# OPTIMAL MODEL REDUCTION OF A PSEUDO-HOMOGENEOUS PLUG-FLOW REACTOR

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**Abstract**
In this preliminary work, the application of a POD/Galerkin empirical spectral method is demonstrated on a pseudo-homogeneous tubular reactor, providing a substantial reduction of the model dimension. The optimal reduction is sought through sampling of chaotic orbits, which contain the maximum amount of information on the system dynamics.

## Introduction

Chemical-looping combustion integrates power production and $CO_2$ capture. Among the available technologies, packed bed reactors have recently been proposed [1]. These units, by holding a bed of oxygen carrier, may operate both for the oxidation and the reduction phase, with the advantage – with respect to fluidized beds – of avoiding the need for separation between carrier particles and gas stream. On the other hand, discontinuous operation is necessary to alternate solid oxidation and reduction. One possibility is the dynamical operation of the packed bed unit. The complex dynamics that may emerge requires accurate and fast modeling prior to design and implementation. Periodically forced systems are amenable to bifurcation analysis by means of specifically developed techniques [2] but models may become unmanageable in terms of state space dimension. In this view, empirical spectral methods based on POD [3] are expected to provide a substantial size reduction.

## Mathematical model

The present study concerns the model of a pseudo-homogenous autothermal reactor, with axial dispersion of mass and heat and external cooling (Figure 1).

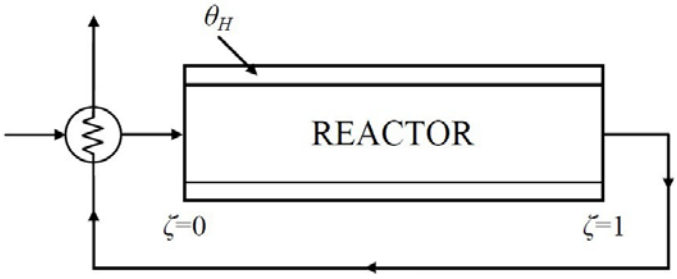


**Figure 1.** Pseudo homogeneous tubular reactor with recycle.

The pseudo-homogeneous model is suitable to describe fixed bed reactors for which intra-particle resistances to heat and mass transfer are small [4]. External heat recovery is the simplest way to implement a feedback mechanism which impacts the dynamics. The present model is taken from Ref. [5], where the authors analyze the dynamics of the system and demonstrate the occurrence of complex oscillatory regimes – including periodic, multiperiodic and chaotic oscillations, under the influence of parameters such as the coolant temperature, the Lewis number, and the Péclet numbers characterizing mass and heat dispersion. The partial differential equations (PDEs) expressing dimensionless mass and energy balances for the system are:

$$\begin{aligned} &\frac{\partial \alpha}{\partial \tau}+\frac{\partial \alpha}{\partial \zeta}=\frac{1}{\mathrm{Pe}_M}\frac{\partial^2 \alpha}{\partial \zeta^2}+\mathrm{Da}\left(1-\alpha\right)^n \exp\left(\gamma\frac{\beta\theta}{1+\beta\theta}\right) \\ &\mathrm{Le}\frac{\partial \theta}{\partial \tau}+\frac{\partial \theta}{\partial \zeta}=\frac{1}{\mathrm{Pe}_T}\frac{\partial^2 \theta}{\partial \zeta^2}+\mathrm{Da}\left(1-\alpha\right)^n \exp\left(\gamma\frac{\beta\theta}{1+\beta\theta}\right)+\delta\left(\theta_H-\theta\right) \end{aligned} \tag{1}$$

where $\alpha$ is a conversion degree and $\theta$ is a dimensionless temperature. The corresponding initial and boundary conditions are given, respectively, by:

$$\begin{aligned} &\alpha(\zeta,0)=\alpha_0(\zeta); \qquad \theta(\zeta,0)=\theta_0(\zeta) \\ &\alpha(0,\tau)=\frac{1}{\mathrm{Pe}_M}\left.\frac{\partial \alpha}{\partial \zeta}\right|_{\zeta=0}; \qquad \left.\frac{\partial \alpha}{\partial \zeta}\right|_{\zeta=1}=0; \\ &\theta(0,\tau)=\frac{1}{\mathrm{Pe}_T}\left.\frac{\partial \varsigma}{\partial \zeta}\right|_{\zeta=0}+f\,\theta(1,\tau); \qquad \left.\frac{\partial \theta}{\partial \zeta}\right|_{\zeta=1}=0 \end{aligned} \tag{2}$$

where $f$ represents the efficiency of the effluent-feed heat exchange. In the present work, the effect of the variation of temperature of the cooling medium $\theta_H$ onto the system dynamics is studied. Table 1 reports the values chosen for the parameters.

