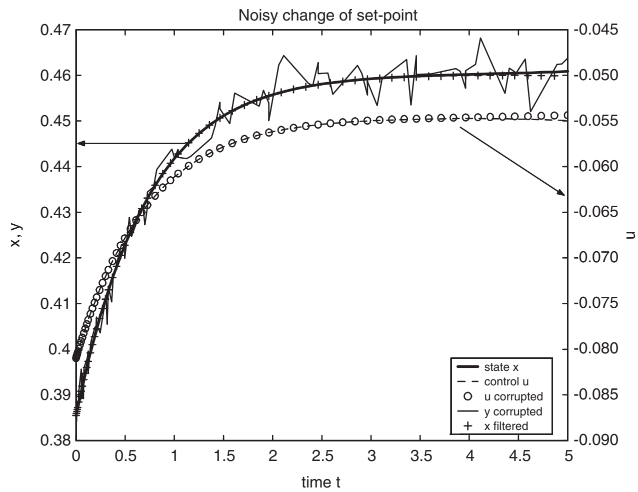

Figure 8. State and control, corrupted and filtered, for a noisy change of set-point. The variable Θ coming from the observer results indistinguishable from the filtered state, so it has not been plotted.

$\zeta_0 = 1$. The observer catches the state in a very small time, so the value of the output variable corresponding to the physical state θ will be nearly the same as the observation function calculated from Θ-values.

Then the appropriate error-measure for updating the parameter C would be

$$\varepsilon_1 \triangleq y_1 - Y \tag{60}$$

where y_1 captures the non-linear structure of the current density through

$$y_1 \triangleq h(\Theta, \eta) - h(\theta_0, \eta_0) \tag{61}$$

and Y denotes the best output computable from the Kalman–Bucy filter equations

$$Y \triangleq CX (= \hat{J} - h(\theta_0, \eta_0)) \tag{62}$$

Finally, after defining the objective to minimize through the updating of C, namely

$$\chi_1^2(t, p) \triangleq \int_0^t \mathrm{e}^{-a(t-\tau)} [y_1(\tau) - CX(\tau)]^2 \, \mathrm{d}\tau \tag{63}$$

the differential equations that should be integrated on line with this purpose (see Section 3) would be

$$\dot{C} = P_1 X \varepsilon_1 \tag{64}$$

$$\dot{P}_1 = aP_1 - P_1 X X' P_1 \tag{65}$$

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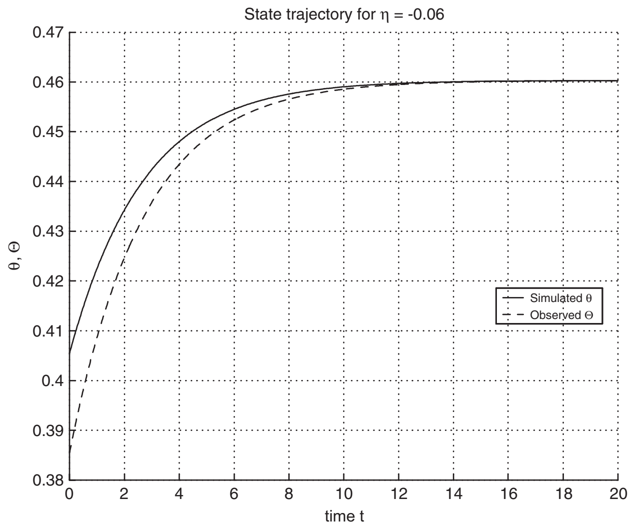

Figure 9. Performance of the non-linear observer.

where

$$P_1(t) \triangleq \left[\int_0^t e^{-a(t-\tau)} X(\tau) X'(\tau)\, d\tau \right]^{-1} \quad \forall t > 0 \tag{66}$$

As explained in Section 3, updating of C may be safely initiated after some small resetting time δ_1, eventually equal to δ, enough for a reliable finite value for $P_1(\delta)$ to be estimated from Equation (66), and then continue integration of the ODEs with

$$C(\delta_1) = \bar{C}$$

6. SIGNALS' FLOW FOR THE COMBINED CONTROL, FILTERING, AND UPDATING STRATEGIES

A flowchart of the adaptive-filtered suboptimal control scheme proposed in this paper is presented in Figure 10. The state variable θ is unavailable in practice, therefore the error ε used for updating purposes is calculated from the filtered state X instead, i.e. $\varepsilon = X_1 - x_1 \approx \theta - \tilde{\theta}$ (the first component of vector x, and consequently the first of X, are representative of the approximation $\tilde{\theta}$ to the original state θ, the other components representing powers of the state). By using X instead of Θ at this stage, the updating of the parameters A, B, N, involved in the approximate dynamics of the state, is kept virtually independent of the form of the observation function h.

