Reduced order modelling of chemical reactors with recycle by means of POD-penalty method

Katarzyna Bizon*, Gaetano Continillo

Department of Engineering, University of Sannio, Piazza Roma 21, 82100 Benevento, Italy

**Abstract**

Spectral method based on POD are an effective approach for model reduction but variable boundary conditions are often a problem, since basis functions must satisfy boundary conditions at all times. In this work we introduce weak imposition of boundary conditions by means of a penalty method and apply the method to develop reduced order models of two different chemical reactors. A quantitative analysis shows that, by changing the values of the penalty parameters, arbitrary accuracy can be achieved at the expense of increasing CPU time. Computations can be reduced by a factor 50 at least, by maintaining still acceptable accuracy. Performance parameters of the approach appear to be model dependent.

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1. Introduction

Mathematical models based on first principles of dynamic and spatially distributed chemical reactors generally take the form of a set of partial differential equations (PDEs) describing mass, momentum and energy transfer along the specified spatial domain and over a specified period of time. For practical computational considerations, the original infinite dimensional PDEs are to be reduced to finite dimensional systems of ordinary differential equations (ODEs). Among classical and commonly used numerical approaches, finite differences and finite volume methods are quite simple but result into relatively large sets of ODEs, hence significant computation time is required. It is then desirable to express the original full order model (FOM) as a set of ODEs of the lowest possible order, namely to create a reduced order model (ROM) of the system. This consists in finding a low dimensional approximation for the system of ODEs. A family of methods largely used is a subspace based projection, and particularly the Galerkin method (Galerkin, 1915), which allows to reduce, by proper choice of the functional basis, the number of ODEs necessary to accurately describe the dynamics of the original PDEs model. In spectral methods, the sought solution is expressed in the form of a linear combination of time dependent coefficients and basis functions of the space variables – usually orthogonal polynomials being solutions of the Sturm–Liouville equation (Hesthaven, Gottlieb, & Gottlieb, 2007). However, classic basis may fail to predict the correct solution, especially when an inappropriate functional basis is chosen, or when the truncation order is too low (think for example of the Gibbs phenomenon, Hesthaven et al., 2007, p. 153). An alternative can be an empirical basis set. In order to choose an optimal orthogonal basis, for building a light yet faithful ROM, the *proper orthogonal decomposition* (POD) method can be used. The POD provides a set of empirical basis functions from an ensemble of observations, originating from an experiment or a numerical simulation, representative of the spatiotemporal complexity of the system (Holmes & Lumley, 1996). Depending of the field of application, the POD is also known as Karhunen–Loève decomposition (KL), principal component analysis (PCA), singular value decomposition (SVD) or Hotelling transformation. The method was proposed in the 1940s independently by several scientists including Karhunen (1947), Loève (1945) and Kosambi (1943), and originates from earlier work by Pearson (1901).

The properties of the POD-derived basis, among which the most striking is its optimality – in the $L^2$ norm sense – suggest that it might be the preferred basis to use in many applications. One of the earliest application of the POD is due to Lorenz (1956) who applied the method of empirical orthogonal functions to statistical weather prediction. Another historically important application is the extraction of so-called *coherent structures* in turbulent flows, first proposed in the late 1960s (Lumley, 1967). POD in its original formulation was very often not applicable due to the amount...
of computation necessary for the analysis of large data sets, until the introduction of the so-called method of snapshots or strobos (Sirovich, 1987). The capabilities of POD – both as an order reduction method and as a tool for feature extraction – resulted in the growing popularity of this technique in many fields: among others, fluid mechanics (Lumley, 1967; Sirisup & Karniadakis, 2005), chemical reactor engineering (Bizon, Continillo, Russo, & Smula, 2008; Park & Cho, 1996) or chemical kinetics (Danby & Echekki, 2005). One example of relatively complex chemically reactive system is the fluidized bed. Cizmas, Palacios, O’Brien, and Syamlal (2003) used POD to extract and analyze dominant spatiotemporal patterns of two-dimensional simulations of a gas–solid interaction in a non-reactive bubbling fluidized bed operating at minimum fluidization velocity. In the continuation of the work Yuan, Cizmas, and O’Brien (2005) constructed a POD-based ROM to simulate transport in a non-reactive fluidized bed.

It must be underlined that the empirical character of the method brings about its biggest limitation. Usually, POD employs sampled data from one transient evolution of the system to an asymptotic regime, whereas the process can often have more than one asymptotic regime. Likewise, when changing operating conditions (parameters), occurrence of bifurcations may lead to drastic changes in the spatiotemporal complexity of the solution. Many different strategies of building a suitable, global POD basis for model reduction have been proposed in the literature. In general, a potentially representative ensemble of data from which the POD eigenfunctions will be extracted can be obtained by the combination of the data from different simulations, run at several values of key parameters and/or with different initial conditions (Bizon et al., 2008; Graham & Kevrekidis, 1996) or extracting the basis from a single orbit containing the maximum amount of information about the system behaviour, such as a chaotic orbit (Bizon et al., 2011; Kerchen, Feeny, & Golinval, 2003).