**Table 1.** Model parameters.

| Da | $\beta$ | $\delta$ | $f$ | $\mathrm{Pe}_M$ | $\mathrm{Pe}_T$ | Le | $n$ |
|---|---|---|---|---|---|---|---|
| 0.15 | 1.4 | 2 | 0.3 | 300 | 300 | 1 | 1 |

The PDEs model is first reduced to a set of ordinary differential equations (ODEs) by approximating it with a cascade of *N* continuous stirred tank reactors (CSTRs). The CSTR cascade is then employed to build a reference solution, and to generate the data needed for the determination of the POD basis.

**Proper orthogonal decomposition**

The POD technique delivers an optimal, in $L^2$ sense, set of empirical orthogonal functions, based on the spatiotemporal set of data obtained from the numerical solution of a full order model (FOM) [6], which in this case is built by means of the CSTR cascade approximation. The set of data, where *t* denotes time and *x* denotes position in space, is extracted (sampled) from the numerical solution and represented in matrix form as:

$$U = \begin{bmatrix} u(x_1,t_1) & u(x_1,t_2) & \cdots & u(x_1,t_M) \\ u(x_2,t_1) & u(x_2,t_2) & \cdots & u(x_2,t_M) \\ \vdots & \vdots & \ddots & \vdots \\ u(x_N,t_1) & u(x_N,t_2) & \cdots & u(x_N,t_M) \end{bmatrix} \tag{3}$$

where $N$ is the number of discretisation points in spatial domain and $M$ is the number of samples taken in time. The POD basis $\Phi = \{\varphi_1, \varphi_2, \ldots, \varphi_N\}$ is then determined by solving the eigenvalue problem $C\Phi = \lambda\Phi$, where $C$ is the autocorrelation matrix, i.e. $C = < U, U^T >$ with angular brackets denoting time-averaging operation. The ordering of the computed eigenvalues from the largest to the smallest induces an ordering of the corresponding eigenvectors, i.e. the POD basis functions, from the most to the least important. Using the determined POD functions, the truncated solution can be expressed as a linear combination of modes and time dependent coefficients as:

$$\tilde{u}(x,t) = \sum_{n=1}^{K} a_n(t)\varphi_n(x) \tag{4}$$

where $K<<N$ is the truncation order, i.e. the number of modes employed, whereas $a_n(t)$ are modal coefficients determined by integration of the system of ODEs obtained by performing the Galerkin projection of the original PDEs – with the solution $u(x,t)$ replaced by its truncation $\tilde{u}(x,t)$ – onto the POD modes [7]:

$$\begin{aligned} &\frac{da_n(t)}{dt} = \left(L\tilde{u}(x,t), \varphi_n(x)\right) \\ &a_n(0) = \left(u(x,0), \varphi_n(x)\right) \qquad n = 1, \ldots, K \end{aligned} \tag{5}$$

with $(\cdot,\cdot)$ denoting inner product and $L$ being a non-linear, parameter dependent operator involving spatial derivatives.

**Results**

Simulations were conducted in the range of $\theta_H \in [-0.07, -0.02]$, by employing the full order model with $N = 150$ tanks in the CSTR cascade, which resulted into a

total of 300 ODEs (150 for each state variable). Figure 2 presents a solution diagram, with bifurcation parameter $\theta_H$, reporting the dimensionless temperature at the outlet of the reactor $\theta_{ex}(1)$. For $-0.0735 < \theta_H < -0.0605$, as also found in Ref. [5], the trajectories have chaotic nature, and become periodic with increasing temperature of the cooling medium. Figure 2 reports also the corresponding values of the orbit entropy, classically defined in the context of information theory as [8]:

$$S = -\sum_{i=1}^{N} p_i \log_2 p_i \,. \tag{6}$$

The reliability of a POD basis depends on the ensemble of observations. Oscillatory – and particularly chaotic – regimes are expected to better span the realm of possible model solutions. The idea is then to generate samples for POD basis construction from the chaotic solution. In fact, the amount of 'information' included in the chaotic orbit is maximum.

