END-USE-ORIENTED DISCRETISATION OF TYPICAL CHEMICAL-PROCESS MODELS

DAVID I. WILSON

Modelling & Simulation Group Department of Electrical Engineering Karlstad University, SE - 651 88 Sweden email: david.wilson@kau.se

Abstract

Estimation and control applications in the chemical process industry require discretisation of continuous-time process models that are usually available from first principles. Simple explicit techniques for discretisation, have rarely been employed for the fear of introducing excessive error in the discrete model. In our view such techniques suffice for most chemical engineering applications given the prior inaccuracy in the continuous-time model and the choice of sampling frequency dictated by the measurement hardware, the plant dynamics and the needs of the estimation or control algorithms. In this work, the Euler and second order Runge-Kutta procedures are shown to provide sufficient accuracy for a wide range of chemical engineering applications. Use of these techniques is recommended for practically all explicit discretisation needs, at least as a first step to establish possible need for more complex alternatives.

Keywords: discretisation, chemical process models, sampling, stability

1. Introduction

The chemical process industry uses continuous-time dynamic process models in a variety of applications including design and control. This work is concerned with the discretisation procedure employed in view of the end-use of these types of models, and the subsequent solution of the ODEs. A critical issue is the choice of the discretising step.

The motivation for this look into discretisation procedures stemmed from our investigation into estimation and optimisation for batch reactors. Crucial for success for these types of applications is to perform the integration rapidly whilst maintaining appropriate accuracy. Batch chemical processes are characterised by significant nonlinear behaviour and continually changing operating conditions. Thus the nonlinearities need be preserved during integration, but the computational demands using library integration routines is prohibitive.

This paper argues that, compared to other fields, the chemical industry uses relatively poor models with high uncertainties and, consequently, relatively relaxed error tolerances. In this environment, the simple, fast, but somewhat inaccurate, explicit low-order Runge-Kutta schemes are shown to be appropriate. The infiltration of additional error is more than compensated by the gain in computational speed. In addition, the explicit nature of the scheme can be exploited further for applications in control, estimation and optimisation particularly in view of the capabilities of symbolic manipulators.

Section 2. outlines the need for explicit discretisation of process models used typically within the chemical industry for applications such as control, estimation and optimisation and reviews the current practice highlighting why this is inadvisable given the nature of these applications. Section 3. presents two simple explicit integration schemes to achieve these needs while avoiding the drawbacks of the current practice. The feasibility of these schemes is assessed in section 4. using several examples covering an array of published models. Conclusions are presented in section 5..

2. Need for discretisation

Dynamic process models encountered in the chemical engineering field are typically systems of nonlinear Ordinary Differential Equations (ODEs) usually comprising of firstprinciple relations and heuristics, of low order, and with no tractable analytical solution. This work restricts the consideration to typical process models suitable for use in estimation, on-line optimisation or control in an industrial environment. Therefore we deliberately exclude large dimensional systems, sparse systems, excessively stiff systems and differential algebraic systems. These problems require specialised solution techniques and software.

Most chemical engineering models were developed originally as continuous models, but there are at least three basic categories that require, or benefit from, a discrete model.

Discrete model required as input

Often an application requires a discrete model as an input to some other algorithm. One example is predictive control where the current input is related to some future desired state. If the model is explicit, one may be able to invert this to create a corresponding explicit control law. If this is not the case, as in the case of a library integration routine, then this complicates the inversion procedure.

Repetitive integration

The second main motivation for explicit discretisation is where the end-use requires repetitive integration so minimising computational speed is a primary objective. This can be further subdivided in two categories: plain iteration of the integration step such as in a simulation or in operator training, or a time critical real-time application, and where the explicit nature of the model can improve the performance of a higher level application such as on-line optimisation.

Contrary to popular belief, library routines are not always as computationally efficient as the simple explicit schemes given chemical engineering applications. While efficient integrating schemes may be able to take large steps, with only a few derivative function evaluations per step, consistent with a user-specified performance, this advantage is lost if the optimised step is larger than the externally imposed sample time. Furthermore multistep methods are not self-starting so incur a penalty at every start and finish of the externally imposed step. For these reasons, for the applications considered here, generally accepted library routines for general purpose numerical integration often perform poorer than expected.