Historically, the treatment of time-dependent boundary conditions has been a matter of some controversy for spectral Galerkin methods. The issue applies to methods employing classic as well as empirical basis functions, and results from the requirement that each function from the chosen basis must individually satisfy the boundary conditions, which often causes the Galerkin formulation to become complicated. An extension of the Galerkin procedure, which allows to handle time-dependent boundary conditions, is known as the tau method (Lanczos, 1938), incorporating boundary conditions by employing additional basis functions. The second, intuitive alternative is the transformation of the problem with time dependent boundary conditions into an equivalent homogenous problem (Park & Cho, 1996; Park & Kim, 2001): this, however, can increase considerably the computational expenses. Another way is the use of a weak form of the Galerkin method (Pahdi & Balakrishnan, 2003), which may account both for the non-homogeneous boundary conditions and for any discontinuities in the spatial domain. The newest approach within POD-based modelling is the use of a penalty function for the enforcement of boundary conditions. This approach was reported so far only in (Sirisup & Karniadakis, 2005), who developed a Galerkin POD-penalty method to construct low-dimensional dynamical systems for non-reacting, unsteady fluid flows with time-dependent boundary conditions. A penalty method, proposed by Lions and Magénès (1972) and studied, among others, by Babuška (1973) in the context of finite element methods, consists of introducing a penalty parameter for imposing the boundary condition weakly. The solution procedure of the original problem may be quite sensitive to the value of the penalty parameter, in terms of relative error with respect to the original problem (Sirisup & Karniadakis, 2005). The same authors also show that the method becomes numerically unstable for too small values of the penalty parameter, whereas, for one example, the relative error tends to increase when the penalty parameter is increased too much in the effort to regain the strong imposition of the boundary conditions. However, it can be said that, in principle, the choice of values of the penalty parameter is arbitrary and, thus, it makes sense, in the construction of optimally reduced order POD models, to determine the optimal value or, at least, a suitable interval for the penalty parameter for the problem under study.

In this paper, we study the application of POD/Galerkin with penalty to the construction of a reduced order model (ROM) of tubular chemical reactors with recycled streams, which imply variable boundary conditions. In our case, the boundary conditions are not only time-dependent but depend on the state variables. The proposed method is first applied, for illustrative purposes, to a simple homogeneous tubular chemical reactor with recycle. The reduced order model of the reactor is built by employing POD coupled with a spectral penalty method, and the influence of the value of the penalty parameter on the accuracy is analyzed. Then, the POD-penalty method is applied for reduction and simulation of a more complex model, namely an idealized model of a circulating fluidized bed combustor (CFBC). Due to the presence of two-time-dependent non-homogeneous boundary conditions for the so-called coarse and fine phases, two penalty parameters are employed in the analysis of this second problem. The two examples reported serve for illustration purposes: the problem size was chosen not too large, in order to make it possible to quickly conduct comparative computations with a standard numerical approach. The latter could not be applied to a full sized problem to obtain results in a reasonable time, as we expect to be possible with our approach.

2. Mathematical models

2.1. Homogeneous plug flow reactor with recycle

The system is described by a simple model of homogeneous tubular chemical reactor with recycle (Fig. 1), in which the second order reaction:

\[ \begin{align*}
A \xrightarrow{r} B \\
r = kC_A^2 
\end{align*} \tag{1} \]

takes place in isothermal mode. The governing equation, in dimensionless form, is the following material balance:

\[ \begin{align*}
\frac{\partial \xi}{\partial \tau} + \frac{\partial \theta}{\partial \zeta} = Da(1-x)^2 
\end{align*} \tag{2} \]

where \( x = (C_A - C_{A0})/C_{A0} \) denotes conversion degree, \( Da = C_{A0}K\tau/Q \) is the Damköhler number, and \( \tau \) and \( \zeta \) are the dimensionless time and space variables respectively. The associated boundary and initial conditions are:

\[ \begin{align*}
x(0, \tau) &= (1 - f)x_{in} + f\theta(1, \tau) \\
x(\zeta, 0) &= x_0(\zeta) 
\end{align*} \tag{3} \]

where \( f \) denotes recycled fraction.

The PDE model is first discretized in space by means of finite difference schemes employing 500 nodes, and then the resulting ODEs are integrated in time. The numerical solutions are then
sampled to construct the POD basis, and also retained as a reference for the validation of the reduced order model. In order to conduct a fair comparison between standard and reduced-order model computations, the main ODE solver employed is the same, i.e. the Adams–Moulton implicit method with adaptive integration time-step, in both cases taken from the public domain VODE library.

### 2.2. Circulating fluidized bed combustor

Starting from the work of Barletta, Marzocchella, Salatino, Kang, and Stromberg (2003), who propose a steady-state model highlighting particle attrition and fragmentation, the CFBC system is idealized as a 1-D distributed, unsteady plug flow tubular reactor followed by a gas–solid separator and a continuous stirred tank to simulate loop-seal (Fig. 2). In the present formulation, the model is isothermal. Quasi-steady approximation is made for interphase momentum exchange. The riser is subdivided vertically into two zones, i.e. dense bed and dilute region (this last including splashing zone and freeboard). Combustion is modelled as a single one-step heterogeneous reaction \((\text{C} + \text{O}_2 \rightarrow \text{CO}_2)\). The ideal fuel has no volatile matter and no ashes. The mass balance for the solids is limited to fixed carbon lumped into two phases of different particle diameter, namely coarse and fine char. Coarse char is consumed by attrition and combustion, whereas fine char is produced by attrition and consumed by combustion.

