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# On the use of chemistry-based slow invariant manifolds in DG methods for reactive flows

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## Abstract

In this paper we investigate a recent method for effectively reducing the dimensionality of high-dimensional nonlinear convection-diffusion-reaction problems. This is known as *model reduction*. We apply a particular chemistry-based model reduction approach based on numerical optimization within a discontinuous Galerkin (DG) framework. In doing so, we obtain a system with (significantly) fewer equations for the chemical species whereat the information loss compared to simulating the full system is kept at an agreeable level.

*Keywords:* chemically reacting flows, model reduction, slow invariant manifold, discontinuous Galerkin, adaptive tabulation

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## 1. Introduction

Modeling realistic physical scenarios such as chemically reacting flows often comprises a large number of differential equations as a result of many chemical species interacting with each other. An example is the simulation of fossil or biomass fuel combustion in an internal combustion engine. Therefore, the numerical solution of such equation systems is inevitably associated with a tremendous increase in computational costs.

In order to face this, plenty of different model reduction (MR) approaches have emerged in recent years. These methods aim at simplifying the setting of a differential equation system in one or another way. Some of these approaches attempt to simplify the physical domain [24] while others assume that a (possibly infinite) dynamical system under consideration possesses an *attracting slow*

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<sup>☆</sup>This document is a collaborative effort.

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*invariant manifold* (SIM) and that this manifold can be parameterized by fixing a few distinct variables of the state space, the so-called *reaction-progress variables* (RPVs). These variables correspond to *slow* dynamics and serve as a means to reconstruct the values of the remaining *fast* variables on the SIM. Ideally, the number of slow variables is much smaller than the number of fast ones.

The choice of the slow variables is user-defined and there seems to be no rule of thumb of how to select appropriate RPVs, so we won't make any further assumptions on them. An algorithm to choose RPVs in an automatic way has been suggested in [13].

Roughly speaking, a SIM represents a long-term evolutionary behavior of a system. To keep technicalities about dynamical systems a minimum here, the reader might envisage a SIM as a bundle of trajectories in phase space moving towards a system's fixed point as time passes by and, avalanche-like, drawing other dynamics with them to that point. That is, once the dynamics are "caught" by the manifold, they will stay on it and are driven to the fixed point of the system. This fixed point corresponds to the chemical equilibrium.

In this paper, we will employ a relatively new MR method for characterizing such a SIM [21]. It is based on minimizing the rate of change of the vector field for the chemical kinetics

$$\dot{y}(t) = S(y(t)), \quad y(0) = y_0. \quad (1.1)$$

In [21] this is described as "maximum slowness" inherent to (1.1) and is to yield an optimal SIM. A variant of this will be briefly discussed in section 4.

The benefits of a SIM-related MR approach are twofold:

First, the underlying differential system can be expressed in the slow variables only with the fast variables being reconstructed via the slow ones whenever needed. Second, beside reducing the dimension of a large system, there arises another advantage of MR: When describing the full system via a SIM, a possibly stiff system is made *less stiff*. As a direct consequence, implicit solvers may be replaced by explicit ones and larger time steps can be employed when solving the reduced system.

SIM-related MR approaches have been subject of intense research and the interested reader, who wishes to gain an overview of existing methods in that field, may consult [10] and the references therein.

## 2. Chemically reacting flows

We will focus here on convection-diffusion-reaction (CDR) systems made up of equations of the form

$$\partial_t u + \nabla \cdot (F(u) - A(u)\nabla u) = S(u), \quad (2.1)$$

accompanied with appropriate initial and boundary conditions. The *chemical source term*  $S(u)$  contains the chemical reactions. As a matter of fact, chemical

reactions in several species proceed on various time scales, resulting in a certain stiffness of the differential system. The distinct separation of time scales is, in turn, needed for a clear determination of slow and fast dynamics in phase space. In general, we assume that there are  $n_{\text{spec}}$  chemical species reacting with each other in  $n_{\text{reac}}$  reactions.

Furthermore, we focus on the equations for the species concentrations and will here neglect the evolution equations for the fluid density and velocity. Thus we assume that the vector of conservative variables is given by  $u = (\rho Y)$ , where  $\rho$  is the *mixture density* and the vector of species *mass fractions* is  $Y = (Y_1, \dots, Y_{n_{\text{spec}}})^T$  whose entries must satisfy the physical constraint  $\sum_{j=1}^{n_{\text{spec}}} Y_j = 1$ . With  $\Omega \subset \mathbb{R}^d$ ,  $d > 0$  the governing equations can be written in the form

$$\begin{aligned} \partial_t u &= \mathcal{L}(u) & \text{in } (0, t_{\text{end}}) \times \Omega, \\ u(0, \cdot) &= u_0(\cdot) & \text{in } \Omega, \end{aligned} \quad (2.2)$$

with  $\mathcal{L}(u) := -\nabla \cdot (F(u) - A(u)\nabla u) + S(u)$  and suitable boundary conditions.

The convection and diffusion parts of the above CDR equations (2.1),

$$F(u) = (F_j(u)) \text{ and } A(u)\nabla u = (A(u)\nabla u)_j$$

for  $i = 1, \dots, d$ , are given as follows:

$$F_j(u) = \begin{pmatrix} \rho Y_1 v_j \\ \vdots \\ \rho Y_{n_{\text{spec}}} v_j \end{pmatrix}, \quad (A(u)\nabla u)_j = \begin{pmatrix} j_{1j} \\ \vdots \\ j_{n_{\text{spec}}j} \end{pmatrix}, \quad (2.3)$$

where the velocity  $v = (v_1, \dots, v_d)^T$  is given and  $\nabla \cdot v = 0$ . The diffusion operator for the species, cf. [7, 17] is given by

$$j_k = \rho \frac{W_k}{\overline{W}} \mathcal{D}_k^{\text{mix}} \left( \nabla X_k + \frac{(X_k - Y_k)}{p} \nabla p \right) \quad (2.4)$$

with *mole fraction* of species  $k$

$$X_k = \frac{Y_k \overline{W}}{W_k} \quad (2.5)$$

and *mean molecular weight of the gas mixture*

$$\overline{W} = \left( \sum_{k=1}^{n_{\text{spec}}} \frac{Y_k}{W_k} \right)^{-1}. \quad (2.6)$$

Here,  $W_k$  denotes the *molecular weight* (also known as *molar mass*) of species  $k$ .

As diffusion coefficients are stated as function of pressure  $p$ , temperature  $T$

and mole fractions  $X$ , it is worth knowing conversion formulae from and to *concentrations*  $[X]$ , namely

$$[X_k] = \rho \frac{Y_k}{W_k} = \rho \frac{X_k}{\bar{W}}. \quad (2.7)$$

Concentrations are stated in  $\text{mol m}^{-3}$ .

An empirical formula for the *mixture-averaged diffusion coefficient*, cf. [7, 17] is given by

$$\mathcal{D}_k^{\text{mix}} = \frac{\sum_{j \neq k} X_j W_j}{\bar{W} \sum_{j \neq k} X_j / \mathcal{D}_{jk}}. \quad (2.8)$$

This expression ensures that the roundoff is accumulated similarly in the numerator and denominator, especially when the mixture approaches pure species situations. We exclude here the possibility of a mixture containing a single species only, because in that case (2.8) isn't defined and also because an MR wouldn't make sense at all.

The *binary diffusion coefficient*,  $\mathcal{D}_{jk}$ , for the  $(j, k)$  species pair in (2.8) can be readily computed via

$$\begin{aligned} \mathcal{D}_{jk} &= \frac{3}{16} \frac{\sqrt{2\pi k_B^3 N_A T^3 \frac{W_j + W_k}{W_j W_k}}}{p \pi \sigma_{jk}^2 \Omega^{(1,1)*}}, \\ \sigma_{jk} &= \frac{1}{2}(\sigma_j + \sigma_k). \end{aligned} \quad (2.9)$$

In order to obtain a well-scaled diffusion coefficient, molecular weights should be inserted in g/mol, pressure in bar (which is  $10^5 \text{ Pa}$ ) and lengths for the collision diameter  $\sigma_{jk}$  in Å (which is  $10^{-10} \text{ m}$ ). We use recommended values from [22] for both the Boltzmann constant  $k_B$  and the Avogadro number  $N_A$ .

We want to point out that there are better ways to compute the diffusion coefficient of species  $k$  in a mixture, as done in (2.8), both in view of accuracy and theoretical validation. However, this comes along with increased computational costs and we refer the interested reader to [7, 16, 17].

Furthermore, for the computation of the binary diffusion coefficient (2.9), the *collision integral*  $\Omega^{(1,1)*}$  is fitted as follows [16]

$$\begin{aligned} \Omega^{(1,1)*}(T^*, \tilde{\delta}_{jk}^*) &\approx (a_1 T^{*-a_2} + (T^* + a_3)^{-a_4}) f^{(1,1)}(T^*, \tilde{\delta}_{jk}^*), \\ f^{(1,1)}(T^*, \tilde{\delta}_{jk}^*) &= 1 + \frac{(\exp(a_5/T^*) - \exp(-a_6/T^*)) (\tilde{\delta}_{jk}^*)^2}{2 + 2.5 \tilde{\delta}_{jk}^*}, \end{aligned}$$

where the coefficients are given in Table 1 and  $f^{(1,1)}$  is a simple extension for the Stockmayer potential (i.e.  $\tilde{\delta}_{jk}^* \neq 0$ ).

