POD-based modelling of the dynamics of a tubular reactor with recycle

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1 Introduction

Many chemically reactive systems are characterized by parametric sensitivity that results in the occurrence a wide variety of static and dynamic phenomena (Aris et al., 1991). Depending on the assumed operating conditions or on the values assumed by different parameters of the model which describes the physical and chemical properties of the system, static equilibrium as well as more complex time–asymptotic regimes such as periodic, quasi–periodic or chaotic oscillations can be observed. The dynamical analysis of distributed reactive systems described by PDE evolutionary equations often becomes very difficult because the order of the dynamical model required to properly describe the system is very high. This, coupled with obvious limitations of the software and hardware, leads to the need of expressing the original model as a set of ordinary differential equations of the lowest possible order. Among classical numerical approaches, finite difference methods are simple but require a relatively large number of ODE as compared for instance to orthogonal collocation. Other spectral methods have been used for reactive systems such as Galerkin projection. Projection methods provide an interesting framework in that by proper choice of the functional basis one can reduce the number of ODEs necessary to accurately describe the dynamics of the original PDE model. The Proper Orthogonal Decomposition (POD) approach delivers an optimal set of empirical basis functions from an ensemble of observations obtained either experimentally or from numerical simulation, which characterize the spatio–temporal complexity of the system. Obtained orthogonal functions can be afterwards used in a Galerkin projection of the original system and as a result a low–dimensional model can be developed (Lumley, 1967). Various strategies aiming at the determination of an optimal set of POD basis functions that are able to capture correctly the dynamics of the system can be found in literature. In fact, the reliability of the basis depends on the ensemble of observation, that is on the generation of this ensemble, namely, time of sampling, location and number or samples as well as varying system conditions such as variation of parameters or initial conditions occurred during sampling (Alonso et al, 2004). Moreover, a POD model is not generally valid far from the parameter range in which the snapshots were taken. On the other hand, Graham and Kevrekidis (1996) were able to determine a POD basis containing global information about the model by integrating from 1000 different initial conditions for one given value of the parameter, for which chaos was known to exist; the resulting dynamical model was reported to approximate well the solutions manifold. Zhang et al (2004) also obtained a POD dynamical model applicable to a wide range of conditions, by collecting data sets from simulations of the original system conducted for different initial conditions and for up to six different values of the bifurcation parameter. It should be noted that the number of basis functions chosen for the approximation is critical: Graham and Kevrekidis (1996) showed that an insufficient number of basis functions – in their case Chebyshev polynomials – can cause that significant phenomena such as period doubling are not detected.

In this work, we apply POD/Galerkin to a model of pseudohomogeneous tubular reactor with mass recycle in oscillatory regime. We analyze the performance of the method by comparing solutions from the reduced model with a “reference” numerical solution.

2 Mathematical model and numerical method

As an example we consider a model of pseudohomogeneous tubular reactor with mass recycle (Berezowski et al, 2000). The mathematical model is given by the following system of mass and heat balance equations:

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$$\frac{\partial \alpha}{\partial \tau}+\frac{\partial \alpha}{\partial \xi}=\frac{1}{Pe_M}\frac{\partial^2 \alpha}{\partial \xi^2}+(1-f)Da(1-\alpha)^n \exp\left(\gamma\frac{\beta\theta}{1+\beta\theta}\right) \tag{1}$$

$$Le\frac{\partial \theta}{\partial \tau}+\frac{\partial \theta}{\partial \xi}=\frac{1}{Pe_H}\frac{\partial^2 \theta}{\partial \xi^2}+(1-f)Da(1-\alpha)^n \exp\left(\gamma\frac{\beta\theta}{1+\beta\theta}\right)+(1-f)\delta(\theta_H-\theta) \tag{2}$$

with Danckwerts boundary conditions resulting from recycle, where α is the conversion degree and θ is the dimensionless temperature. In the present work the PDE model is first reduced to a system of ODE by approximating it with a cascade of 50 CSTR which is then employed to build reference solutions by time integration, to conduct bifurcation analysis, and finally to collect data needed for the generation of POD basis functions. In the POD scheme, the objective is to determine a set of orthogonal basis functions which minimize, on average, the least square error between the truncated representation of the model and the true solution. By collecting time series obtained by simulation, the sampled data set is a vector–valued function given as a matrix:

$$U=\begin{bmatrix} u_1(x_1) & u_2(x_1) & \cdots & u_M(x_1) \\ u_1(x_2) & u_2(x_2) & \cdots & u_M(x_2) \\ \vdots & \vdots & \ddots & \vdots \\ u_1(x_N) & u_2(x_N) & \cdots & u_M(x_N) \end{bmatrix} \tag{3}$$