The material balances for coarse and fine char are given, respectively, in dimensionless form, by:

\[
\begin{align*}
\frac{\partial \alpha_c}{\partial \tau} + \frac{\partial (v_i \alpha_c)}{\partial z} &= -\sigma_0 \alpha_c - \sigma_{f \alpha_c} \\
\frac{\partial \alpha_f}{\partial \tau} + \frac{\partial (v_i \alpha_f)}{\partial z} &= \sigma_0 \alpha_c - \sigma_{f \alpha_f}
\end{align*}
\]

(4)

where \(\alpha_i\) is the dimensionless suspension density of the solids, while \(v_i\) and \(\sigma_i\) are the respective dimensionless velocity and consumption/production rates. The corresponding boundary conditions at the bottom of the bed, resulting from the feed of coarse particles and the recycle of both unburned coarse and fine particles, are expressed as:

\[
\begin{align*}
v_i(0, \tau) \alpha_c(0, \tau) &= k_1 F_i + F_{\text{rec}} \alpha_{c,\text{rec}}(\tau) \\
v_i(0, \tau) \alpha_f(0, \tau) &= F_{\text{rec}} \alpha_{f,\text{rec}}(\tau)
\end{align*}
\]

(5)

where the densities of recycled solids \(\alpha_{i,\text{rec}}\) are calculated from the material balances written for the continuous stirred tank located in the recycle loop:

\[
\begin{align*}
\frac{d\alpha_{c,\text{rec}}}{d\tau} &= k_2 [\eta_i v_i(1, \tau) \alpha_c(1, \tau) - F_{\text{rec}} \alpha_{c,\text{rec}}] \\
\frac{d\alpha_{f,\text{rec}}}{d\tau} &= k_2 [\eta_i v_f(1, \tau) \alpha_f(1, \tau) - F_{\text{rec}} \alpha_{f,\text{rec}}]
\end{align*}
\]

(6)

where \(\eta_i\) represents the efficiencies of the cyclone with respect of the two size classes (coarse and fine).

The material balance for \(\text{CO}_2\) is given by:

\[
\frac{\partial (\alpha_{\text{CO}_2})}{\partial \tau} + \frac{\partial (v_i \alpha_{\text{CO}_2})}{\partial z} = k_3 (\sigma_0 \alpha_c + \sigma_{c \alpha_c})
\]

(7)

with the corresponding (homogeneous) boundary and continuity conditions formulated at the secondary air injection level:

\[
\begin{align*}
\alpha_{\text{CO}_2}(0, \tau) &= 0 \\
[\alpha_{\text{CO}_2}(\zeta, \tau) \rho_{\text{CO}_2}(\zeta, \tau)]_{\zeta = \zeta_{bottom}} &= [\alpha_{\text{CO}_2}(\zeta, \tau) \rho_{\text{CO}_2}(\zeta, \tau)]_{\zeta = \zeta_{top}}
\end{align*}
\]

(8)

and the initial conditions given as follows:

\[
\begin{align*}
\alpha_c(\zeta, 0) &= \alpha_{c,0}(\zeta), & \alpha_f(\zeta, 0) &= \alpha_{f,0}(\zeta), & \alpha_{\text{CO}_2}(\zeta, 0) &= \alpha_{\text{CO}_2,0}(\zeta)
\end{align*}
\]

(9)

The constitutive equations and correlations used in the model of CFBC are reported in Appendix A.

As done earlier for the simple tubular reactor model, the governing equations are first discretized in space by means of finite difference schemes discretization employing 500 spatial nodes – using a staggered grid in this case – and then integrated in time by means of an adaptive Adams–Moulton method. Again, the numerical solutions are sampled to construct the POD basis, and also retained as a reference for the validation of the reduced order model.

### 3. POD-penalty method

#### 3.1. Proper orthogonal decomposition

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 (Holmes & Lumley, 1996). Having a finite dimensional case of \(M\) observations (in time) of the \(N\)-dimensional vector \(x\), we can form a matrix:

\[
U = u(x, t) = \begin{pmatrix} u(x_1, t_1) & \cdots & u(x_1, t_M) \\ \vdots & \ddots & \vdots \\ u(x_N, t_1) & \cdots & u(x_N, t_M) \end{pmatrix}
\]

(10)

It can be shown that a POD basis \(\phi_n(x)_{n=1}^N\) can be obtained by solving the eigenvalue problem:

\[
R\phi = \lambda\phi
\]

(11)

where \(R\) is the autocorrelation matrix defined as:

\[
R(x, x') = \langle u(x, t) u^T(x', t) \rangle
\]

(12)

with the angular brackets denoting time-averaging.

In case of a large number of grid points \(N\), the matrix \(R\) can become too large for the machine capacity or too expensive in terms of computational expenses for the solution of the eigenvalue problem. It was suggested by Sirovich (1987) that, when \(N \gg M\),
the \( N \)-dimensional eigenvalue problem can be reduced to an \( M \)-dimensional eigenvalue problem; this method is called method of snapshots or strobes. The method is based on the hypothesis that data vectors \( u(x,t_m) \) and eigenvectors \( \phi_n \) span the same linear space (Holmes & Lumerly, 1996). This implies that the eigenvectors can be written as a linear combination of the data vectors:

\[
\phi_n(x) = \sum_{m=1}^{M} b_n(t_m) u(x, t_m)
\]  

(13)

After performing some transformations, the coefficients \( b_n \) can be obtained from the solution of:

\[
Cb = \lambda b
\]  

(14)

where \( C \) is autocorrelation matrix defined as:

\[
C(x,x') = [u(x, t)u(x', t)]
\]  

(15)

Using the POD modes, the solution \( u(x,t) \) can be expressed, in a truncated form, by a linear combination of the eigenfunctions:

\[
u_k(x, t) = \sum_{n=1}^{K} a_n(t) \phi_n(x)
\]  

(16)

where \( K \ll N \) is the number of POD modes used for truncation, while \( a_n(t) \) are time-dependent modal coefficients. The Galerkin projection of the original PDE system onto the POD modes yields a set of ODE in the unknowns \( a_n(t) \), that can hence be computed by a standard numerical method for sets of ODE, e.g. Adams–Moulton.