For the sake of simplicity, we assume that all collision partners in a gas mixture are polar. This deliberates us from distinguishing different cases of

Table 1: Coefficients for interpolating  $\Omega^{(1,1)*}$  as stated in [16]

| $i$   | 1      | 2       | 3       | 4      | 5        | 6   |
|-------|--------|---------|---------|--------|----------|-----|
| $a_i$ | 1.0548 | 0.15504 | 0.55909 | 2.1705 | 0.093193 | 1.5 |

molecular interaction when computing the subsequent quantities for an approximate  $\Omega^{(1,1)*}$ :

$$\begin{aligned}\frac{\varepsilon_{jk}}{k_B} &= \sqrt{\frac{\varepsilon_j}{k_B} \frac{\varepsilon_k}{k_B}}, \\ T^* &= \frac{k_B T}{\varepsilon_{jk}}, \\ \tilde{\delta}_{jk}^* &= \frac{1}{2} \frac{\mu_j^{\text{dip}} \mu_k^{\text{dip}}}{\varepsilon_{jk} \sigma_{jk}^3}.\end{aligned}$$

Data for molecular interaction such as Lennard-Jones potentials  $\varepsilon_k/k_B$  (K), Lennard-Jones collision diameters  $\sigma_k$  (Å) and dipole moments  $\mu_k^{\text{dip}}$  (Debye  $\approx 3.34 \times 10^{-30}$  C m) have been taken from the transport database of [18]. For a thorough discussion and detailed derivation of the above transport concepts, the reader may wish to consult [16, 14].

To close the system we define the pressure  $p$  in accordance with the ideal gas law

$$p = \frac{\rho R T}{\overline{W}}. \quad (2.10)$$

Chemical kinetics cast in the source term  $S(u)$  are modeled with the aid of concentrations (2.7). The differential equations from (1.1) read as

$$\dot{y}_k := [\dot{X}_k] = \sum_{i=1}^{n_{\text{reac}}} (\nu''_{ki} - \nu'_{ki}) q_i, \quad k = 1, \dots, n_{\text{spec}}, \quad (2.11)$$

where  $\nu''_{ki}, \nu'_{ki}$  are the *reverse* and *forward stoichiometric coefficients of species  $k$  in reaction  $i$* , respectively. The *rate of progress of reaction  $i$*  is defined as

$$q_i = \beth_i \left( k_i(T) \left( \prod_{k=1}^{n_{\text{spec}}} [X_k]^{\nu'_{ki}} - \prod_{k=1}^{n_{\text{spec}}} [X_k]^{\nu''_{ki}} \right) \right), \quad i = 1, \dots, n_{\text{reac}}, \quad (2.12)$$

where  $\beth_i = \sum_{k=1}^{n_{\text{spec}}} a_{ki} [X_k]$ . The collision efficiencies  $a_{ki}$  are set to 1 for all  $k$  by default but they may vary depending on the chemical mechanism under consideration.

The *reaction rate constants* in (2.12) for both the forward and reverse direction of reaction  $i$  are given by a *modified 3-parameter Arrhenius law*

$$k_i(T) = A_i T^{\beta_i} \exp \left( -\frac{E_{a,i}}{R_{\text{gas}} T} \right) \quad (2.13)$$

with known values for the prefactor  $A_i$ , temperature exponent  $\beta_i$  and activation energy  $E_{a,i}$ .

### 3. Discontinuous Galerkin approximation

The considered discretization is based on the Discontinuous Galerkin (DG) approach and implemented in DUNE-FEM [9] a module of the DUNE framework [4, 5]. The current state of development allows for simulation of convection dominated (cf. [8]) as well as viscous flow (cf. [6]). We consider the CDG2 method from [6] of up to 2nd order in space and 3rd order in time for the numerical investigations carried out in this paper.

#### 3.1. Spatial discretization

The spatial discretization is derived in the following way. Given a tessellation  $\mathcal{T}_h$  of the domain  $\Omega$  with  $\cup_{K \in \mathcal{T}_h} K = \Omega$  the discrete solution  $u_h$  is sought in the piecewise polynomial space

$$V_h = \{v \in L^2(\Omega, \mathbb{R}^{n_{\text{spec}}}) : v|_K \in [\mathcal{P}_k(K)]^{n_{\text{spec}}}, K \in \mathcal{T}_h\} \quad \text{for some } k \in \mathbb{N},$$

where  $\mathcal{P}_k(K)$  is a space containing polynomials up to degree  $k$ . On quadrilateral or hexahedral elements we replace  $\mathcal{P}_k$  with  $\mathcal{Q}_k$  build by products of Legendre polynomials of up to degree  $k$  in each coordinate.

We denote with  $\Gamma_i$  the set of all intersections between two elements of the grid  $\mathcal{T}_h$  and accordingly with  $\Gamma$  the set of all intersections, also with the boundary of the domain  $\Omega$ . The following discrete form is not the most general but still covers a wide range of well established DG methods. For all basis functions  $\varphi \in V_h$  we define

$$\langle \varphi, \mathcal{L}_h(u_h) \rangle := \langle \varphi, \mathcal{K}_h(u_h) \rangle + \langle \varphi, \mathcal{I}_h(u_h) \rangle \quad (3.1)$$

with the element integrals

$$\langle \varphi, \mathcal{K}_h(u_h) \rangle := \sum_{K \in \mathcal{T}_h} \int_K ((F(u_h) - A(u_h) \nabla u_h) : \nabla \varphi + S(u_h) \cdot \varphi),$$

and the surface integrals (by introducing appropriate numerical fluxes  $\widehat{F}_e, \widehat{A}_e$  for the convection and diffusion terms, respectively)

$$\begin{aligned} \langle \varphi, \mathcal{I}_h(u_h) \rangle &:= \sum_{e \in \Gamma_i} \int_e (\{A(u_h)^T \nabla \varphi\}_e : \llbracket u_h \rrbracket_e + \{A(u_h) \nabla u_h\}_e : \llbracket \varphi \rrbracket_e) \\ &\quad - \sum_{e \in \Gamma} \int_e (\widehat{F}_e(u_h) - \widehat{A}_e(u_h)) : \llbracket \varphi \rrbracket_e, \end{aligned}$$

where  $\{v\}_e = \frac{1}{2}(v^+ + v^-)$  denotes the average and  $\llbracket v \rrbracket_e = (\mathbf{n}^+ \otimes v^+ + \mathbf{n}^- \otimes v^-)$  the jump of the discontinuous function  $v \in V_h$  over element boundaries. For matrices  $\sigma, \tau \in \mathbb{R}^{m \times n}$  we use standard notation  $\sigma : \tau = \sum_{j=1}^m \sum_{l=1}^n \sigma_{jl} \tau_{jl}$ . Additionally, for vectors  $v \in \mathbb{R}^m, w \in \mathbb{R}^n$ , we define  $v \otimes w \in \mathbb{R}^{m \times n}$  according to  $(v \otimes w)_{jl} = v_j w_l$  for  $1 \leq j \leq m, 1 \leq l \leq n$ .

The convective numerical flux  $\widehat{F}_e$  can be an standard upwind flux or the Local Lax-Friedrichs Flux function.

A wide range of diffusion fluxes  $\widehat{A}_e$  can be found in the literature, for a summary see [3]. We choose the CDG2 flux

$$\widehat{A}_e(v) := 2\chi_e(A(v)\mathbf{r}_e(\llbracket v \rrbracket_e))|_{K_e^-} \quad \text{for } v \in V_h, \quad (3.2)$$

which was shown to be highly efficient for advection-diffusion equations and also for the Navier-Stokes equations (cf. [6]). Based on stability results, we choose  $K_e^-$  to be the element adjacent to the edge  $e$  with the smaller volume.  $\mathbf{r}_e(\llbracket v \rrbracket_e) \in [V_h]^d$  is the lifting of the jump of  $v$  defined by

$$\int_{\Omega} \mathbf{r}_e(\llbracket v \rrbracket_e) : \tau = - \int_e \llbracket v \rrbracket_e : \llbracket \tau \rrbracket_e \quad \text{for all } \tau \in [V_h]^d. \quad (3.3)$$

For the numerical experiments done in this paper we use  $\chi_e = \frac{1}{2}\mathcal{N}_{\mathcal{T}_h}$ , where  $\mathcal{N}_{\mathcal{T}_h}$  is the maximal number of intersections one element in the grid can have (cf. [6]).

### 3.2. Temporal discretization

The discrete solution  $u_h(t) \in V_h$  has the form  $u_h(t, x) = \sum_i u_i(t) \varphi_i(x)$ . We get a system of ODEs for the coefficients of  $u(t)$  which reads

$$u'(t) = f(u(t), t) \text{ in } (0, t_{\text{end}}] \quad (3.4)$$

with  $f(u(t), t) = M^{-1}\mathcal{L}_h(u_h(t), t)$ ,  $M$  being the mass matrix which is in our case block diagonal or even the identity, depending on the choice of basis functions.  $u(0)$  is given by the projection of  $u_0$  onto  $V_h$ .

In this paper we use diagonally implicit Runge-Kutta (DIRK) solvers to solve the resulting ODE system, e.g. implicit Euler, Crank-Nicholson, or DIRK34 [1]. These solvers are based on Jacobian-free Newton-Krylov methods (see [19]).