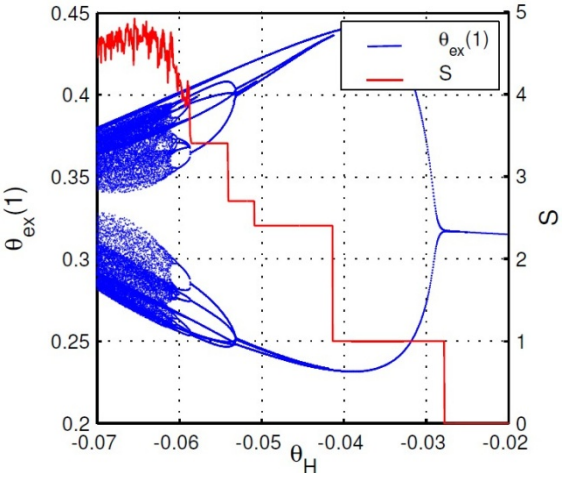


**Figure 2.** Effect of $\theta_H$ on outlet temperature $\theta_{ex}(1)$ and information entropy *S*.

where $p_i$ is the probability that the maximum value of the orbit falls into the $i^{\text{th}}$ interval, and $N$ is the (conveniently high) number of intervals. It can be seen that the entropy of the orbit – which is a non-smooth function of the bifurcation parameter $\theta_H$ – increases gradually with increasing value of the orbits' periodicity and reaches its maximum values for values of $\theta_H$ for which the chaotic character of the solution is observed.

With the purpose of determining a POD basis able to capture the global dynamics of the system, the solution trajectories were sampled at $\theta_H = -0.065$, in the chaos

window. A set of POD basis functions were determined (called here 'POD-A' basis), employing 250 equally spaced samples collected in the transient and at steady state. The reduced models are able to reconstruct the chaotic attractor and, also, to robustly reconstruct the system behaviour for values of $\theta_H$ far from the chaotic window, i.e. far from the sampled condition.

Figure 3 shows a comparison of time series (a) and attractor (b) of a periodic solution for $\theta_H = -0.04$, obtained by means of the full order model (FOM) and its projection onto 24 POD modes – for each state variable – obtained from the chaotic orbit (POD-A/24). The solution obtained by the POD-based model, which has a dimension (48) much smaller than the FOM (300), gives a good approximation of the solution even from a quantitative point of view. For comparison, the results of a standard orthogonal collocation method with 8x3=24 nodes per state variable (same dimension as POD-A/24) are also reported: results are no better than POD-A/24.

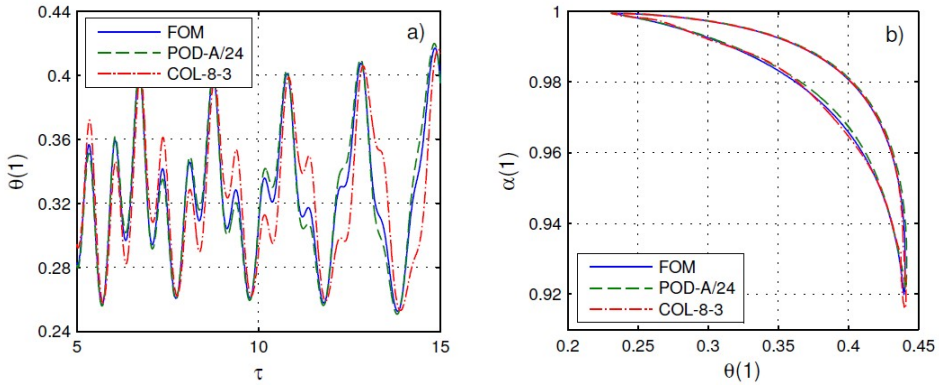


**Figure 3.** Comparison of the time series in early transient for $\theta_H = -0.04$: FOM vs POD-A/24 (POD basis determined at $\theta_H = -0.065$) and COL-8-3 (orthogonal collocation on 3 elements with 8 nodes) (a); corresponding limit cycles (b).

**Conclusions**

Model reduction via empirical spectral methods such as POD/Galerkin proves effective with a pseudo-homogeneous tubular reactor model, suitable to represent oxidation and reduction phases of a chemical looping combustion process based on dynamical operation of packed bed reactors. Chaos sampling is a successful way of optimizing the model reduction process, and provides robust reduced order models that can be employed for values of the operating parameters far from the sampled conditions.

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