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 can define the cumulative correlation energy captured by the \( K \) successive modes which is given by:

\[
E_K = \sum_{n=1}^{K} \lambda_n
\]  

(17)

In order-reduction application, as a guiding criterion for minimal order selection it is commonly recommended to choose a set made of the first \( K \) eigenfunctions for which \( E_K \geq 99\% \) (Sirovich, 1987). However, as it was reported in Bizon et al. (2008), this criterion is not always reliable, hence the number \( K \) should not be chosen only based on energy estimation but also on a comprehensive analysis of the error of the ROM.

3.2. Spectral penalty method

In spectral methods, the resolution of PDE involves the approximation of the solution of the equation as a truncated series of known, prescribed \textit{a priori}, basis functions \( \{\phi_n(x)\}_{n=1}^{N} \), usually orthonormal functions. The spectral approximation is then achieved by the determination of the expansion coefficients \( \{a_n(t)\}_{n=1}^{N} \). Let us consider the problem (Hesthaven et al., 2007):

\[
\frac{\partial u(x, t)}{\partial t} = Du(x, t), \quad x \in [a, b], \quad t \geq 0
\]  

\[
u(a, t) = g, \quad u(x, 0) = f(x)
\]

(18)

where \( D \) is a non-linear operator that involves spatial derivatives of the dependent variable. We seek a solution in the form:

\[
u_n(x, t) = \sum_{n=1}^{N} a_n(t) \phi_n(x)
\]  

(19)

where \( N \) denotes the order of spectral approximation. In the fundamental Galerkin method – which demands that all basis functions satisfy the boundary conditions individually – we require then that the error of the approximation, defined as:

\[
R_n(x, t) = \frac{\partial u_n(x, t)}{\partial t} - Du_n(x, t)
\]  

(20)

be orthogonal to \( \{\phi_m(x)\}_{m=1}^{N} \), that is:

\[
(R_n(x, t), \phi_m(x)) = 0, \quad m = 1, \ldots, N
\]

(21)

If the prescribed basis is orthonormal, then the time-dependent coefficients can be determined by solving the following set of ODE:

\[
\frac{\partial a_n}{\partial t} = \left[ D \sum_{n=1}^{N} a_n(t) \phi_n(x), \phi_n(x) \right] - p \left[ \sum_{n=1}^{N} a_n(t) \phi_n(a) \right]
\]

(22)

with the initial conditions determined from the initial condition of the dependent variable (Eq. (18)) by projection onto the basis, i.e.: \( a_n(0) = f(x), \phi_n(x) \), \( m = 1, \ldots, N \).

As already mentioned, each function from the chosen basis must individually satisfy boundary conditions. This causes the Galerkin formulation to become complicated, especially when the boundary conditions depend on the state variables, or even just on the independent variable \( t \). Chemical reactors with recycle are one such case. The modification of the Galerkin method in which the basis functions are not required to satisfy the boundary conditions is known as penalty method. The Galerkin projection of the governing equation (Eq. (18)) with the penalty term included, and under the assumption of orthonormality for the chosen basis, is given by:

\[
\frac{\partial a_n}{\partial t} = \left[ D \sum_{n=1}^{N} a_n(t) \phi_n(x), \phi_n(x) \right] - g(t) \phi_n(a), \quad m = 1, \ldots, N
\]  

(23)

where \( g(t) \) signifies the presence of a variable boundary condition, and \( p \) is the penalty parameter enforcing the boundary condition. In general, as already mentioned, the accuracy of the solution scales as \( p^{-1} \) in well resolved simulations, as with \( p \rightarrow \infty \) we have strong imposition of the boundary conditions whereas for small values of \( p \) we have weak imposition (Sirisup & Karniadakis, 2005). Moreover, it is well known (Epshtein & Riviére, 2007) that there exists a threshold value of \( p \) above which the scheme is stable and convergent.

4. Results

4.1. Homogenous tubular reactor with recycle

Our goal is to evaluate the performance of the POD-penalty method, and particularly the influence of the value of the penalty parameter onto the accuracy of the ROM solution. To this aim, the solutions obtained from ROM – constructed by means of the POD method and Galerkin projection with the penalty term for the enforcement of time-variable boundary conditions – are compared with the results obtained from the FOM (full order model) simulation, i.e. the numerical model built using finite differences with a high number of spatial grid points.

In order to build the ROM we first collected snapshots from the FOM simulation to build the POD basis. Simulations were performed for \( f = 0.5 \) and \( Da = 0.4 \). Fig. 3a reports a sequence of snapshots of the conversion degree \( x \) collected during the transient and at steady state of a single simulation. The total number of snapshots (samples) used for the determination of the POD basis was \( M = 100 \), which is lower than the number of state variables (\( N = 500 \)), hence the Sirovich method of snapshots was employed. Fig. 3b presents
the three leading modes, i.e. those corresponding to the three highest eigenvalues. It can be observed that the first mode (blue solid line) closely reflects the conversion degree profile at steady state.

Fig. 4a reports the portion of the eigenvalue spectrum corresponding to the first 50 POD modes. The presence of a single, strictly dominant eigenvalue can be observed: this phenomenon, reported earlier by Bizon et al. (2008), is consistent with the strictly positive entries of the data correlation matrix. Fig. 4b reports the cumulative correlation energy (Eq. (17)) of the leading 15 modes. It is seen that the first mode, coupled with the dominant eigenvalue, already captures more than 99.9% of the cumulative energy. This, according to the common advice, would indicate that using just one mode in the ROM should give acceptable accuracy of the solution. However, this is not going to work in the case of transient behaviour or parametric variations.