## 4. Computation of slow invariant manifolds

Chemical processes often involve a large amount of chemical species that interact with each other. The question arises whether the number of species and hence the number of differential equations can be reduced without destroying too much information. Methods that are concerned with this question are called *chemistry-based model reduction* methods. Thus whenever we speak of model reduction (MR) methods, we exclusively mean chemistry-based model reduction approaches. Often (but not always [15]) such MR methods are reliant on the existence of *slow invariant (attracting) manifolds*, SIMs for short, [12] that specify system dynamics for a long period, cf. Fig. 1. For recent reactive flow applications in conjunction with non-SIM methods, see [26]. A SIM is usually parametrized by a distinct number of state variables. These state variables usually describe, e.g., the concentrations or mass fractions  $y$  of chemical species and their dynamics are commonly given by an initial value problem (1.1). Here, we



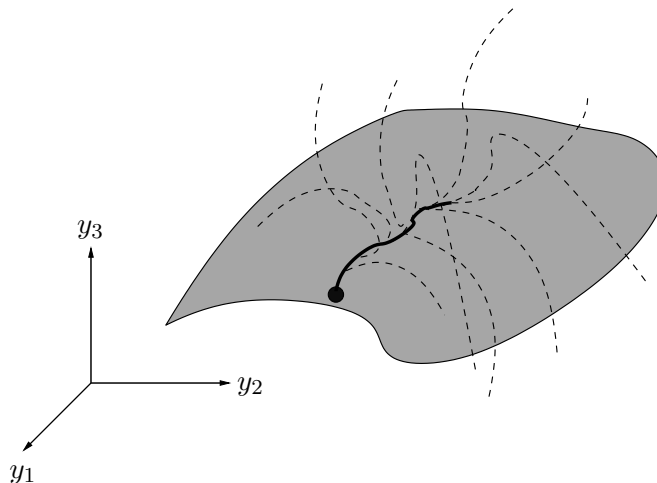


Figure 1: Visualization of a SIM: Trajectories (dashed curves) in phase space – here consisting of three states  $y_1, y_2$  and  $y_3$  – successively bundle on lower-dimensional manifolds (shaded area, bold solid line) until reaching the chemical equilibrium (black dot). This corresponds to a 2D, 1D and 0D manifold.

assume  $y \in \mathbb{R}^{n_{\text{spec}}}$ ,  $n_{\text{spec}} > 1$ . At the very beginning, one selects a distinct and fixed subset of variables of  $y$ , i.e.  $\{r_1, \dots, r_{n_{\text{red}}}\} \subset \{y_1, \dots, y_{n_{\text{spec}}}\}$ , where ideally  $n_{\text{red}} \ll n_{\text{spec}}$ . This subset of state variables that we collect in a vector  $r \in \mathbb{R}^{n_{\text{red}}}$  are called *reaction progress variables* (RPVs) and are used to parametrize the SIM. Roughly speaking, a SIM can be viewed as a function of  $r$ . For it, we define a constant matrix  $B \in \{0, 1\}^{n_{\text{spec}} \times n_{\text{red}}}$  such that the RPVs are related to the full state via

$$r = B^T y. \quad (4.1)$$

Thus each column of  $B$  is made up of a unit vector, with a 1 denoting the species that is to be used as a RPV. The exact choice of  $B$  is *user-defined* and it is out of the scope of this presentation to discuss ways of choosing “good” candidates for RPVs in chemical systems.

As soon as the system dynamics approach the SIM  $\mathcal{M}$ , the RPVs are used to approximate the remaining variables in order to obtain an estimate of the full state  $y$ . The estimation of  $y$  is designated by  $z^{\mathcal{M}}(r)$  and the  $n_{\text{red}}$ -dimensional chemistry-based manifold that is used in the dimension reduction is given by

$$\mathcal{M} = \{y \mid y = z^{\mathcal{M}}(r)\}. \quad (4.2)$$

This process is also referred to as *species reconstruction*.

The RPVs are sometimes associated with the slow dynamics of a dynamical system (1.1) and the unrepresented state variables with the unknown fast modes. This originally goes back to the idea of separating different time scales and lower-dimensional long-term dynamics in order to describe complex systems [10].

For further investigations, we frequently require the computation of the tangent space for a given  $r$ . The matrix whose columns span the tangent space of the manifold  $\mathcal{M}$  at  $z^{\mathcal{M}}(r)$  is denoted by  $T(r) \in \mathbb{R}^{n_{\text{spec}} \times n_{\text{red}}}$ , in accordance with [27]. This matrix can be computed by taking the derivatives of  $z^{\mathcal{M}}(r)$  with respect to  $r$ . Fig. 2 serves as an illustration of a SIM in phase space (the space spanned by the species).

For the computation of such a SIM, we employ an approach that has recently been launched by Lebedz et al. [21]. However, we use a computationally cheaper variation of it. In particular, we do *not* solve  $\dot{y}(t) = S(y(t))$ ,  $t \leq t_{\text{end}}$  in order to get a new “time-averaged” approximation for the full state but evaluate the following minimization problem:

Given current values of the RPVs  $r$  (fixed throughout optimization) and of the full state  $y$ , solve the minimization problem to obtain a new  $y = z^{\mathcal{M}}(r)$ , viz.

$$\min_y \mathcal{J}(y) := \sigma \|J(y)S(y)\|_2^2 \quad (4.3a)$$

subject to

$$Cy - b = 0, \quad (4.3b)$$

$$B^T y - r = 0, \quad (4.3c)$$

$$y_{\text{low}} \leq y \leq y_{\text{up}}. \quad (4.3d)$$

The last statement (4.3d) is to be understood componentwise and, for given lower and upper bounds,  $y_{\text{low}}$  and  $y_{\text{up}}$  respectively, on the variables, ensures that physical properties are maintained during the optimization process (e.g. positivity of mass fractions that might be violated during the solution of (4.3)). The notation from [21] has been slightly adapted for the above problem:  $J(y)$  represents the Jacobian of the source term  $S(y)$ , both being evaluated at the current full state approximation  $y$ . In addition the minimization problem is run with the current approximation  $r$  of the reduced system and represents values of RPVs that parameterize the SIM. Constraint (4.3b) expresses element mass conservation:  $C = (\beta_{kl})$  is a constant mass balance matrix. The vector  $b$  represents the sum of masses, i.e.

$$\sum_{k=1}^{n_{\text{spec}}} \beta_{lk} y_k - b_l = 0, \quad l = 1, \dots, n_{\text{elem}}, \quad (4.4)$$

with  $n_{\text{elem}}$  denoting the number of chemical elements involved in the reactions (i.e. C, H, O, ...) and  $\beta_{kl}$  denoting the number of occurrences of chemical element  $l$  in chemical species  $k$ . Consider  $\text{CH}_4$ , then  $\beta_{\text{C}, \text{CH}_4} = 1$  and  $\beta_{\text{H}, \text{CH}_4} = 4$ . In the current discussion, we assume  $n_{\text{red}} = n_{\text{elem}}$ , but these dimensions need not necessarily be equal. The matrix  $B^T$  has already been introduced in (4.1). The usual Euclidean norm in  $\mathbb{R}^{n_{\text{spec}}}$  is given by  $\|\cdot\|_2$ . A parameter  $\sigma$  in (4.3a) is used for scaling the objective function. A quite similar approach is described in [11].

Due to the continuity of  $\mathcal{J}$  and the compactness of the constraint set (4.3b)–(4.3d), the existence of a solution of (4.3) is guaranteed. Therefore, it may be solved by *any* standard method for nonlinear programming, see e.g. [23]. We use a code by J. Siehr [20] that implements the generalized Gauß-Newton (GGN) to solve (4.3). Although having a linear convergence rate only, the GGN is not affected by a loss of positive definiteness of the underlying linear equation systems. In particular no expensive regularizations of such systems need to be performed as is the case with the conventional Newton’s method.

## 5. Handling invariances

Generally, computing a SIM may be accompanied by small aberrations leading the composition away from the SIM and introducing invariances. These perturbations can result from accumulation of computational errors such as truncation and round-off errors as well as from considering physical transport processes such as diffusion (convection/advection is not assumed to contribute any perturbations in  $u$ ). Diffusion introduces additional time scales into the dynamics thus contributing to the aforementioned drawn-off effects.