Software availability

Finally, a simple one step explicit discretisation of the model is the easiest to implement and maintain on current industrial distributed control hardware. If library routines are called for, they are usually executed on a networked auxiliary workstation where often, the DCS-computer communication link further increases computation time, and decreases reliability. [1] notes that even the current OPC link incurs a 5 second penalty.

Given these needs for explicit discrete models, and the unique environment of the applications, it is worthwhile to critically analyse the current practice of approaching estimation, optimisation and control problems.

2.1 Current practice

For a justification of why the current and convenient practice of employing library integration routines (such as [2] or [3]) are often overused, it is necessary to look in more detail at the type of processes, and environment common in applications of control, estimation and optimisation in the chemical industry.

Firstly, relatively poor models with high levels of uncertainty are used in applications where moderately large errors are acceptable. These modelling errors are often understated or ill-characterised in much chemical engineering literature and thus there seems a large discrepancy between academic research simulation studies and actual industrial feasibility. There is a danger that over-enthusiastic researchers develop control algorithms with stringent model requirements that simulate perfectly, but fail in industrial tests. Despite the maturity of linear control invariably however the underlying dynamics are nonlinear leading to model/plant mismatch. For example [4, p219] report that the controller gains that gave the best *simulated* performance using a 5 state linear model were one order of magnitude too high to be implemented on the pilot equipment. Three decades ago, [5] concludes that applying methods developed elsewhere to chemical processes that differ so significantly mean that these solutions have little practical value. His recommendation then was to apply simple techniques to low order models and we would argue that this advice is still relevant today.

Nevertheless many practitioners avoid the crude Euler type integration techniques for fear of introducing excessive error, or excessive computation time due to the small stepsize requirement. It is worth noting that computational speed and storage is still at a premium in typical industrial processing plants overburdened with supervisory duties coupled with crude low level operating environments.

In summary, the current practice is divided either to use linear models, that possibly do not adequately reflect the nonlinear process, or overcompensate by using state of the art numerical library routines that are computationally inefficient, unwieldy, and not explicit.

3. Explicit discretisation

Assuming we desire to retain the underlying nonlinearities, we wish to discretise initial value ODES of the form

$$\frac{d\mathbf{x}}{dt} = \mathbf{f}(\mathbf{x}, \mathbf{u}, t), \qquad \mathbf{x}(t=0) = \mathbf{x}_0 \tag{1}$$

where in the control context, the input, **u**, is assumed bounded and constant over the period of integration (the sample time), and thus for the purposes of this analysis, the input may be neglected. Most numerical analysis texts for engineering type applications advise that a 4th order Runge-Kutta (RK) ODE integrator including various improvements is adequate for all but the most demanding problems. We argue that even this algorithm is an overkill for most chemical engineering applications and low order explicit RK ODE solvers are more suitable.

The Euler scheme approximates the derivative as a first order finite difference, advancing the state vector \mathbf{x}_i over one time step $h \stackrel{\text{def}}{=} t_{i+1} - t_i$ to \mathbf{x}_{i+1} as

$$\mathbf{x}_{i+1} = \mathbf{x}_i + h \, \mathbf{f}(\mathbf{x}_i, t_i) \tag{2}$$

This algorithm is first order accurate and requires one function evaluation per step. Most practitioners discourage its use because of stability problems at larger step sizes and relatively poor accuracy at a given step size. A second order variant, RK_2 ,

$$\mathbf{x}_{i+1} = \mathbf{x}_i + h \mathbf{f} \left(\mathbf{x}_i + \frac{h}{2} \mathbf{f} \left(\mathbf{x}_i, t_i \right), t_i + \frac{h}{2} \right) \quad (3)$$

has similar stability restrictions as the Euler scheme, but has improved accuracy. Both Eqns 2 and 3 are explicit, simple self starting one step integrators. For most chemical engineering processes, the accuracy is improved with decreasing step size.

With such trivial discretisation methods (as opposed to say methods proposed in [6]), we have the option, if we desire an eventual linear discrete model, to either linearise first, then perform an (exact) discretisation, or discretise first using either Eqns 2 or 3 and then linearise by a Taylor series. In general the final expressions are different, and it is worth noting that the former case, with its exact discretisation step, was not necessarily the most accurate.