The governing equations, modified with the penalty term, were then Galerkin-projected onto the POD basis. To examine the stability and accuracy of the POD-penalty method, a parametric analysis was performed, with the penalty parameter $p$ varied in the range $10^{-4}$ to $10^6$. Fig. 5 compares the solution at the outlet of the reactor at steady state obtained using FOM and ROMs employing 1, 2 and 3 modes for the approximation, as a function of $p$. No difficulties with the convergence of the numerical scheme are observed even when employing very small values of the penalty parameter, corresponding to weak imposition of the boundary conditions. When one POD mode is employed, it can be observed that increasing the value of $p$ leads to very significant divergence of the approximated solution from the reference solution obtained from the FOM simulation.

For all models presented above, the most accurate solutions correspond to $p \approx 1000$, whereas accuracy is found to deteriorate for higher values of $p$. This finding of the lack of improvement of the ROM solution with increasing value of the penalty parameter confirms earlier findings by Sirisup and Karniadakis (2005).

Fig. 6a shows the comparison of time series of the conversion degree at the outlet obtained from FOM and ROM employing 3 modes and two different values of the penalty, that is $p = 1$ and $p = 10^3$. The response is reasonably accurate for both ROM and both values of the penalty parameter (Fig. 6a). Fig. 6b displays the absolute error for the two ROM as a function of time: it appears that both ROM solutions are less accurate in the transient than at steady state, confirming the well known fact that more modes are required for the correct approximation of the transient. Fig. 6b also shows that ROM3, $p = 1000$ provides a better approximation than ROM3, $p = 1$, both during the transient (except at very early times) and at steady state.

Fig. 7 shows the comparison of conversion degree profiles along the spatial coordinate obtained by FOM, ROM3, $p = 1$ and ROM3, $p = 1000$, at one instant during the early transient (at $t = 1$) and at steady state. This figure confirms how ROM3, $p = 1000$
provides a better approximation along the reactor. The solution is acceptable at steady state (Fig. 7b) whereas significant discrepancies are present at early transient (Fig. 7a). To evaluate in detail the accuracy of the ROM with penalty, we calculate a

mean relative error of the approximation at steady state, defined as:

\[
\varepsilon_K = \sqrt{\frac{\sum_{n=1}^{N}(x_{FOM} - x_{ROM,K})^2}{N \sum_{n=1}^{N} |x_{FOM}|}}
\]  

(25)

where \(x_{FOM}\) is the reference solution, \(x_{ROM,K}\) is the approximated solution of order \(K\), and \(N\) is the number of spatial grid points. Fig. 8a shows \(\varepsilon_K\) as a function of the penalty parameter, for ROMs of different order, calculated with respect to the FOM solution. It appears that, even when using only one POD mode, the mean relative error at steady state is smaller than \(10^{-4}\) in the whole examined range of the penalty parameter. For the highest reported order approximation, employing 8 POD modes, the error appears to decrease monotonically with increasing \(p\), while for models of lower order (\(K = 1\) and \(K = 3\)) the minimum of the error occurs for \(p = 1000\), after which it increases drastically.

If we change parameters in the model, namely if we increase the value of Da from 0.4 to 2, meaning that we move relatively far from the conditions for which the POD basis was determined, the error of the approximation increases approximately by one order of magnitude for all ROMs (Fig. 8b) – but still remains lower than \(10^{-3}\) in the whole investigated range of \(p\). A non-monotonic trend is observed now also for ROM8. As with Da = 0.4, characteristic minima for the error are found in a neighborhood of \(p \approx 1000\). Detailed results of the error analysis – the best values of the penalty parameters for the particular cases and the corresponding values of the error – are listed in Table 1. As already mentioned, no monotonic trend for the error was observed, except with ROM4 and ROM8 with Da = 0.4, that is with the value of Damköhler number in the reduced model equal to the one used for the determination of the POD basis.

Obviously, objective of dynamical model reduction is not only the substantial reduction of the number of the degrees of freedom – achieved here quite successfully – but the reduction of the computational expenses. It should be underlined that the reduction of
the order of the model by employing spectral-type methods often – but not necessarily – results in the reduction of the computational time. For example, for linear and bilinear systems most operations, including the projection onto the basis, are executed only once, and then constant matrices are multiplied by time-dependent coefficients during integration (see for example Eq. (24) for constant D). On the other hand, when more complex nonlinearities exist this simplification is not possible, since the presence of such nonlinear terms requires re-evaluation of all approximation sums at each time step of the integration.

To investigate the performance of the ROMs, the CPU time required for the integration of the reduced models was compared with the CPU time elapsed for full order model. The ROMs – of order between 1 and 40 – were compared over a dimensionless time $\tau = 20$ with $Da = 0.4$ and $f = 0.5$. A fixed value of the penalty parameter, $p = 10^3$, was used in all simulations. The results are presented graphically in Fig. 9. The solid line in the figure corresponds to the CPU time for the FOM, that was equal to $t_{CPU} = 5.59$ s, while diamonds correspond to CPU time for ROMs as a function of the order. As it can be observed in Fig. 9, the CPU time for all ROMs up to ROM37 is lower than the CPU time required by the FOM model. Very significant time savings can be obtained with lower order ROMs: note that the CPU time for ROM1 is approximately $10^5$ times shorter than for FOM. The CPU time is not always monotonic with the order variable $K$. This can be related to numerical conditioning issues brought up by the use of an adaptive time step integrator for the ODE set. It must also be recalled that instabilities may be introduced by progressively adding modes, for example when consecutive paired eigenvalues are not both included (Bizon et al., 2008).

It has to be mentioned that not only the number of employed POD modes but also the value of the penalty parameter, $p$, influences significantly the computational time (as mentioned above, for the cases reported in Fig. 9 a constant value was used). In general, the higher the value of the penalty parameter, the higher the computational time: this is due to the fact that increasing $p$ results in a stronger imposition of the boundary conditions, which can lead to the occurrence of steep gradients at the boundary, hence the necessity of smaller integration time steps.