However, the presumed analytical dependence of the current density J on the state and control plays its role at the observer stage, where h enters the calculations through $\check{C}$ and $\check{E}$. So the observed state Θ, calculated from J, is the appropriate value to correct the observation

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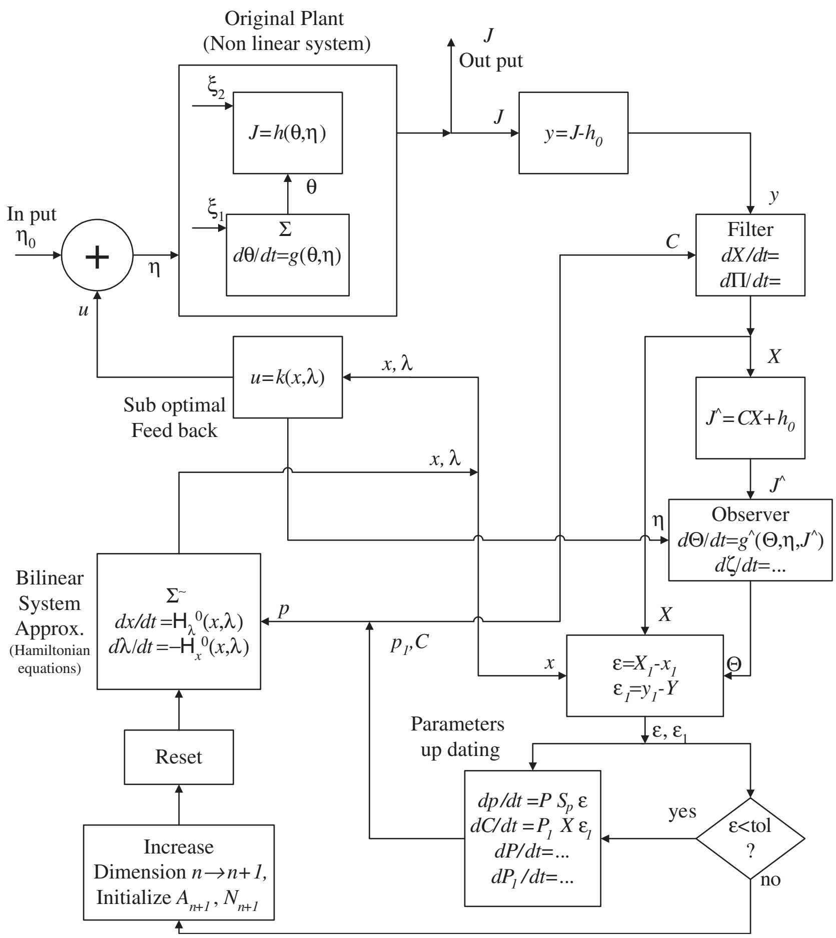

Figure 10. Flow of signals for the complete strategy.

parameter C of the filter, which is made through the error $\varepsilon_1 = h(\Theta, \eta) - \hat{J} \approx J - CX \approx J - Cx = J - \tilde{J}$. Here $\tilde{J}$ denotes the output corresponding to the observed bilinear approximation

$$\dot{x} = Ax + Bu + Nxu$$

$$y = Cx$$

7. CONCLUSIONS

In this paper the optimal control of HER reactions in the presence of uncertainty and noise has been treated from an engineering viewpoint. In doing so a rather complete methodology has been developed to treat the same problem for non-linear processes whose dynamics are accurately modelled by smooth differential equations.

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The strategy relies on bilinear approximations of the dynamics, whose dimensions are increased as needed by adding new terms of the Taylor expansion corresponding to the original non-linear plant, *via* Carleman's technique. The parameters of these approximations are continuously updated by using sensitivity matrices. The error between observables of the plant and its approximation is corrupted by noise, so a filter becomes necessary. It is shown that the Kalman–Bucy structure is suitable for the bilinear model, provided an observation matrix C is efficiently estimated. This forces to run a non-linear observer in parallel, since the original output (the current density J) theoretically depends on the state in a complicated non-linear fashion.

Each stage of the scheme has been individually illustrated through numerical simulations, in regulation and servo problems with quadratic optimality criteria. The complete control procedure requires the following ODEs to be integrated on-line (for a fixed dimension n of the bilinear approximation):

(i) the bilinear dynamics in Hamiltonian form for x, λ ($2n$ equations);
(ii) the updated parameters p (approximately $2n + 1$), sensitivity matrix S_p ($2n + 1$ equations) and covariance matrices P ($2n + 1$ equations);
(iii) the filter (n equations for the state X) and its corresponding matrix Riccati ODE for Π (n equations, by neglecting non-diagonal elements), and $2n$ equations for updating C (includes P_1);
(iv) the non-linear observer Θ and its corresponding correction factor μ; 2 equations.

This makes a total of $12n + 5$ coupled ODEs running on-line. In this paper the whole strategy has been put to work in dimension one, but the number of equations for reasonably low dimensions remain easily tractable with modern programmable equipment used in process control.

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