Model reduction and control of multi-scale reaction–convection processes

Marie Nathalie Contou-Carrere[1], Prodromos Daoutidis*

Department of Chemical Engineering and Materials Science, University of Minnesota, Minneapolis, MN 55455, USA

A B S T R A C T

This paper focuses on non-isothermal plug flow reactors with fast and slow reactions. These processes are modeled by first order hyperbolic partial differential equations with large and small parameters, which results in stiffness and multi-scale behavior. Through a combination of the method of characteristics and singular perturbation techniques, a model reduction method is developed for obtaining a non-stiff representation of the dominant dynamics of these systems. The application of the method and the advantages of using the reduced models for model-based control are illustrated through a case study.

© 2008 Elsevier Ltd. All rights reserved.

1. Introduction

Chemical processes typically involve physical and chemical phenomena occurring over widespread time and length scales. Such processes are usually modeled by stiff systems of differential equations. The numerical solution of such systems is computationally intensive, especially in the case of PDEs (see e.g., Miranker, 1981; Verwer, 1996); furthermore, the direct use of such stiff systems as the basis for controller design is problematic due to the strong sensitivity of the resulting controllers to measurement and modeling errors (Kumar and Daoutidis, 1999). Therefore, there is a need for methods to derive non-stiff models that capture the essential dynamical features of such systems and can be used for efficient numerical simulation and control.

This problem of model reduction (or coarse graining at a broader level) has attracted a lot of attention in the literature recently (see e.g., the recent volume of Gorban et al., 2006, for a representative and broad collection of state of the art results). One direction of research is motivated by the dissipative nature of certain classes of distributed systems (e.g., reaction–diffusion or Navier–Stokes equations) which allows seeking low-order ODE systems that can adequately approximate the dominant dynamics of the PDE system, following a variety of analytical or numerical modal decomposition approaches (see e.g., Christofides, 2000; Christofides and Daoutidis, 1997 and the references therein). However, these techniques do not account for stiffness originating from the source term

in the underlying PDEs. Another direction concerns the efficient simulation and systems analysis of macroscopic phenomena based on model descriptions at a fine microscopic scale (see e.g., Kevrekidis et al., 2004, and the references therein). There also exists a substantial research body which addresses model stiffness due to chemical reactions whose rates span widely different orders of magnitude (see e.g., Maas and Pope, 1992; Lam and Goussis, 1994; Vora and Daoutidis, 2001; Gorban and Karlin, 2003; Goussis and Valorani, 2006 for representative approaches). Among them, the computational singular perturbation (CSP) method, originally developed for stiff ODEs, has been also extended to reaction-diffusion systems (Hadjinicolaou and Goussis, 1999; Goussis et al., 2005), aiming to address stiffness due to both the diffusion and the reaction terms.

In this work, we focus on transport-reaction systems where convective mechanisms dominate over diffusive ones. We focus in particular on non-isothermal plug flow reactors (PFRs) (a very common class of chemical reactors) with fast and slow reactions. These processes are modeled by first order hyperbolic partial differential equations (PDEs) with large and small parameters, which results in stiffness and multi-scale behavior. Unlike reaction–convection–diffusion processes, all the eigenmodes of the spatial differential operator of these systems contain the same amount of energy, which precludes the application of modal decomposition techniques (Christofides and Daoutidis, 1997). Moreover, contrary to the systems studied in Christofides and Daoutidis (1998), there is no clear distinction between fast and slow variables. In addition, the multi-scale behavior occurs both in time and space (i.e., all variables may present temporal and spatial boundary layers) owing to the distributed nature of the systems.

For the processes under consideration, first, we outline a dimensionless formulation of the standard material and energy balance

---

* Corresponding author. Tel.: +1 612 625 8818.
  E-mail address: daoutidi@cems.umn.edu (P. Daoutidis).
[1] M.N. Contou-Carrere currently with INEOS.

model. The resulting PDE models are then recast in the geometric context of the method of characteristics to provide insights on their multi-scale dynamical behavior, and specifically on the occurrence of temporal and spatial boundary layers. Then, by using singular perturbation arguments, we present a systematic framework to obtain a non-stiff representation of the slow (dominant) dynamics. Various cases are considered to account for fast heat transfer or fast convection phenomena too. The obtained reduced models can be used for controller design, by application of control methods available for hyperbolic PDEs (Sira-Ramirez, 1989; Hanczyc and Palazoglu, 1995; Christofides and Daoutidis, 1996; Gundepudi and Friedly, 1998; Orlov and Dochain, 2002; Karafyllis and Daoutidis, 2002; Shang et al., 2004; Smets et al., 2004). As an illustration, we apply the analysis framework to a representative chemical reaction system, for which we derive a non-stiff model of the slow dynamics using the proposed method, formulate a control problem and illustrate the advantages of using the non-stiff reduced model as the basis for controller design.

2. The stiff PDE model

Consider a PFR where the following R reactions involving N species take place

$$\sum_{j=1}^{N} v_{ij} A_j = 0 \quad i = 1, \ldots, R \tag{1}$$

where A_j denotes the species j and v_{ij} denotes the stoichiometric coefficient of the species j in the reaction i. The material and energy balances describing the evolution of the species compositions and temperature take the form

$$\frac{\partial C_j}{\partial t'} = -\widetilde{V} \frac{\partial C_j}{\partial z'} + \sum_{i=1}^{R} v_{ij} R_i \quad j = 1, \ldots, N$$

$$\frac{\partial T}{\partial t'} = -\widetilde{V} \frac{\partial T}{\partial z'} - \frac{1}{\rho c_p} \sum_{i=1}^{R} \Delta H_i R_i + \frac{UA}{\rho c_p V_R}(T^h - T) \tag{2}$$

with $C_j(t'=0, z') = f_j(z')$ as initial conditions and $C_j(t', z'=0) = g_j(t')$ as feed data for $j = 1, \ldots, N$, where t' denotes time, z' denotes the position with respect to the entrance of the reactor, C_j denotes the molar concentration of species j, T denotes the temperature, $\widetilde{V}$ is the axial velocity of the reacting mixture, ρ the density of the reacting mixture, c_p the mass heat capacity of the mixture, U the heat transfer coefficient, A the area of heat transfer, V_R the volume of the reactor, T^h the temperature of the heat-transfer medium, ΔH_i is the heat of reaction (with the usual convention of $\Delta H_i < 0$ for an exothermic reaction), and R_i is the reaction rate of the reaction i (in moles per unit of time per unit volume). For simplicity, it is assumed that the temperature T^h is constant and uniform.