3.1 Sample time and integration step size

The potentially most concerning aspect of using the first or second order scheme is not their accuracy, but their stability. The maximum allowable step size to still maintain stability when integrating $\dot{\mathbf{x}} = \mathbf{A}\mathbf{x}$ is a function of the eigenvalues (λ) of \mathbf{A} or the Jacobian for a nonlinear system. For the Euler scheme to be stable when applied to a single variable process, the step size, (h), must be chosen such that

$$|1+h\lambda| \le 1 \tag{4}$$

while for the second order scheme, the stability region is defined by

$$\left|1 + h\lambda + \frac{h^2\lambda^2}{2}\right| \le 1 \tag{5}$$

For multivariable processes, each eigenvalue is tested in turn, and the smallest resulting stepsize is the stability limit, since the eigenvalue with the largest magnitude is not necessarily the one that contributes to the limiting step size. Indeed it is possible that different eigenvalues are governing depending on the integrator scheme. Given a system with the 4 eigenvalues $-0.2\pm0.75i$, $-2\pm0.75i$, then the largest magnitude governs the maximum allowable step size for RK₂ scheme, but the other smaller eigenvalue dictates the maximum allowable step size when using the first order Euler scheme.

If stability governs one upper limit of the stepsize, then sample time selection provides another practical limit. For these trivial integration schemes to be attractive, this maximum allowable stepsize should be of the same order of magnitude (or larger) than the employed sample time. Depending on the application, various recommendations for the sample time selection are given ranging from 0.1–0.01 τ where τ is the open loop dominant time constant, (see e.g. [7] or [8, p37]). Naturally there are complications given nonlinear systems, batch processes with integrating states and so forth.

Aside from considerations of process dynamics, most industrial process-control hardware cannot readily sample faster than about 100 ms^{-1} , nor is there often any advantage to sample slower than about 1–5 minutes⁻¹ when using continuous sensors. Therefore sampling rates of the order of s⁻¹ irrespective of the process are common, [9, p339].

In practice, we chose a sample rate based on the process dynamics such as 1-10% of the dominant time constant. If the system has integrators, then we selected the smallest nonzero eigenvalue as the dominant time constant. If that proved unacceptable, then one must resort to choosing the sampling rate based on some process specific reason such as hardware cost, batch time, or engineering judgement.

Many chemical engineering processes are, or are very close to being overdamped. With this approximation, one can relate the appropriate sampling time to the maximum allowable step size. Since the sampling recommendation is based on the dominant time constant (smallest λ), and the stability is based on the largest λ , then the stiffness (ratio of eigenvalues) is an important consideration. Roughly to maintain stability of an explicit RK scheme with a step size the same as the sample rate (which is $0.1-0.01\tau$), then stiffness should be less than $20-200.^1$

In summary, this work makes the assumption that the explicit schemes are attractive only if the step size to achieve some reasonable tolerance (say 1% error) is in the same order of magnitude as the sampling rate. In other words, the stiffness of the problems of interest, should be less than around 200.

4. Example chemical processes

To support the claims made in sections 1. and 2., we have selected 15 well known and oft cited systems from the chemical engineering literature. For each of these systems, open loop simulation runs employing each of the two explicit algorithms from section 3. are compared to that employing a reliable library integration routine, namely algorithm DC03 from the Harwell library of codes, [10]. Where possible, runs in the original publication are closely followed. For originally closed-loop examples, a preset input was used to approximate the closed-loop run. The aim of these simulations is to verify that the simple explicit schemes are applicable across a variety of typical process models using appropriate integration steps.

An overview of the processes citing the original reference, the type of chemical process, the size (number of states) of the system is given in Table 1. Implementation details such as the sample time (possibly only units), integration algorithm, and hardware used in the original study are given if known. Space precludes results for all examples, but two representative processes are given starting in section 4.1 while further details are given in [11].

To aid the performance comparison, the states are normalised using a nominal range (x_{range}) and a nominal midpoint (x_{mid}) giving a dimensionless state that roughly lies between ± 0.5 . The error for a particular step size between

$$\frac{2}{\lambda_{\text{max}}|} = 0.1 \frac{1}{|\lambda_{\text{min}}|} \tag{6}$$