In this section we briefly review a method, the so-called *Close-Parallel Assumption*, CPA for short, which “projects” the composition back onto the SIM. It was first laid out by Ren and Pope [27] and we will stay close to their presentation. The concept is graphically highlighted in Fig. 2. Without loss of generality, we

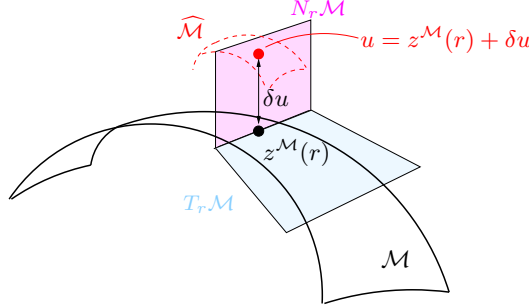


Figure 2: Close-Parallel Assumption: The composition  $u$  on a nearby manifold  $\widehat{\mathcal{M}}$  is thought to be a linear combination of a point  $z^{\mathcal{M}}(r)$  lying on the manifold  $\mathcal{M}$  of interest and an aberration  $\delta u$  into the unrepresented space spanned by the columns of  $U$ . The manifold  $\mathcal{M}$  is parameterized by  $r$ .  $T_r \mathcal{M}$  and  $N_r \mathcal{M}$  represent tangent and normal space at the *manifold point*  $z^{\mathcal{M}}(r)$ , respectively. The figure has been adapted from [27].

assume that a composition  $u$  can be represented via an aberration  $\delta u$  in the form

$$u = z^{\mathcal{M}}(r) + \delta u, \quad (5.1)$$

where  $z^{\mathcal{M}}(r)$  stands for the computed point on the manifold  $\mathcal{M}$  and

$$r := B^T u \quad (5.2)$$

are chosen entries out of  $u \in \mathbb{R}^{n_{\text{spec}}}$  via a predefined transposed *species-selection matrix*  $B \in \{0, 1\}^{n_{\text{spec}} \times n_{\text{red}}}$  whose entries serve as parametrization for the manifold. We won't make any further assumptions about the special structure of  $B$  since the proper selection of slow mode species is a topic of its own and won't be addressed here.

We assume that the aberration takes place in the phase space of unrepresented species, spanned by the columns of  $U \in \{0, 1\}^{n_{\text{spec}} \times (n_{\text{spec}} - n_{\text{red}})}$ . Therefore we set

$$\delta u := U\delta, \quad (5.3)$$

where  $\delta \in \mathbb{R}^{(n_{\text{spec}} - n_{\text{red}})}$  is an unknown to be determined in what follows. Consider the general CDR system

$$\partial_t u + \nabla \cdot (F(u) - A(u)\nabla u) = S(u). \quad (5.4a)$$

together with elementary mass balances

$$Cu = b, \quad (5.4b)$$

with given conservation matrix  $C \in \mathbb{R}^{n_{\text{red}} \times n_{\text{spec}}}$  and  $b \in \mathbb{R}^{n_{\text{red}}}$  a given vector of sum balances.

Premultiplying system (5.4a) with  $B^T$  yields

$$\partial_t B^T u + \nabla \cdot B^T F(u) = B^T \nabla \cdot (A(u)\nabla u) + B^T S(u). \quad (5.5)$$

Using (5.1) and (5.2) we can write

$$\begin{aligned} \partial_t r + \nabla \cdot B^T F(z^{\mathcal{M}}(r) + U\delta) &= B^T \nabla \cdot (A(z^{\mathcal{M}}(r) + U\delta)\nabla(z^{\mathcal{M}}(r) + U\delta)) \\ &+ B^T S(z^{\mathcal{M}}(r) + U\delta). \end{aligned} \quad (5.6)$$

The CPA now states that the composition is drawn onto another manifold that is close by and similar in structure as those being computed (hence the term parallel – quasi-isomorphic would probably be a better-suited description), cf. Figure 2.

We now apply the CPA to (5.6): First we note that diffusion depends only on derivatives of  $u$ . The composition  $u$  in turn is assumed to lie on a nearby-manifold. Thus we have

$$B^T \nabla \cdot (A(z^{\mathcal{M}}(r) + U\delta)\nabla(z^{\mathcal{M}}(r) + U\delta)) \approx B^T \nabla \cdot (A(z^{\mathcal{M}}(r))\nabla z^{\mathcal{M}}(r)). \quad (5.7)$$

As a matter of fact, chemical kinetics cast in the source term  $S(\cdot)$  are sensitive to perturbations. Applying again the CPA, namely the property of being *nearby*,

we can perform a first order Taylor expansion around  $z^{\mathcal{M}}(r)$  w.r.t. the second term of the RHS of (5.6):

$$B^T S(z^{\mathcal{M}}(r) + U\delta) \approx B^T S(z^{\mathcal{M}}(r)) + B^T J(z^{\mathcal{M}}(r))U\delta \quad (5.8)$$

where  $J(z^{\mathcal{M}}(r)) = DS(z^{\mathcal{M}}(r))$  denotes the Jacobian of the chemical source term.

Using (5.7) and (5.8) and the fact that the convective part depends on derivatives of  $u$  and since we assume that  $u$  is close to  $z^{\mathcal{M}}(r)$ , compare (5.7) for the same argumentation, we achieve the *CPA formulation* of system (5.4a)

$$\begin{aligned} \partial_t r + B^T \nabla \cdot F(z^{\mathcal{M}}(r)) &= \underbrace{B^T (\nabla \cdot (A(z^{\mathcal{M}}(r)) \nabla z^{\mathcal{M}}(r)) + S(z^{\mathcal{M}}(r)))}_{=:Q} \\ &+ \underbrace{B^T J(z^{\mathcal{M}}(r))U\delta}_{=:R}. \end{aligned} \quad (5.9)$$

We refer to  $Q$  as “*first approximation*” and to  $Q + R$  as “*second approximation*” or *CPA approximation*. We frequently write *1st* and *2nd* as mnemonic for these approximations.

The first approximation neglects possible drawn-off effects whereas the second approximation takes them into account via a “*correction term*”  $R$ .

Up to now, we have not specified how to calculate the unknown perturbation  $\delta$  of (5.9). This will be done in what follows.

Assuming that the aberration  $\delta u$  lies in the normal space of the manifold  $\mathcal{M}$ , we multiply (5.4a) by  $N^T(r) \in \mathbb{R}^{((n_{\text{spec}} - n_{\text{red}}) \times n_{\text{spec}})}$  (note that the columns of  $N(r)$  span the normal space of  $\mathcal{M}$  at  $z^{\mathcal{M}}(r)$ ):

$$\begin{aligned} N^T(r) \partial_t (z^{\mathcal{M}}(r) + U\delta) + N^T(r) \nabla \cdot F(z^{\mathcal{M}}(r) + U\delta) &= \\ N^T(r) \nabla \cdot (A(z^{\mathcal{M}}(r) + U\delta) \nabla (z^{\mathcal{M}}(r) + U\delta)) + N^T(r) S(z^{\mathcal{M}}(r) + U\delta). \end{aligned} \quad (5.10)$$

Obviously,

$$N^T(r) \partial_t (z^{\mathcal{M}}(r) + \delta u) = N^T(r) \underbrace{Dz^{\mathcal{M}}(r)}_{=:T(r)} \partial_t r + N^T(r) \partial_t \delta u. \quad (5.11)$$

Noting that  $N^T(r)T(r) = 0$  and applying the CPA on (5.11) again, we get

$$N^T(r) \partial_t \delta u \approx 0. \quad (5.12)$$

Recall

$$\nabla \cdot F(z^{\mathcal{M}}(r) + U\delta) \approx \nabla \cdot F(z^{\mathcal{M}}(r)).$$

Moreover, we reason in the sense of the CPA that the convective part makes the composition  $u$  evolve only along the tangent space  $T(r) \forall z^{\mathcal{M}}(r)$ , hence

$$N^T \nabla \cdot F(z^{\mathcal{M}}(r)) \approx 0.$$

Thus we end up with

$$N^T(r)\partial_t u = N^T(r)\nabla \cdot (A(z^{\mathcal{M}}(r) + U\delta)\nabla(z^{\mathcal{M}}(r) + U\delta)) + N^T(r)S(z^{\mathcal{M}}(r) + U\delta) \approx 0.$$

Using the same argumentation as above, see (5.8), we obtain

$$0 \approx N^T(r)\nabla \cdot (A(z^{\mathcal{M}}(r))\nabla z^{\mathcal{M}}(r)) + N^T S(z^{\mathcal{M}}(r)) + N^T J(z^{\mathcal{M}}(r))U\delta. \quad (5.13)$$

After some rearrangement we arrive at

$$N^T(r)J(z^{\mathcal{M}}(r))\delta u = -N^T(r)(\nabla \cdot (A(z^{\mathcal{M}}(r))\nabla z^{\mathcal{M}}(r)) + S(z^{\mathcal{M}}(r))) \quad (5.14)$$

for the unknown  $\delta u = U\delta$ . The dynamics cast in the chemical source term  $S(\cdot)$  are formed using the law of mass action. As a result, the Jacobian  $J(\cdot)$  of that source term is *singular*. This makes matrix  $N^T(r)J(z^{\mathcal{M}}(r))$  rank-deficient. To remove some of the ambiguity in the definition of  $\delta u$  we make use of the elementary mass balance  $Cu = b$ , recall (4.3b), where  $C \in \mathbb{R}^{n_{\text{red}} \times n_{\text{spec}}}$ . Using representation (5.1) we arrive at  $Cz^{\mathcal{M}}(r) + C\delta u = b$ . The value  $z^{\mathcal{M}}(r)$  on the slow manifold also satisfies the same mass balance law, i.e.  $Cz^{\mathcal{M}}(r) = b$  so that the abbreviation satisfies  $C\delta u = 0$ . Summing up,  $\delta u$  solves the following  $n_{\text{spec}} \times n_{\text{spec}}$  linear system

$$\begin{bmatrix} N^T(r)J(z^{\mathcal{M}}(r)) \\ C \end{bmatrix} \delta u = - \begin{bmatrix} N^T(r)(\nabla \cdot (A(z^{\mathcal{M}}(r))\nabla z^{\mathcal{M}}(r)) + S(z^{\mathcal{M}}(r))) \\ 0 \end{bmatrix}. \quad (5.15)$$

Finally, it should be said that the matrix  $N(r)$  can be obtained by an orthonormalization of  $T(r) := Dz^{\mathcal{M}}(r)$ . Once the matrix  $T(r)$  has been computed, we use a QR factorization  $T(r) \equiv T = [Q_1, Q_2][R, 0]^T$ , where  $N(r)$  agrees with  $Q_1$  and it holds  $N^T(r)T(r) = 0 \in \mathbb{R}^{((n_{\text{spec}} - n_{\text{red}})) \times n_{\text{red}}}$ .