### 4.2. Idealized model of circulating fluidized bed combustor

Following the procedure employed in the previous section for the homogenous tubular reactor with recycle, the POD-penalty method was employed in the construction of the reduced order model of an idealized model of circulating fluidized bed combustor for solid fuel combustion. As before, in order to obtain the reference solution and collect snapshots for the determination of the POD basis, the full order model – employing finite differences on the staggered grid with 500 nodes (resulting in total of 1500 ODEs) – was integrated. The basic model's parameters used in the FOM simulation are reported in Table 2. The number of snapshots used for the determination of the POD basis was equal to $M = 250$, for each of the state variables, i.e. coarse and fine char dimensionless suspension density, and carbon dioxide dimensionless concentration. Fig. 10a shows a sequence of snapshots of the axial fine char concentration whereas Fig. 10b the corresponding eigenvalue spectrum of the POD basis determined from these snapshots. Similar profiles and spectra were obtained for the remaining state variable

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**Table 1**

<table>
<thead>
<tr>
<th>Damköhler number, Da</th>
<th>Modes</th>
<th>Best penalty parameter, $p^*$</th>
<th>Error $\varepsilon$, corresponding to $p^*$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.4</td>
<td>1</td>
<td>$\approx 1.260$</td>
<td>$6.706 \times 10^{-6}$</td>
</tr>
<tr>
<td>0.4</td>
<td>3</td>
<td>$\approx 1.000$</td>
<td>$1.545 \times 10^{-6}$</td>
</tr>
<tr>
<td>0.4</td>
<td>4</td>
<td>$\approx 2.5 \times 10^4$</td>
<td>$2.348 \times 10^{-6}$</td>
</tr>
<tr>
<td>0.4</td>
<td>8</td>
<td>$\approx 18 \times 10^4$</td>
<td>$6.098 \times 10^{-7}$</td>
</tr>
<tr>
<td>2.0</td>
<td>1</td>
<td>$\approx 400$</td>
<td>$4.042 \times 10^{-5}$</td>
</tr>
<tr>
<td>2.0</td>
<td>3</td>
<td>$\approx 560$</td>
<td>$1.815 \times 10^{-5}$</td>
</tr>
<tr>
<td>2.0</td>
<td>4</td>
<td>$\approx 560$</td>
<td>$1.725 \times 10^{-5}$</td>
</tr>
<tr>
<td>2.0</td>
<td>8</td>
<td>$\approx 560$</td>
<td>$9.417 \times 10^{-6}$</td>
</tr>
</tbody>
</table>

---

**Fig. 9.** CPU time of ROM models, for $p = 10^3$, compared with CPU time of FOM simulations.

**Fig. 10.** Snapshots of the fine char concentration from the full order model simulation (a) and corresponding eigenvalue spectrum (b).
Table 2
Basic parameters used in the reference solution calculation.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cyclone properties</td>
<td></td>
</tr>
<tr>
<td>Cyclone efficiency for coarse</td>
<td>1</td>
</tr>
<tr>
<td>Cyclone efficiency for fine</td>
<td>0.9</td>
</tr>
<tr>
<td>Cyclone efficiency for inert</td>
<td>1</td>
</tr>
<tr>
<td>Bed inert properties</td>
<td></td>
</tr>
<tr>
<td>Particle diameter, (d_0) (µm)</td>
<td>300</td>
</tr>
<tr>
<td>Mass density, (\rho_b) (kg/m³)</td>
<td>2600</td>
</tr>
<tr>
<td>Coarse char particle diameter, (d_c) (mm)</td>
<td>3</td>
</tr>
<tr>
<td>Fine char particle diameter, (d_f) (µm)</td>
<td>100</td>
</tr>
<tr>
<td>Mass density, (\rho_c) (kg/m³)</td>
<td>1500</td>
</tr>
<tr>
<td>Coarse sphericity, (\psi_c)</td>
<td>0.63</td>
</tr>
<tr>
<td>Fine sphericity, (\psi_f)</td>
<td>0.73</td>
</tr>
<tr>
<td>Attrition constant, (k_a)</td>
<td>4.9 x 10⁻⁷</td>
</tr>
<tr>
<td>Multiplication factors n₁ and n₂</td>
<td>1</td>
</tr>
<tr>
<td>Coarse slip multiplier, (\gamma_c)</td>
<td>0.5</td>
</tr>
<tr>
<td>Fine slip multiplier, (\gamma_f)</td>
<td>1</td>
</tr>
<tr>
<td>Fluidization velocity, (u_0) (m/s)</td>
<td>5</td>
</tr>
<tr>
<td>Temperature, (T) (K)</td>
<td>1123</td>
</tr>
<tr>
<td>Pressure, (P(P_a))</td>
<td>101,325</td>
</tr>
<tr>
<td>Excess air factor, (\lambda)</td>
<td>1.2</td>
</tr>
</tbody>
</table>

(not reported here). Due to the occurrence of two competing processes, namely fines generation by attrition of coarse char and their burn out the axial concentration profiles (Fig. 10a) manifest a non-monotonic trend, peaking inside the dense zone in the early transient (first, low concentration profiles), while at steady state (highest concentration profiles) two maxima of concentration can be observed – one at the inlet of the reactor, due to recycle of unburned fines, and another between the dense bed and splashing zone (i.e. at \(\xi \approx 0.92\)).