In order to analyze such systems in the presence of fast and slow reactions, we derive a dimensionless form of Eq. (2), in appropriate normalized variables. We define $\widehat{\underline{C}}$ to be the vector of dimensionless concentrations, $\widehat{\underline{C}} = [C_1/C_{10} \cdots C_N/C_{N0}]^T$ where, for simplicity, the reference concentrations are all taken to be equal to unity ($C_{j0} = C_0 = 1$ for $j = 1, \ldots, N$), $\widehat{T}$ to be the dimensionless temperature $\widehat{T} = T/T_0$ (where T_0 is the reference temperature), $\widehat{T}^h$ to be the dimensionless temperature of the cooling medium $\widehat{T}^h = T^h/T_0$, x to be the vector of dimensionless concentrations and temperature $x = [\widehat{\underline{C}}^T \widehat{T}]^T$, z to be the dimensionless axial position $z = z'/L$ (where L denotes the length of the reactor), and t to be the dimensionless time scaled by the characteristic time of the reactor $t = (\widetilde{V}/L)t'$. Since, typically, the reaction rate R_i is expressed as $R_i(x) = k_i(T)r_i(C)$, where $k_i(T)$ is the reaction rate constant, we introduce the dimensionless Damköhler number, defined as the ratio of the characteristic residence time to

the characteristic reaction time

$$Da_i(T, C_0) = \frac{L}{\widetilde{V}} \frac{k_i(T) r_i(C_0)}{C_0}$$

We also introduce the dimensionless Stanton number which captures the characteristic time of heat transfer compared to the convection one

$$St = \frac{L}{\widetilde{V}} \frac{UA}{\rho c_p V_R}$$

In dimensionless variables, the model in Eq. (2) can be written in matrix form as

$$\frac{\partial}{\partial t} \begin{bmatrix} \widehat{C}_1 \\ \vdots \\ \widehat{C}_N \\ \widehat{T} \end{bmatrix} = -\frac{\partial}{\partial z} \begin{bmatrix} \widehat{C}_1 \\ \vdots \\ \widehat{C}_N \\ \widehat{T} \end{bmatrix} + \frac{L}{\widetilde{V}} \frac{UA}{\rho c_p V_R} \begin{bmatrix} 0 \\ \vdots \\ 0 \\ \widehat{T}^h - \widehat{T} \end{bmatrix}$$

$$+ \begin{bmatrix} v_{11} & \cdots & v_{R1} \\ \vdots & & \vdots \\ v_{1N} & \cdots & v_{RN} \\ -\dfrac{\Delta H_1 C_0}{\rho c_p T_0} & \cdots & -\dfrac{\Delta H_R C_0}{\rho c_p T_0} \end{bmatrix}$$

$$\times \begin{bmatrix} Da_1(T, C_0)\bar{r}_1(C) & & \\ & \ddots & \\ & & Da_R(T, C_0)\bar{r}_R(C) \end{bmatrix}$$

where

$$\bar{r}_i(C) = \frac{r_i(C)}{r_i(C_0)} \quad i = 1, \ldots, R$$

It is assumed that, in the temperature range of interest, p fast reactions have been identified; without loss of generality, we assume that the last p reactions are fast. More specifically, the Damköhler number of the $(R-p+1)$th reaction evaluated at T_0 and C_0, denoted Da_{R-p+1}^0, is assumed to be a large parameter (much greater than one) such that the Damköhler numbers $Da_i(T, C_0)$, for $1 \leqslant i \leqslant (R-p)$, are of much smaller order of magnitude than Da_{R-p+1}^0, and of the same order of magnitude as $Da_1(T_0, C_0)$, denoted Da_1^0, and the Damköhler number of the remaining p reactions are of the same order of magnitude as Da_{R-p+1}^0, i.e.,

$$\frac{Da_{R-p+1}^0}{Da_1^0} \gg 1$$

$$\frac{Da_i(T, C_0)}{Da_{R-p+1}^0} \simeq O(1) \quad R-p+1 \leqslant i \leqslant R$$

$$\frac{Da_i(T, C_0)}{Da_1^0} \simeq O(1) \quad 1 \leqslant i \leqslant R-p \tag{3}$$

Following this classification of slow and fast reactions, in order to isolate the source of stiffness to single parameters, we extract the representative dimensionless numbers Da_1^0, Da_{R-p+1}^0, and St so that the system takes the form

$$\frac{\partial x}{\partial t} = -\frac{\partial x}{\partial z} + \frac{1}{\varepsilon_h} \underline{g}(x) + \frac{1}{\varepsilon_s} \mathcal{V}_s(x) \underline{r}_s(x) + \frac{1}{\varepsilon_f} \mathcal{V}_f(x) \underline{r}_f(x) \tag{4}$$

where the dimensionless parameters ε_s, ε_f, ε_h characterize, respectively, the slow reactions, the fast reactions, and the heat transfer