¹Setting the maximum allowable step size equal to the recommended sample rate gives

	dim.	h	$\frac{\lambda_{\max}}{\lambda_{\min}}$	$h_{ m max}$	h^{\star}	stepsize at 1% error			
Application & Author						RK_2		Euler	
						$\overline{\epsilon}$	$\epsilon_{\rm max}$	$\overline{\epsilon}$	$\epsilon_{\rm max}$
CSTR, Seinfeld (1970), [12]	2	s	12	6.6	0.6	> 6	5.5	1	0.5
CSTR (Sodium acetate), Litchfield et al (1979), [13]	3	30 s	6.8	42	1.4	20	10	11	2
CSTR, Morningred et al (1992), [14]	2	0.1min	2.4	0.42	0.005	0.25	0.1	0.05	0.005
CSTR fermentation, Henson & Seborg (1991), [15]	3	h	74	9.9	0.1	> 8	5	2	1
CSTR (methanol), Tátrai et al (1992), [16]	3	S	250	0.73	0.9	0.6	0.3	0.6	0.1
CSTR (pH), Henson & Seborg (1991), [17]	2	0.1s	1	169	9	44	9	9	5
Fed-batch (Bio), Rosen & Luus (1991), [18]	5	h	80	0.11	0.003	> 0.1	> 0.1	0.03	0.01
Batch (Bio), Caminal et al (1987), [19]	3	min	65	100	0.8	38	25	8	4
Batch (pulp), Venkateswarlu & Gangiah (1992), [20]	3	h	1	1.25	0.03	0.4	0.2	0.2	< 0.05
Batch PVC, Kiparissides & Shah (1983), [21]	7	S	$\approx 10^3$	32	0.2	26	6	14	< 5
Evaporator, Newell & Lee (1989), [22]	3	1min	1.7	21	0.2	> 2	> 2	0.8	< 0.5
Fed-batch crystalliser, Wilson (1990), [23]	6	0.005h	820	0.048	0.2	0.038	0.02	0.03	0.004
Furnace, Coggan & Noton (1970), [24]	3	0.1 h	7	0.2	0.007	0.09	0.05	0.05	0.01
Arc-furnace steel refining, Woodside et al (1970), [25]	2	h	50	0.5	0.12	> 0.2	> 0.2	0.08	0.03
Chaotic (autocatalytic), Lynch (1992), [26]	3	-	10^{4}		Inappropriate				

Table 1. A summary of the example chemical processes

the normalised RK results $\mathbf{x}_n^{\mathrm{RK}}$ and the Gear results $\mathbf{x}_n^{\mathrm{G}}$) is characterised in two ways: the maximum instantaneous absolute error $\boldsymbol{\epsilon}_{\mathrm{max}}$, and an average absolute error over the run,

$$\bar{\boldsymbol{\epsilon}} = \frac{1}{t_f} \int_0^{t_f} |\mathbf{x}_n^{\mathrm{G}} - \mathbf{x}_n^{\mathrm{RK}}| \, dt \tag{7}$$

From the 'exact' (Gear) profile, the eigenvalues of the locally linearised system are calculated to indicate the extent of nonlinearity and stiffness. The maximum and average state errors using the RK₂ (solid) and Euler (dashed) schemes are plotted against the integration step size. A reasonable error acceptance level (considering the uncertainty in the actual models used) is $\approx 1\%$. The proposed scheme is considered successful if the error is less than 1% using a step length that can serve as an appropriate sample time.

4.1 A batch bio-reactor

An example of enzymatic hydrolysis of cellulose with enzyme deactivation in a bio-reactor is given in [19]. The three state model is

$$\frac{dC_a}{dt} = -\frac{r_m C_a}{K_m (1 + C_b/K_i) + C_a} \tag{8}$$

$$\frac{dC_c}{dt} = \frac{r'_m C_b}{K'_m (1 + C_c / K'_i) + C_b}$$
(9)

$$\frac{dC_b}{dt} = -\frac{dC_a}{dt} + \frac{dC_c}{dt} \tag{10}$$

where the deactivation is explicitly dependent on time with a grace period and a deactivation period as

$$t < 400, \quad r'_m = r_m^{0'}$$
 (11)

$$t > 400, \quad r'_m = r_m^{0'} \exp\left(-1.5 \cdot 10^{-3} (t - 400)\right) \quad (12)$$

This system is stable, and the stiffness ranges from 2–11. The maximum stable step size 100–400 min is larger than the recommended 0.8–8min based on the dominant time constant. The RK_2 scheme will achieve the 1% error criteria with any recommended sampling rate, while the Euler's performance is borderline.