The very fast descent of the eigenspectrum (Fig. 10b) confirms that only a very small number of leading modes is carrying significant information about the system behaviour, with the first eigenvalue being strictly dominant: indeed, the first POD mode (also for the remaining state variables) captures more than 99.9% of the cumulative correlation energy.

The determined POD bases were consecutively employed in the construction of the ROM employing Galerkin projection coupled with the penalty method. Since the boundary condition for carbon dioxide is homogeneous (i.e. we set \(\alpha_{CO_2}(0, \tau) = 0\)), the standard Galerkin projection (Eq. (22)) was employed for Eq. (7), whereas for the mass balances for coarse and fine char (Eq. (4)), for which time-dependent and non-homogeneous boundary conditions are imposed, the penalty method was used (Eq. (24)). This results in a system of ODEs containing two penalty parameters for coarse and fine char particles mass balances, \(p_1\) and \(p_2\), respectively.

As for the homogenous reactor model, the stability and the accuracy of the POD-penalty ROM were analyzed for varying number of POD modes employed in the projection and varying values of penalty – two this time – parameters. Values of \(p_1\) and \(p_2\) were varied independently in the range from \(10^{-4}\) to \(10^4\). The values of the error for the reduced model employing 4 modes, computed for the fine char concentration, are reported in Fig. 11a. It was observed that the method is unstable for values of \(p_2 < 0.224\) (white zone in the bottom of the contour plot in Fig. 11a), where \(p_2\) is the penalty parameter used for the imposition of the boundary condition for fine char particles mass balance. No stability problems were observed when employing the minimal value of \(p_1\), i.e. \(\sim 10^{-1}\), both for this and the ROMs of other orders (not reported here). This is due to the fact that too strong imposition of the boundary conditions for the fine particle mass balance, being initially \(\eta_f(0, \tau = 0) = 0\), can bring about negative, unphysical values of the fines concentration at the reactor inlet. This never happens for the coarse phase, because of the relatively high value of the ratio of feed-to-recycled particles. Fig. 11b reports the corresponding CPU time (for the integration over dimensionless time \(\tau = 5000\)), in the logarithmic color scale, and varying from 0.61 for low values of penalty parameters up to value as high as \(2.5 \times 10^4\) when the value of at least one of the penalty parameters increase substantially. Both for the mean error value and computational time, a universal trend was observed: beyond the zone of instability of the model for the lowest values of \(p_2\) and low values of \(p_2\) – for which the model is already stable but not accurate enough – error convergence (Fig. 11a) and increase of CPU time (Fig. 11b) are almost symmetric, hence a simpler analysis can be conducted by choosing \(p_1 = p_2\).

Fig. 12 reports the values of mean error – calculate for ROM2, ROM4 and ROM7 – at steady state for all state variables (Fig. 12a–c) and the corresponding computational time (Fig. 12d). ROM4 is unstable for the values of \(p_1 = p_2 < 0.224\). The interval of instability for the lower order model, i.e. ROM2, is even larger (\(p_1 = p_2 < 2.818\)), however there is no general rule for the dependence of the ROM stability on the order of the model. On the other hand, unlike the case of homogenous reactor, a quite general trend in terms of dependence of models’ accuracy on the value of the penalty parameter can be observed. Namely, after the instability interval and irregular error behaviour for the lowest values of the penalty parameters, errors start to converge and settle down relatively fast to asymptotic values. This gives the possibility of a rather arbitrary choice of the value of penalty parameter, which should then be picked around the point at which error becomes insensitive. In fact, if we look at Fig. 12d we can observe that, after a relatively large interval of approximately constant value of the CPU time, it starts to increase drastically, with a clear threshold value, whereas error does not change any more. For the cases presented in Fig. 12, the best lower and higher limits of the penalty parameters as well as corresponding values of the CPU time in seconds (for the integration of the system over the dimensionless time \(\tau = 5000\), the corresponding CPU time for the full order model simulation was 462.53 s) are reported in Table 3. These value ranges correspond to the best compromise between ROM accuracy and CPU time.

5. Summary and conclusions

The distributed models of two reactors – a tubular reactor with recycle and a more complex model of a circulating fluidized bed combustor (CFBC) – are considered to the aim of constructing reduced order models via an empirical spectral method. The models are characterized by variable boundary conditions, which represent a known difficulty for a Galerkin approach. The reduced order models are thus built by means of a POD-penalty approach. POD-penalty generates a reduced order model of arbitrary accuracy depending on the value of the penalty parameter. The influence of the value of
the penalty parameter on the accuracy and CPU time is analyzed. Solutions obtained from POD ROMs are compared with the results obtained from the FOM simulation. To examine the stability and accuracy of the POD-penalty method, a parametric analysis is performed, with the penalty parameter \( p \) varied in a wide range (up to 10 orders of magnitude). To evaluate in detail the accuracy of the ROM with penalty, a mean relative error of the approximation at steady state is considered, whereas to assess the computational performance of the ROMs, the CPU time required for the integration of the reduced models is compared with the CPU time elapsed for the FOM. For the tubular reactor, no difficulties with the convergence of the numerical scheme are observed even when employing