where N is the number of positions in the spatial domain and M is the number of samples taken in time. A POD basis $\Phi=\{\varphi_1,\varphi_2,\ldots,\varphi_N\}$ is obtained by solving the eigenvalue problem $C\Phi=\lambda\Phi$ where $C(x,x')=\langle U_t(x),U_t(x')\rangle$ is the averaged autocorrelation matrix and angular brackets denote time–averaging operation. Using the POD modes, the solution $u_t(x)$ can be expressed as

$$\tilde{u}_t(x)=\sum_{k=1}^{K} a_k(t)\varphi_k(x) \tag{4}$$

where $K<N$ is the number of modes used for truncation, whereas $a_k(t)$ are modal coefficients that can be calculated by Galerkin projection of the original PDE on the POD modes. The ordering of the eigenvalues from the largest to the smallest induces an ordering in the corresponding eigenfunctions, from the most to the least important. Hence, in order to determine the truncation degree of the POD reduced model, we define the *cumulative correlation energy* captured by the *K* successive modes which is given by:

$$E_K=\sum_{k=1}^{K}\lambda_i \Big/ \sum_{k=1}^{N}\lambda_i \,. \tag{5}$$

Thus if the minimum required cumulative correlation energy is set, then the number of necessary modes is determined according to Eq. 5.

3 Results

Our goal is to evaluate the ability of POD/Galerkin to reproduce the dynamics of the system under study. To this aim, solutions obtained by POD/Galerkin are compared with those obtained with the CSTR cascade model.

Table 1. Eigenvalues, % cumulative correlation energy, error from "reference" solution and Hausdorff distance between limit sets of CSTR and POD solutions. Values computed for conversion degree.

Modes	1	3	5	10	13	19	23	36	49
Eigenvalue	23.26	0.30	6.79e-2	6.31e-3	2.76e-3	4.81e-4	1.68e-4	7.70e-6	8.17e-15
% Energy	95.39	98.93	99.57	99.92	99.97	99.99	100.00	100.00	100.00
Error	2.56e-2	2.48e-2	2.58e-2	3.01e-2	2.91e-2	6.62e-3	4.93e-5	7.41e-6	5.77e-6
$d_H(A,B)$	8.76e-1	4.34e-1	3.83e-1	1.15e-1	7.25e-2	1.91e-2	6.92e-3	1.92e-3	1.90e-3

First we must decide a criterion to decide how many modes are necessary for a satisfactory description. We evaluate the cumulative correlation energy as a function of the number of modes (Eq. 5). Then, the relative error between the POD solution and the CSTR "reference" solution defined as the mean square error of truncation

$\left\langle \left\| u-\tilde{u} \right\|^2 \right\rangle$, where $\|\cdot\|$ denotes Euclidean norm. Finally, we introduce the Hausdorff distance between sets defined as follows (Stuart and Humphries, 1998) $d_H(A,B) = \max\{dist(A,B), dist(B,A)\}$, where $dist(B,A) = \sup_{u\in B} dist(u,A)$ is called the Hausdorff semi–distance, and $dist(u,A) = \inf_{v\in A} \|u-v\|$ is the distance of one point from a set. Hence, we calculate the distance between two limit sets, namely the limit set obtained from CSTR simulation and its approximation by POD. The cumulative correlation energy proved insufficient to provide a criterion for selection of number of modes. The relative error provides a more direct evaluation of differences between the CSTR cascade solution and the reduced solution; however, even small frequency drifts can yield a large relative error for solutions which are essentially the same in the phase space, as reported in the example of Figure 1, which shows initial times (left) and steady states (right).

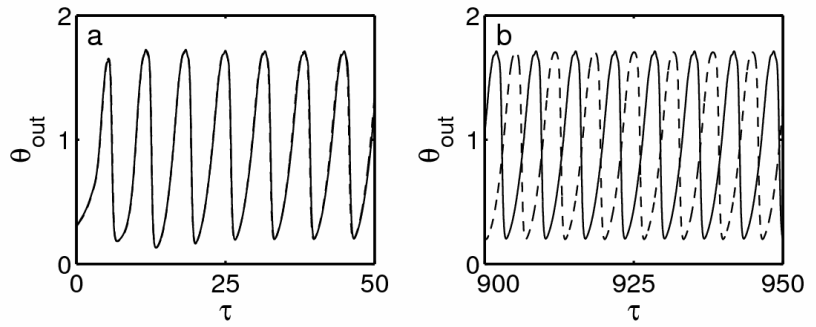

Figure 1. Time series, POD/Galerkin w 23 basis functions (solid line) and CSTR cascade (dashed line).