Note however that Eq. (5.15) differs from the presentation in [27] insofar as we have added constraints to take violations of the mass balance into account. However for the simple system considered in the numerical experiments in [27] there are no mass balance equations and the Jacobian is not singular, so that in this case the system (5.15) corresponds to the definition given in [27]. For more complicated systems however we found that adding mass conservation was essential for obtaining a well defined method. The matrix  $C$  can be identified with the “element matrix” because it counts the number of atoms of chemical elements (such as O, H, ...) in each of the  $n_{\text{spec}}$  chemical species. Thus our approach bears a certain resemblance with that presented in [28].

### 5.1. Implementation of the manifold

Every time  $z^{\mathcal{M}}(r)$  is to be evaluated, equation (4.3a) has to be solved. This is computationally expensive, especially in the 2d or 3d computations where similar states  $r$  can be present in the system (see also Section 7.3 and 7.4). Therefore, an appropriate way of tabulating already computed similar states needs to be used. In this work we used an adaptive interpolation approach.

We discretize the space spanned by the reduced variables using a linear Lagrange Finite Element approach on a Cartesian grid where the computational domain is given by  $[0, 1]^r$ . We start with a very coarse grid that contains one or four cells. For every Lagrange Point, here the vertices of the cells, that lies below the line of mass conservation (see Figure 4), the optimization problem (4.3a) is solved. We skip points above the line of mass conservation by setting the interpolation values to zero.

Whenever  $z^{\mathcal{M}}(r)$  is to be evaluate, instead of solving equation (4.3a) we interpolate the values for  $z^{\mathcal{M}}(r)$  on the cell of the Cartesian grid the point  $r$  is located in. The search of this cell can be obtained by simple integer arithmetic since we rely on a Cartesian grid.

To resolve  $z^{\mathcal{M}}(r)$  appropriately we adaptively refine the grid cells until the interpolation of the manifold at the cell center  $r_c$  is close enough to the computed solution or a maximal refinement level is reached. The criterion reads as follows:

$$\|z^{\mathcal{M}}(r_c) - \Pi_{z^{\mathcal{M}}}(r_c)\| \leq \alpha \text{ or } l(E_r) = l_{\max}. \quad (5.16)$$

For points  $r$  within cells that satisfy this criterion the interpolation  $\Pi_{z^{\mathcal{M}}}$  is used to approximate  $z^{\mathcal{M}}(r)$ .

A resulting interpolation grid is presented in Figure 4 with threshold  $\alpha = 10^{-8}$  and maximal refinement level  $l_{\max} = 5$ . In this example we started with one cell and cells are refined by quartering.

Another tabularization strategy frequently applied in practical CFD simulations is described in [25].

## 6. Discretization of the reduced system

We now need to discretize the evolution equation (5.9) for the reduced variable  $r$ . Using operator notation, we have to solve  $\partial_t r = \mathcal{L}_2(r)$  with

$$\begin{aligned} \mathcal{L}_2(r) := & -\nabla \cdot (B^T F(z^{\mathcal{M}}(r)) - A(z^{\mathcal{M}}(r)) \nabla z^{\mathcal{M}}(r)) \\ & + B^T (S(z^{\mathcal{M}}(r)) + J(z^{\mathcal{M}}(r)) U \delta) \end{aligned}$$

where  $\delta$  is given by (5.15):

$$\begin{bmatrix} N^T(r) J(z^{\mathcal{M}}(r)) U \\ CU \end{bmatrix} \delta = - \begin{bmatrix} N^T(r) (\nabla \cdot (A(z^{\mathcal{M}}(r)) \nabla z^{\mathcal{M}}(r)) + S(z^{\mathcal{M}}(r))) \\ 0 \end{bmatrix}.$$

We will concentrate on the second approximation which is given by the operator  $\mathcal{L}_2$ . The discretization of the first approximation is simpler and can be inferred from the following discussion. We assume in the following that the parametrizations  $z^{\mathcal{M}}$  of the slow manifold and of its normal space  $N^T$  are available for example using the tabularization discussed in the previous section.

First we need to discuss the computation of the correction  $\delta = \delta(r)$ . As already mentioned  $N^T(r) J(z^{\mathcal{M}}(r))$  is rank-deficient. But we assume that by adding the mass balance constraints  $CU\delta = 0$  the full matrix

$$\begin{bmatrix} N^T(r) J(z^{\mathcal{M}}(r)) U \\ CU \end{bmatrix}$$

has full column rank. In this way the linear least squares solution is unique. We must, however, be able to cope with the situation when the left hand side matrix of (5.15) is rank deficient (up to machine precision), i.e. its rank is smaller than the minimum of its row and column number. In such a case, we can solve the linear system (5.15) by using the pseudo inverse. In our implementation we use a linear least squares method based on singular value decomposition, employing LAPACK's `dge1sd` routine [2]. This can handle a possible rank deficiency.

The evolution operator for  $r$  differs from the general form discussed in Sec. 3 in two ways:

1. The diffusion is not simply of the form  $A(r)\nabla r$  but involves a non-linearity  $A(z^{\mathcal{M}}(r))\nabla z^{\mathcal{M}}(r)$ . Thus our DG approximation can not be directly used for this equation.
2. The operator is not in divergence form due to the second order correction.

To overcome these problems, we study the following discrete weak formulation of the evolution equation for  $r$ :

$$\begin{aligned} u_h &= z^{\mathcal{M}}(r_h) \\ \langle \varphi, w_h \rangle &= \langle \varphi, \mathcal{L}_a(u_h) \rangle \\ \delta_h &= - \begin{bmatrix} N^T(r_h)J(u_h)U \\ CU \end{bmatrix}^{-1} \begin{bmatrix} N^T(r_h)w_h \\ 0 \end{bmatrix} \\ \langle \varphi, \partial_t r_h \rangle &= \langle \varphi, \mathcal{L}_b(r_h, u_h, w_h, \delta_h) \rangle \end{aligned}$$

where  $\mathcal{L}_a$  and  $\mathcal{L}_b$  are advection-diffusion-reaction operators of the form discussed in Sec. 3:

$$\begin{aligned} \mathcal{L}_a(u_h) &:= \nabla \cdot (A(u_h)\nabla u_h) + S(u_h) \\ \mathcal{L}_b(r_h, u_h, w_h, \delta_h) &:= -\nabla \cdot B^T F(u_h) + B^T(w_h + J(u_h)U\delta_h) \end{aligned}$$

Note that this formulation is formally equivalent to the original equation and the operators are of the right form. In fact  $\mathcal{L}_a$  is equivalent to the original evolution operator for the full system (neglecting the advection term). The second operator  $\mathcal{L}_b$  is a convection-reaction operator. Note in our implementation we do not store the auxiliary variables  $u_h, w_h, \delta_h$ . Both  $u_h$  and  $\delta_h$  can be directly computed pointwise for given values of  $r_h$  and  $w_h$  so they can be obtained as needed. The same also holds for  $w_h$  which can be computed on a single element  $K \in \mathcal{T}_h$  due to the local structure of the discontinuous Galerkin method. Thus  $\mathcal{L}_2$  can be approximated using a single grid traversal and without storing additional variables - especially no variables of the size of the full system are required.

## 7. Numerical results

We will apply the approaches from the preceding sections to some examples. The first one is a rather simple one with a known SIM that allows direct comparison with numerically generated results. The second example is a notional isothermal mechanism paving the way for future investigations on realistic combustion chemistry.



### 7.1. A generalized 1d Davis-Skodje-like toy problem

The following academic example of a simple diffusion-reaction system in 2 variables has been taken from [27]. This model has the advantage that an *analytical* SIM is known.

Let  $z \equiv z(x, t)$  where  $z = (z_1(x, t), z_2(x, t))^T$ . We skip the dependence on  $x$  and  $t$  until further notice in order to make the representation more readable. Defined on  $\Omega := (0, 1)$ , the test model is given by

$$\begin{aligned}\partial_t z_1 &= \frac{c}{\varepsilon} w(z) - dz_1 + \partial_x(\mathcal{D}_1 \partial_x z_1) \\ \partial_t z_2 &= -\frac{1}{\varepsilon} w(z) - \frac{z_1}{(1 + bz_1)^2} + \partial_x(\mathcal{D}_2 \partial_x z_2),\end{aligned}\tag{7.1}$$

where  $w(z) = z_2 - z_1/(1 + az_1)$  and  $\mathcal{D}_2(x) = \mathcal{D}_1 + ex$ . Both the “stiffness gap” and the diffusion coefficient for  $z_1$  are held fixed in all cases of interest, namely  $\varepsilon = 0.01$  and  $\mathcal{D}_1 = 1$ . Parameters  $a$  to  $e$  vary and are displayed in Table 2. System (7.1) is equipped with the following boundary and initial conditions

$$z(t, x = 0) = \begin{bmatrix} 0 \\ 0 \end{bmatrix} \quad z(t, x = 1) = \begin{bmatrix} 1 \\ \frac{1}{1+a} \end{bmatrix}$$

and

$$z(t = 0, x) = \begin{bmatrix} x \\ \frac{z_1}{1+az_1} \end{bmatrix}.$$

These conditions are already on the SIM.