4.2 A batch pulp digester

Venkateswarlu and Gangiah, [20], describe a low order model for delignification in a Kraft pulping digester.

$$\frac{dC}{dt} = -q_1 \exp\left(b - \frac{a}{T}\right) CL \tag{13}$$

$$\frac{dL}{dt} = -q_2 \exp\left(b - \frac{a}{T}\right) CL \tag{14}$$

$$\frac{d[CO]}{dt} = q_3 \frac{dL}{dt} \tag{15}$$

The states are alkali, lignin and carbohydrate concentrations. The parameters q_1-q_3 were fitted to experimental data from a laboratory scale batch digester. The input for this system was an open loop temperature program specified as a function in time. This is system has two zero eigenvalues, which leaves only one nonzero eigenvalue, thus stiffness is not relevant for this example. The batch time is quite long (10 hours), and the time constant based sample time is 0.03–0.3 h. In practice, these are relatively large sample times, and one would expect to use much smaller, however pulp digesters are well known to be difficult units to control, primarily due to the lack of any good on-line transducers. Common pseudo on-line transducers such as kappa number meters have measurement cycles in the order of 20 minutes. The stability limit is larger at 2–7



Figure 1. An enzyme deactivation. Left: A nominal openloop trajectory at h = 0.8 (Gear —, $RK_2 \cdots$, Euler – –), and lower, the absolute values of the eigenvalues. Right: The average (upper) and maximum (lower) errors incurred at different step sizes ($RK_2 - - \circ - -$, Euler - - * - -)

hours. The RK₂ scheme can achieve the required accuracy with step sizes inside this recommended region, but the Euler scheme cannot achieve a maximum error of < 1% with h > 0.05.

4.3 General simulated results

Table 1 summarises of the results for all the considered processes. The approximate stiffness and maximum allowable integrator stepsize, $h_{\rm max}$ that still preserves stability are tabulated. The lower recommended sample time, h^* , is noted using the guidelines presented in section 3.1. The final four columns list the stepsize at which the 2 explicit integrator schemes, over the simulation presented, reach the standard 1% error. A large number here indicates that the explicit scheme can take relatively large steps, and still meet the required accuracy.

Most problems have ratios less than 200, so one may expect the explicit scheme to be sufficient. Actually for some processes with stiffness > 200, the explicit scheme is still sufficient, and these are discussed in more detail in [11].

For our implementation, the explicit schemes only used 5-10% of the computational load of the library routine, with the Euler scheme half that of the RK₂. The order of the average error with respect to stepsize for each of the processes studied is approximately 1 for the Euler and approximately 2 for the RK₂ scheme as expected from theory.



Figure 2. A batch pulp digester. (Legend as for Fig. 1.)

5. Conclusions

Many reported applications of control, optimisation or estimation in the chemical processing industries use cumbersome state of the art numerical techniques to integrate the dynamic system. For some applications, this is necessary, but for many applications particularly in on-line control or estimation of industrial processes, the dynamic model is small, moderately stiff with a relatively high degree of uncertainty. For these applications, we favour the simple explicit RK₂ scheme, that for reasonable time steps (roughly equal to the existing sampling rate), the accuracy and stability is quite sufficient. The advantages of the explicit scheme are the simple coding, fast execution and the possibility of deriving simple expressions such as analytical partial derivatives that would otherwise require solving the general nonlinear ODE. The disadvantage of the RK2 implementation is the sometimes tedious algebra required to obtain an explicit expression for the discrete model.

These simple explicit schemes are not a general panacea for all problems. They are unworkable for chaotic systems, and unsuitable for very stiff systems. One general solution approach for excessively stiff systems is to simplify the model eliminating the very fast (or perhaps very slow) dynamics. (Often this is done during the model construction stage.) With the recast problem now much less stiff, the explicit schemes can be used.

In summary, even for nonlinear, moderately illbehaved, stiff systems the second order scheme represents a good trade off between the accuracy at step sizes in the order of the sampling rate, and complexity of implementation. The simpler popular Euler implementation is only of borderline accuracy while the third and higher order RK explicit schemes are too complicated to extract analytical expressions for only marginal accuracy improvement.