The same solution reported in Fig. 1 as a time series (POD with 23 basis functions) is seen in Fig. 2b in the phase space. The agreement with CSTR cascade (dashed line) is good, and almost no amplitude error is observed. On the contrary, Figure 2a reports the POD solution phase portrait for only 13 basis functions, where it is seen that there are unacceptable quantitative and qualitative discrepancies between the two solutions.

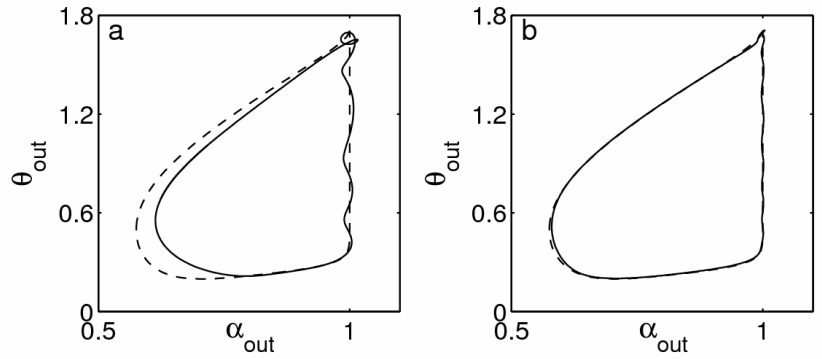

Figure 2. Phase portraits of limit sets (a) 13 basis functions (b) 23 basis functions.

Performance of POD reduced model was also evaluated with respect to qualitative dynamical features. Solution diagrams were obtained by parameter continuation of the CSTR approximation. Parameters of the model were kept constant as follows: $Pe_M = Pe_H = 100$, $f = 0.2$, $\beta = 0.75$, $Le = 5$, $\gamma = 15$, $\delta = 0$, whereas the value of Damköhler number Da was varied as the bifurcation parameter. As it is seen, for the assumed above set of parameters, multiple solutions exist i.e. one stable fixed point and stable and unstable oscillations. In Figure 3, the solution diagram computed for the system by brute force with POD/Galerkin using 19 modes is compared to the diagram obtained by automatic parameter continuation on the CSTR cascade model. POD basis functions were obtained by collecting snapshots from stable oscillations only in the range of Da in between the values 0.03 to 0.08. It is seen that the results match in almost the whole range of stable limit cycles. The first bifurcation is also captured, whereas the supercritical Hopf bifurcation is not reproduced.

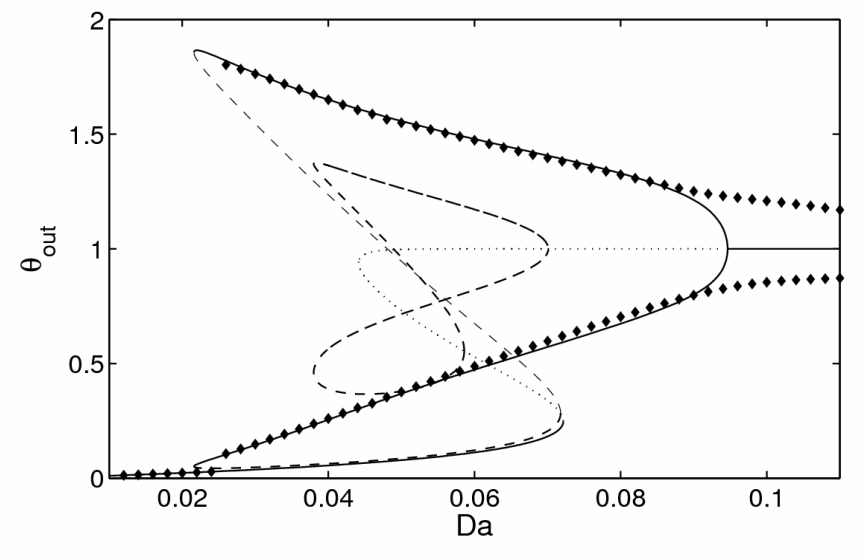

Figure 3. Solution diagram for conversion degree vs. Damköhler. Stable stationary solutions and stable oscillations (—); unstable oscillations (– –); unstable stationary solutions (…). Diamonds: POD/Galerkin.

4 Conclusions

In the attempt of building an accurate reduced dynamical of model of a pseudohomogeneous tubular reactor with mass recycle, a POD/Galerkin approach was developed and applied to simulations in the oscillatory regimes. Three different ways of comparing the solutions were employed to highlight features of the reduced model. It was found that the cumulative correlation energy is not a reliable criterion for such system. Since frequency drift can appear in the solution, also a conventional norm computed for the difference of time series is not a valid indication. The Hausdorff distance between limit sets is found to be the best means to assess accuracy of the reduced model although it does not carry information on time series.

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