The reaction-progress variable (RPV) will be  $z_1$ , hence

$$B = \begin{bmatrix} 1 \\ 0 \end{bmatrix} \quad U = \begin{bmatrix} 0 \\ 1 \end{bmatrix}.$$

An analytical SIM is given by

$$z = z^{\mathcal{M}}(z_1) = \begin{bmatrix} z_1 \\ \frac{z_1}{1+az_1} \end{bmatrix},\tag{7.2}$$

so that in a fully analytical scenario,  $z_2 = z_1/(1 + az_1)$ .

Differentiation of (7.2) with respect to the RPV  $z_1$  yields the matrix  $T(z_1)$  whose column spans the (analytical) tangent space at  $z^{\mathcal{M}}(z_1)$ ,

$$T(z_1) := Dz^{\mathcal{M}}(z_1) = \begin{bmatrix} 1 \\ \varphi(z_1) \end{bmatrix},$$

where  $\varphi(z_1) := 1/(1 + az_1)^2$ .

Since  $N^T(z_1)T(z_1) = 0$ , the  $2 \times 1$  matrix  $N(z_1)$  for the analytical normal space can be readily computed via

$$N(z_1) = \frac{1}{\sqrt{\varphi(z_1)^2 + 1}} \begin{bmatrix} -\varphi(z_1) \\ 1 \end{bmatrix}.$$

Table 2: Parameters for system (7.1), taken from [27]

|     | case 1 | case 2 | case 3 | case 4 |
|-----|--------|--------|--------|--------|
| $a$ | 0      | 1      | 0      | 1      |
| $b$ | 1      | 1      | 0      | 1      |
| $c$ | 1      | 1      | 1      | 1      |
| $d$ | 2      | 1      | 1      | 2      |
| $e$ | 0      | 0      | 3      | 3      |

In terms of (5.9), we obtain

$$\begin{aligned} \partial_t z_1 &= -dz_1 + \mathcal{D}_1 \partial_x^2 z_1 \\ &+ \frac{c}{c\varphi(z_1) + 1} \left( dz_1 \varphi(z_1) - \frac{z_1}{(1 + bz_1)^2} - \varphi(z_1) \mathcal{D}_1 \partial_x^2 z_1 + \partial_x \mathcal{D}_2(x) \partial_x z_2 \right). \end{aligned} \quad (7.3)$$

The first term in the RHS of (7.3) corresponds to the first approximation while the second to the correction term introduced by the CPA. The complete RHS denotes the second approximation. In Fig. 3, we compare our numerical results with the analytical ones obtained by (7.3).

Note however that in this special case we have no mass balance equations so that the system defining the correction  $\delta$  reduces to  $N^T J(z^{\mathcal{M}})U$  which is always not equal to zero, i.e., in contrast to realistic chemical reaction source terms, the Jacobian is *not* singular for this model.

Our results shown in Fig. 3 agree quite well with the results presented in [27]. In all considered cases, the second approximation obtained with the CPA is closer to the full solution than the analytical first approximation. The numerical second approximation has been computed via *in situ* calculations and storage in an adaptive look-up table as described in preceding sections. The *in situ* calculations have been performed by solving an optimization problem of the form (4.3) at the finest refinement level.

## 7.2. Hydrogen combustion model

We consider an isothermal ( $T = 1000$  K) hydrogen mechanism that can be found in Table 3 and we compute numerically a two-dimensional SIM. The mechanism has been taken from [12]. Unfortunately, it does not possess any known analytical SIM so that any simulations concerning a reduced description can only be verified by comparing them to the computational solution of the full system.

The species in the above mechanism are put into a vector in order of appearance

$$U = (u_1, u_2, u_3, u_4, u_5, u_6)^T.$$

Thus,  $u_1, u_2, \dots, u_6$  corresponds to  $\text{H}_2, \text{H}, \text{O}_2, \text{O}, \text{H}_2\text{O}$  and  $\text{OH}$ , respectively. In order to derive the mass balance matrix from Eqs. (4.3b) and (4.4), we count

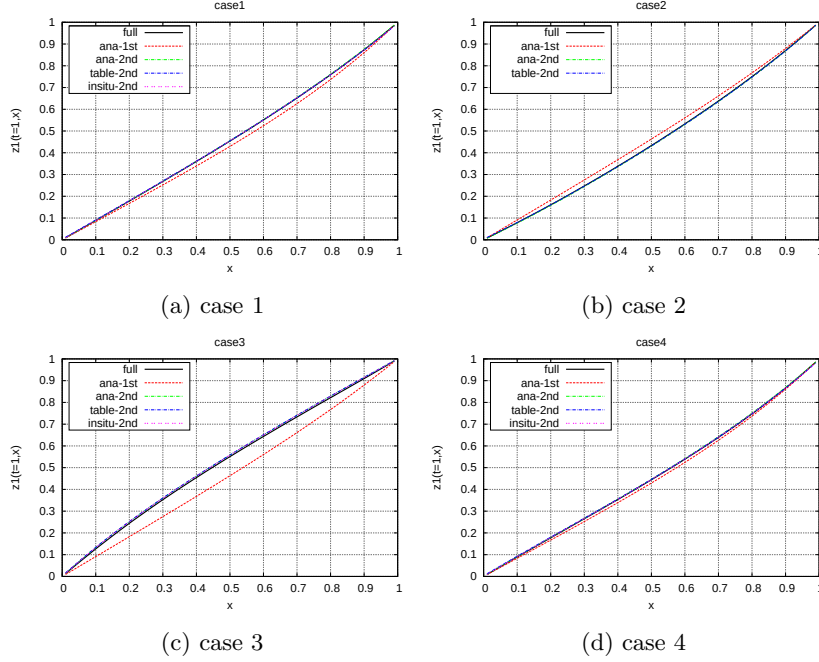


Figure 3: Results for the four test cases, cf. Table 2. For each case, the solution of the full set of equations (7.1) at  $t_{\text{end}} = 1$  is plotted and compared with the solution of the analytical first and second approximation (7.3) as well as with fully numerically obtained solutions for the second approximation (this includes computation of normal spaces, too).

the number of occurrences of the two chemical elements H and O in each of the six chemical species, see. Tab. 5.

The conservations laws, e.g. in concentrations (4.4), are given by

$$\begin{aligned} 2u_1 + u_2 + 2u_5 + u_6 &= b_1, \\ 2u_3 + u_4 + u_5 + u_6 &= b_2. \end{aligned}$$

We take  $b_1 = 2$  and  $b_2 = 1$  as in [12].

Had the conservation laws been stated in mass fractions  $Y_k$ , then physical constraints would have been imposed by  $\sum_k Y_k = 1$ .

The equilibrium (in concentrations) is

$$u_{\text{eq}} = (0.27, 0.05, 0.135, 0.02, 0.7, 0.01)^T. \quad (7.4)$$

We take  $r = (u_1, u_5)^T$ , i.e.  $\text{H}_2$  and  $\text{H}_2\text{O}$  as RPVs.

We start our investigation by studying the pure ODE, i.e. neglecting advection or diffusion. As can be seen from Fig. 5, the CPA approximation 5.9 yields

Table 3: Simple test mechanism. Forward ( $k_f$ ) and reverse ( $k_r$ ) rate coefficients are assumed to be temperature-independent. The rate coefficients have been computed via (2.13), with  $\beta_i = E_{a,i} = 0$ .

| Reaction                                                        | $k_f$ | $k_r$  |
|-----------------------------------------------------------------|-------|--------|
| $\text{H}_2 \rightleftharpoons 2 \text{H}$                      | 2.    | 216.   |
| $\text{O}_2 \rightleftharpoons 2 \text{O}$                      | 1.    | 337.5  |
| $\text{H}_2\text{O} \rightleftharpoons \text{H} + \text{OH}$    | 1.    | 1400.  |
| $\text{H}_2 + \text{O} \rightleftharpoons \text{H} + \text{OH}$ | 1000. | 10800. |
| $\text{O}_2 + \text{H} \rightleftharpoons \text{O} + \text{OH}$ | 1000. | 33750. |
| $\text{H}_2 + \text{O} \rightleftharpoons \text{H}_2\text{O}$   | 100.  | 0.7714 |

Table 4: Hydrogen combustion mechanism

| Elements | Chemical species |            |              |            |                      |             |
|----------|------------------|------------|--------------|------------|----------------------|-------------|
|          | $\text{H}_2$     | $\text{H}$ | $\text{O}_2$ | $\text{O}$ | $\text{H}_2\text{O}$ | $\text{OH}$ |
| H        | 2                | 1          | 0            | 0          | 2                    | 1           |
| O        | 0                | 0          | 2            | 1          | 1                    | 1           |

Table 5: Define matrix  $C$  from Eq. (4.3b)

(slightly) better results than simply applying the first approximation for different initial data. From Tab. 6 one can also observe that the aim of reducing the stiffness in the system by the reduction to the slow manifold has been achieved. To obtain an upper bound for the stability of the method we used a nested interval approach, integrating the system to the equilibrium state for different time steps. A method is termed stable for a given time step if in each step the mass balance equations is satisfied. In the case of an implicit method, the Newton scheme converged, and the method approximated the correct equilibrium state at  $t_{\text{end}} = 2$ . It turns out that the reduced system can be solved using an explicit method with a larger time step than with an implicit treatment of the full system. For example, the forward Euler method for the reduced system is stable with twice the time step as for the full system using the implicit Euler method. To clarify this issue, we should point out that the Newton method used for treating the nonlinear system was quite basic and more sophisticated

|                | first  | second | full   |
|----------------|--------|--------|--------|
| explicit RK(1) | 0.0021 | 0.0021 | 0.0001 |
| implicit RK(1) | 0.0041 | 0.0040 | 0.0011 |
| explicit RK(2) | 0.0028 | 0.0028 | 0.0001 |
| implicit RK(2) | 0.0083 | 0.0080 | 0.0006 |

Table 6: Numerically estimated maximum time step for the full ODE and the first and second order approximation using different time stepping methods.