**Table 3**

<table>
<thead>
<tr>
<th>Modes</th>
<th>Lowest pen. par., ( p^{*\text{min}} )</th>
<th>Highest pen. par., ( p^{*\text{max}} )</th>
<th>CPU time for ( p^{*\text{min}} ) (s)</th>
<th>CPU time for ( p^{*\text{max}} ) (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>( \approx 12.5 )</td>
<td>( \approx 63 )</td>
<td>0.172</td>
<td>0.156</td>
</tr>
<tr>
<td>4</td>
<td>( \approx 1.25 )</td>
<td>( \approx 18 )</td>
<td>0.656</td>
<td>0.625</td>
</tr>
<tr>
<td>7</td>
<td>( \approx 0.45 )</td>
<td>( \approx 71 )</td>
<td>6.592</td>
<td>9.188</td>
</tr>
</tbody>
</table>
very small values of the penalty parameter, corresponding to weak imposition of the boundary conditions. For the tubular reactor, the CPU time for all ROMs up to ROM37 is lower than the CPU time required by the FOM model, and very significant time savings, up to 10^2 times, can be obtained with lower order ROMs. For the circulating fluidized bed combustor, the ROMs contain two penalty parameters, p_1 and p_2, deriving from variable boundary conditions associated with coarse and fine char particles mass balances respectively. As for the homogenous reactor model, stability and accuracy of the POD-penalty ROM are analyzed for varying number of POD modes employed in the projection and varying values – by 5 orders of magnitude – of penalty parameters. Error convergence and increase of CPU time are almost symmetric with respect to the chosen values of the two penalty parameters p_1 and p_2, hence in this case a simpler analysis can be conducted by setting p_1 = p_2. After an instability interval and irregular error behaviour observed at small values of the penalty parameters, errors converge to low asymptotic values whereas the CPU time increases linearly in a log scale. The optimal value can then be picked around the point at which error becomes insensitive.

The improvement in performance is only due to our approach of model reduction: as stated above, the main ODE solver, common to our standard and penalty computations, was in both cases Adams–Moulton from the public domain VODE library. In conclusion, the POD-penalty method proves generally efficient in overcoming the varying boundary difficulty, however quantitative aspects are model dependent.

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Appendix A. Constitutive relationships and correlations used in the model of CFBC

Voidage ε_bub in the dense bed is estimated from the correlation proposed by Pallarés and Johnsson (2002), taking into account fraction of the bubbles δ_bub:

\[ \varepsilon_b = \delta_{bub} + (1 - \delta_{bub}) \varepsilon_{b} \]

\[ \delta_{bub} = \frac{1}{1 + 1.3 / (u_0 - u_{mf})} \]

\[ f = 0.26 + 0.70 \exp(-3.27)(0.15 + (u_0 - u_{mf}))^{0.13} \]

The voidage profile \( \varepsilon \) above the bottom bed is given by Johnsson and Leckner (1995):

\[ \varepsilon = 1 - (\varepsilon_{b,2} - \varepsilon_b) \exp\left[-a(z - H_b)\right] - (1 - \varepsilon_{ex}) \exp(K[H_{ex} - z]) \]

\[ \varepsilon_{b,2} = 1 - (1 - \varepsilon_{ex}) \exp(K[H_{ex} - H_b]) \]

where \( a = 4u_0/u_0 \) and \( K = 0.23/(u_0 - u_{mf}) \) are decay constants.

The solid entrainment rate \( G_s \) is calculated as (Tasirin & Geldart, 1998):

\[ G_s = 23.7 \rho_s u_0^{2.5} \exp(-5.4u_0 / u_0) \]

The actual average diameter of coarse fuel particles which are fed to the riser is expressed according to (Arena, Chirone, D’Amore, Miccio, & Salatino, 1995):

\[ d = 0.8d_0 \left[ n_1 \left( \frac{n_2}{2} \right) \right]^{-1/3} \]

where \( n_1 \) and \( n_2 \) represent the char multiplication factors due to primary and secondary fragmentation, respectively, while the coefficient 0.8 accounts for particle shrinkage during combustion (Arena et al., 1995).

The rate of production of fine particles per unit mass of coarse char, \( r_a \), is calculated as (Merrick & Highley, 1974):

\[ r_a = \frac{k_a (u_0 - u_{mf})}{d} \quad (A.5) \]

with \( k_a \) being an attrition constant.

Char particle velocities \( u_c \) and \( u_f \) are related to the bed solids velocity \( u_b = G_s / (1 - \varepsilon) \rho_b \) by means of slip multipliers \( \gamma_c \) and \( \gamma_f \) (Barletta et al., 2003).

The combustion rate depends both on the oxygen mass transfer rate and the chemical reaction rate; under the assumption of first order reaction, it can be expressed as:

\[ r_{char} = \frac{6}{\delta d} \frac{M_c}{\delta_{char}} C_{O_2, bulk} \]

with the burning constant \( k_{char} \) defined as:

\[ k_{char} = \frac{1}{(1/K_{rea}) + (1/K_{diff})} \]

The diffusion constant \( K_{diff} \) is obtained from the Sherwood number \( \text{Sh} = K_{diff} / D \) which is calculated from empirical correlations, given respectively by:

\[ \text{Sh} = 2e + 0.695 \text{Sc}^{0.33} \text{Re}^{0.5} \]

for fine particles (Chakraborty & Howard, 1981), and:

\[ \text{Sh} = 2e_{mf} + 0.117 A_b^{0.39} \left( \frac{d_c}{\delta_c} \right) 0.87 \left( \frac{\beta_c}{\rho_c} \right) 0.15 \text{Sc}^{0.33} \]

for coarse char particles (Palchonok, Dolidovich, Andersson, & Leckner, 1992), where indices b and f correspond to char and inert bed particles, respectively.

The kinetic constant of the reaction of oxygen with carbon at the particle surface is modelled in the usual way by the Arrhenius law:

\[ k_{rea} = k_{rea,0} \exp\left(-\frac{E}{RT}\right) \]

with the kinetic parameters chosen as in (Smith, 1971) for anthracite char:

\[ k_{rea,0} = 0.99 \text{kg} / (\text{m}^2 \text{s} \text{kJ}^0), \quad \frac{E}{R} = 8555 \text{K}, \quad n = 1 \]

References