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References

- Jarno Rinta-Valkama, Martti Välisuo, Tommi Karhela, Pasi Laakso, and Matti Paljakka. Simulation Aided Process Automation. In *Computer Aided Control System Design 2000*, University of Salford, UK, 11–13 September 2000.
- [2] C.W. Gear. Numerical Initial Value Problems in Ordinary Differential Equations. Prentice–Hall, Englewood Cliffs, NJ, 1971.
- [3] Hans Olsson. Runge-Kutta Solution of Initial Value Problems: Methods, Algorithms and Implementation. PhD thesis, Lund University, November 1998.
- [4] Robert B. Newell and D. Grant Fisher. Model development, reduction, and experimental evaluation for an evaporator. *Ind. Eng. Chem. Process Des. Develop.*, 11(2):213– 221, 1972.
- [5] A. S. Foss. Critique of chemical process control theory. *American Inst. Chemical Engineers J*, 19(2):209–214, 1973.
- [6] Sören Hohmann, Alexej Konrad, and Volker Krebs. Exact Sampled Data Representation of Continuous Time Nonlinear Systems by Finite polynomials with Exactly Determined Coefficients. In *American Control Conference*, pages 1628– 1633, Arlington, VA, 25–27 June 2001.
- [7] D.E. Seborg, T.F. Edgar, and D.A. Mellichamp. Process Dynamics and Control. Wiley, 1989.
- [8] K.J. Åström and B. Wittenmark. Computer controlled systems: Theory and design. Prentice–Hall, 1984.
- [9] Ivan Nagy. Introduction to Chemical Process Instrumentation. Elsevier, 1992.
- [10] United Kingdom Atomic Energy Authority. *Harwell Subroutine Library*, 9 edition, April 1989.
- [11] David I. Wilson and Mukul Agarwal. End-Use–Oriented Discretisation of Typical Chemical-Process Models. Technical Report SEG/R/284, Systems Engineering Group, ETH Zurich, CH-8092 Switzerland, 1993.
- [12] John H. Seinfeld. Optimal stochastic control of nonlinear systems. American Inst. Chemical Engineers J, 16:1016– 1022, 1970.
- [13] R.J. Litchfield, K.S. Campbell, and A. Locke. The application of several Kalman filters to the control of a real chemical reactor. *Trans. IChemE*, 57(2):113–120, April 1979.

- [14] J. Duane Morningred, Bradley E. Paden, Dale E. Seborg, and Duncan A. Mellichamp. An adaptive nonlinear predictive controller. *Chemical Engineering Science*, 47(4):755– 762, 1992.
- [15] Michael A. Henson and Dale E. Seborg. An internal model control strategy for nonlinear systems. *American Inst. Chemical Engineers J*, 37(7):1065–1081, 1991.
- [16] Ferenc Z. Tátrai, Ernö Varga, and Paul Benedek. Dynamic simulation of catalytic reactors using the UCKRON– I test problem as the kinetic model. *Ind. Eng. Chem. Res.*, 31(3):868–876, 1992.
- [17] Michael A. Henson and Dale E. Seborg. Time delay compensation for nonlinear processes. In *DYCORD*+, pages 151–156. Intl Federation of Automatic Control, 1992.
- [18] O. Rosen and R. Luus. Evaluation of gradients for piecewise constant optimal control. *Computers in Chemical Engineering*, 15(4):273–281, 1991.
- [19] G. Caminal, J. Lafuente, J. López-Santin, M. Poch, and C. Solà. Application of Extended Kalman filter to identification of enzymatic deactivation. *Biotechnology and Bioengineering*, 29:366–369, 1987.
- [20] Chimmiri Venkateswarlu and Kota Gangiah. Dynamic modelling and optimal state estimation using Extended Kalman filter for a kraft pulping digester. *Ind. Eng. Chem. Res.*, 31(3):848–855, 1992.
- [21] C. Kiparissides and S.L. Shah. Self-tuning and stable adaptive control of a batch polymerization reactor. *Automatica*, 19(3):225–235, 1983.
- [22] R.B. Newell and P.L. Lee. Applied Process Control A Case Study. Prentice–Hall, 1989.
- [23] David I. Wilson. Advanced Control of a Batch Raw Sugar Crystalliser. PhD thesis, The University of Queensland, St Lucia 4067, Australia, February 1990.
- [24] G. C. Coggan and A. R. M. Noton. Discrete-time sequential state and parameter estimation in chemical engineering. *Trans. Instn. Chem. Engrs.*, 48:T255–T264, 1970.
- [25] Charles M. Woodside, B. Pagurek, John Pauksens, and Arun N. Ogale. Singular arcs occurring in optimal electric steel refining. *IEEE transactions on automatic control*, AC–15(5):549–556, 1970.
- [26] David T. Lynch. Chaotic behaviour of reaction systems: Parallel cubic autocatalators. *Chemical Engineering Science*, 47(2), 1992.