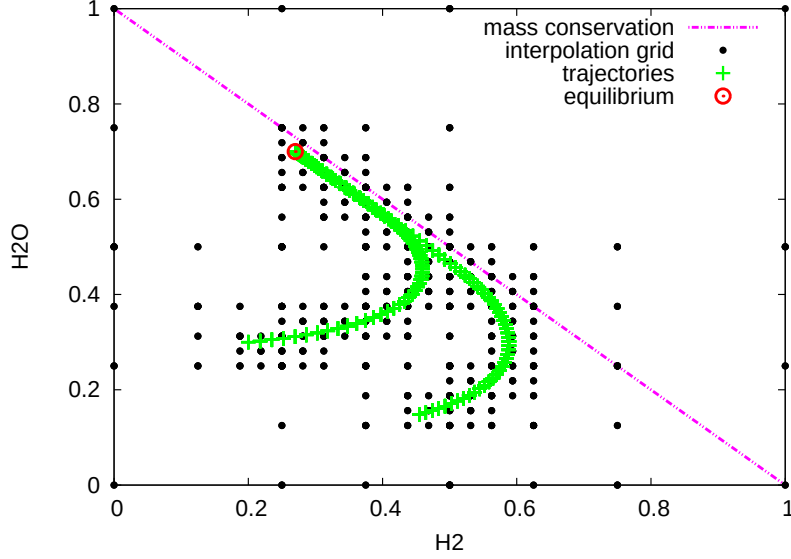


Figure 4: Interpolation grid used to store the evaluation of the manifold.

approaches could lead to higher time steps for the implicit method. The stability of the explicit and implicit methods applied to the reduced system turns out to be quite similar.

As expected, the reduced system allows the use of larger time steps and even explicit methods can successfully be used to integrate the system. The maximal time steps for the CPA approach are very similar to the first approximation but the CPA produces slightly better results. However, the CPA approach is much more expensive because of the formation of  $N(r)$ , i.e. the computation of the derivatives of  $z^{\mathcal{M}}$  with respect to  $r$ . To make things worse, unlike in the case of the first approximation, the transport coefficients have to be calculated for *all* species (represented and unrepresented) and finally a linear system, cf. (5.15), has to be solved. Therefore, one should carefully weigh up the necessity of applying more elaborate (and possibly more accurate) methods in the light of computational performance.

We used two sets of initial values (IVs) which are in the “vicinity” of the chemical equilibrium (7.4). The left column of Fig. 5 shows results for IV

$$u_0 = (0.455, 0.779, 0.237, 0.363, 0.148, 0.015)^T. \quad (7.5)$$

In the right column, results are depicted for IV

$$u_1 = (0.2, 0.95, 0.31, 0.03, 0.3, 0.05)^T. \quad (7.6)$$

In the next subsections, we endow mechanism 3 with transport coefficients. Values needed for the computation of these coefficients can be obtained upon request, if the reader wishes to compare our results with his own.

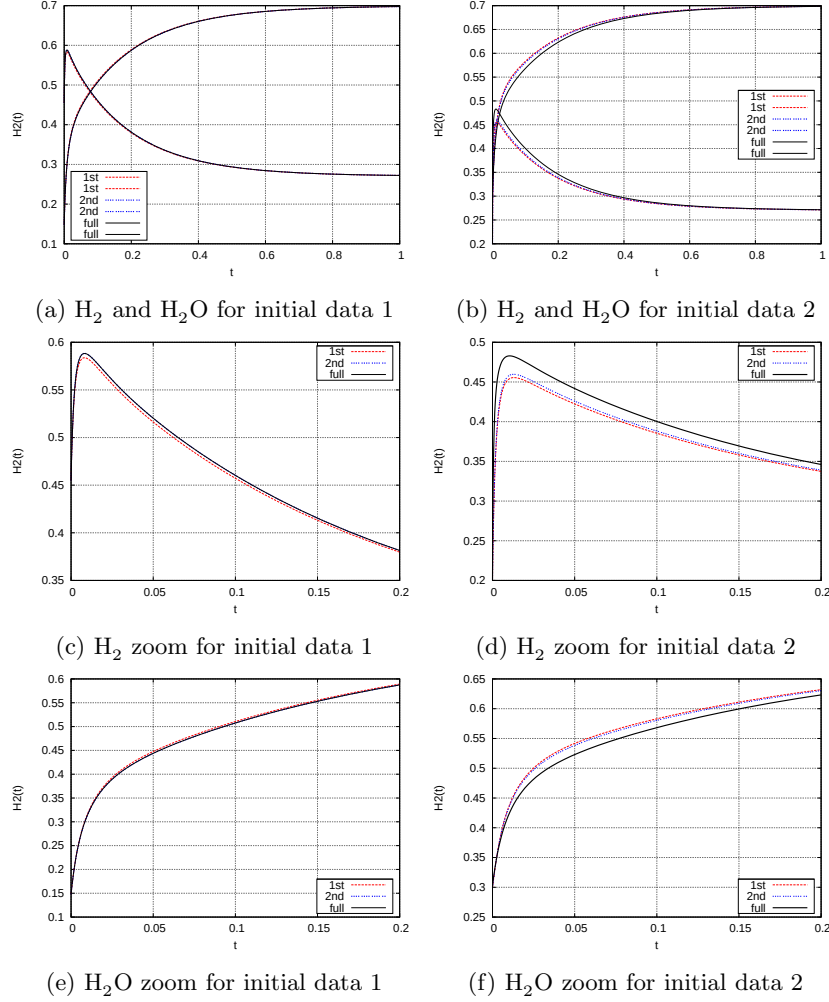


Figure 5: Results for ODE test for data set 1 and 2.

### 7.3. Hydrogen combustion with diffusion in 1d

We study the convection-free ( $v = 0$ ) transport problem described in section 2, in an isobaric ( $p = \text{const.}$ ) and isothermal ( $T = \text{const.} = 1000\text{K}$ ) setting defined on  $\Omega_{t_{\text{end}}} = (0, 0.01) \times (0, t_{\text{end}})$ , where  $t_{\text{end}} = 0.5$ . The governing equations read as

$$\partial_t(\rho Y_k) - \partial_x \left( \rho \frac{W_k}{\overline{W}} \mathcal{D}_k^{\text{mix}} \partial_x X_k \right) - [\dot{X}_k] W_k = 0, \quad k = 1, \dots, n_{\text{spec}}, \quad (7.7)$$

where  $[\dot{X}_k]$  is given by Eq. (2.11). By virtue of (2.7), the governing equations can also be stated in concentrations by simply dividing (7.7) by  $W_k$ .

As initial condition, we take

$$u(x, t = 0) = \begin{cases} u_0 & \text{if } 0.002 \leq x \leq 0.008 \\ u_1 & \text{otherwise,} \end{cases}$$

where  $u_0$  is given in (7.5) and  $u_1$  is given in (7.6). For the sake of simplicity we impose periodic boundary conditions, i.e.

$$u(0, t) = u(0.01, t).$$

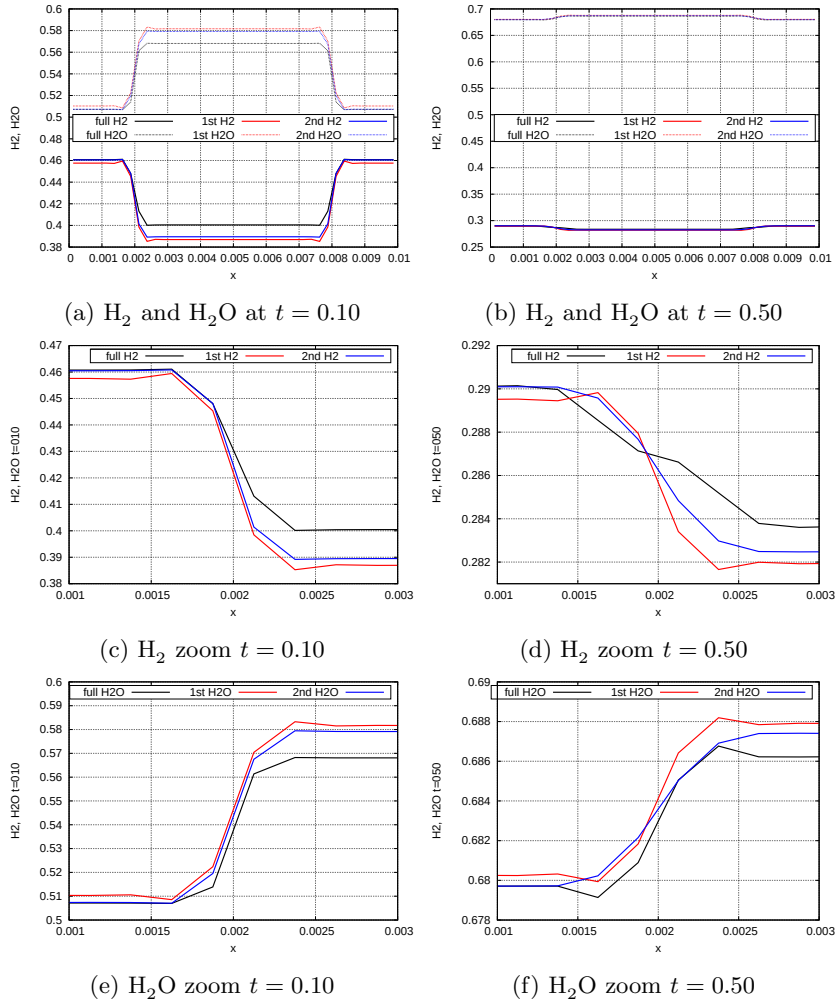


Figure 6: Results for ODE test for data set 1 and 2.

As can be seen in Fig. 6, diffusive and computational invariance effects can be extenuated by using the CPA approach. However, the improvements relative to the result obtained by solving the full set of differential equations is moderate. At the same time, the computational costs increase tremendously. Nevertheless, the CPA remains a viable method for improving the accuracy of our model reduction method.

#### 7.4. Hydrogen combustion with diffusion in 2d

In this section we consider the full transport problem described in section 2 in an isobaric ( $p = \text{const.}$ ) and isothermal ( $T = \text{const.} = 1000\text{K}$ ) scenario with diffusion defined on  $\Omega_{t_{\text{end}}} = (0, 0.01)^2 \times (0, t_{\text{end}})$ , where  $t_{\text{end}} = 0.5$ . The governing equations are the ones from the previous section, this time in 2d and with a convection term. They read as

$$\partial_t(\rho Y_k) + \nabla \cdot \left( \rho Y_k v - \rho \frac{W_k}{\overline{W}} \mathcal{D}_k^{\text{mix}} \nabla X_k \right) - [\dot{X}_k] W_k = 0, \quad k = 1, \dots, n_{\text{spec}}, \quad (7.8)$$

where  $[\dot{X}_k]$  is given by Eq. (2.11). By virtue of (2.7), the governing equations can also be stated in concentrations by simply dividing (7.8) by  $W_k$ . We choose a rotational velocity field given by

$$v(x, y) = (-2\pi(y - 0.005), 2\pi(x - 0.005))^T.$$

The initial conditions are shown in Fig. 7. Again, for the sake of simplicity we impose periodic boundary conditions, i.e.

$$u(0, y, t) = u(0.01, y, t) \text{ and } u(x, 0, t) = u(x, 0.01, t).$$

As with one spatial dimension, the CPA approach works well in the 2d case, compare Figs. 8 and 9 for simulations of the RPVs  $\text{H}_2$  and  $\text{H}_2\text{O}$ , respectively. To get a better impression of the quality of the particular approximations with respect to the full solution, Fig. 10 shows cross-sectional plots for both  $\text{H}_2$  and  $\text{H}_2\text{O}$ . The CPA (2nd) approximation yields again slightly better results than the 1st, but is computationally much more expensive.

In this paper we only highlight the quality of the approximations. A full efficiency study will be carried out in a separate paper together with the coupling to the fully compressible Navier-Stokes equations.



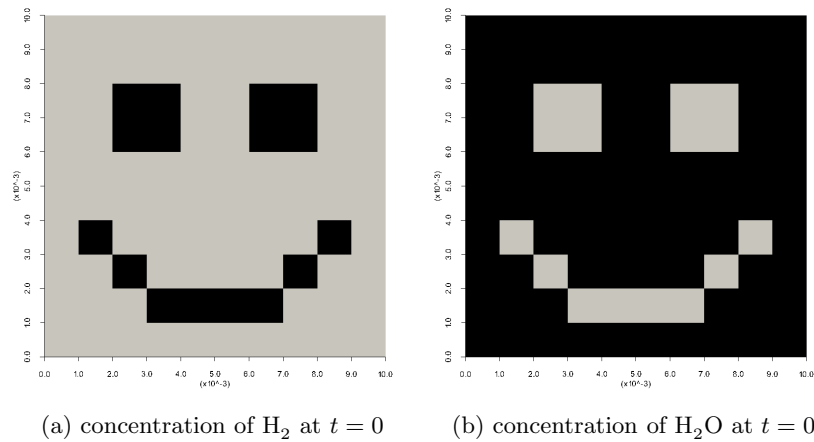


Figure 7: Initial data for the 2d simulation. The dark color corresponds to the concentration of  $u_{H_2}(\cdot, 0) = 0.2$  and  $u_{H_2O}(\cdot, 0) = 0.148$  whereas the bright colors correspond to the concentrations  $u_{H_2}(\cdot, 0) = 0.455$  and  $u_{H_2O}(\cdot, 0) = 0.3$ .

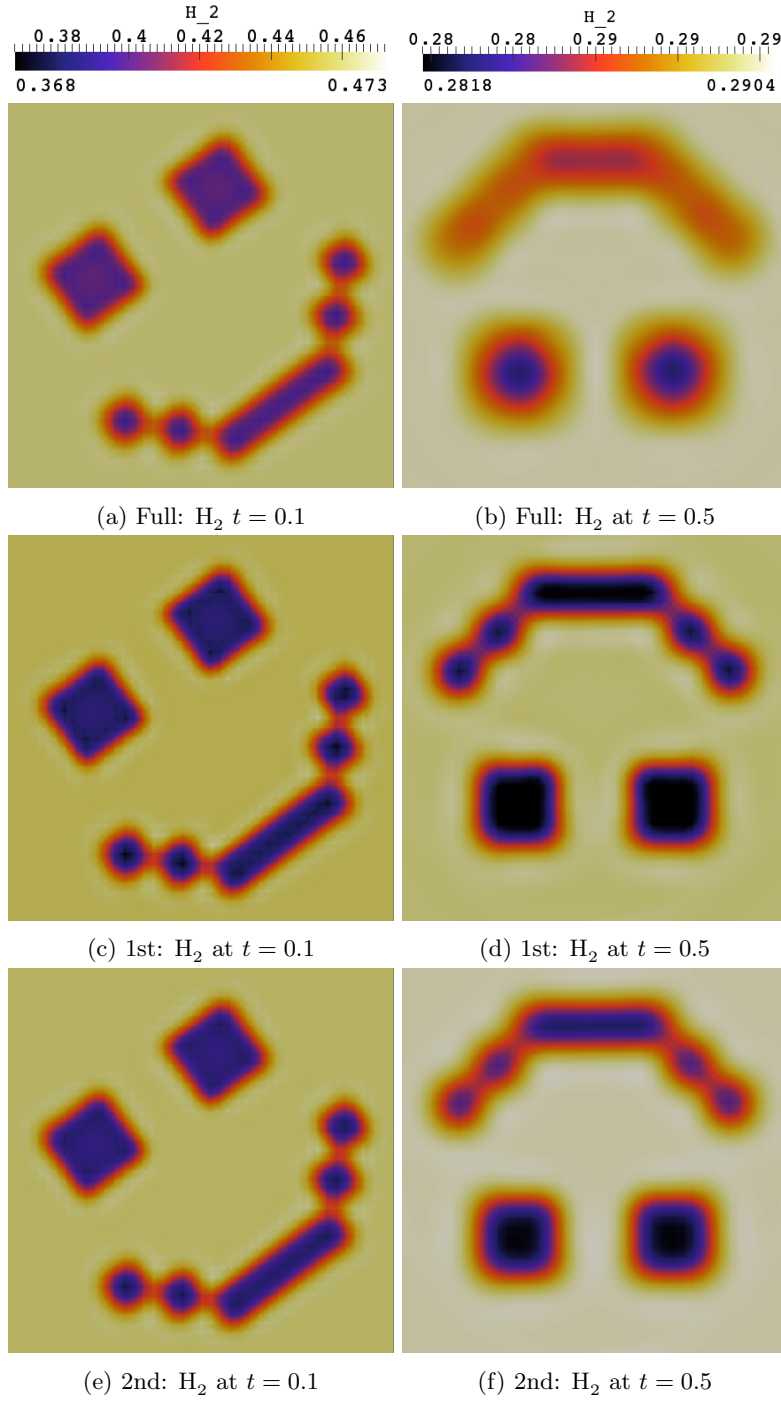


Figure 8: Simulations of  $H_2$ . From top to down: full, first and second approximation. Left column: Snapshot at  $t = 0.1$ . Right column: Snapshot at  $t = 0.5$ .

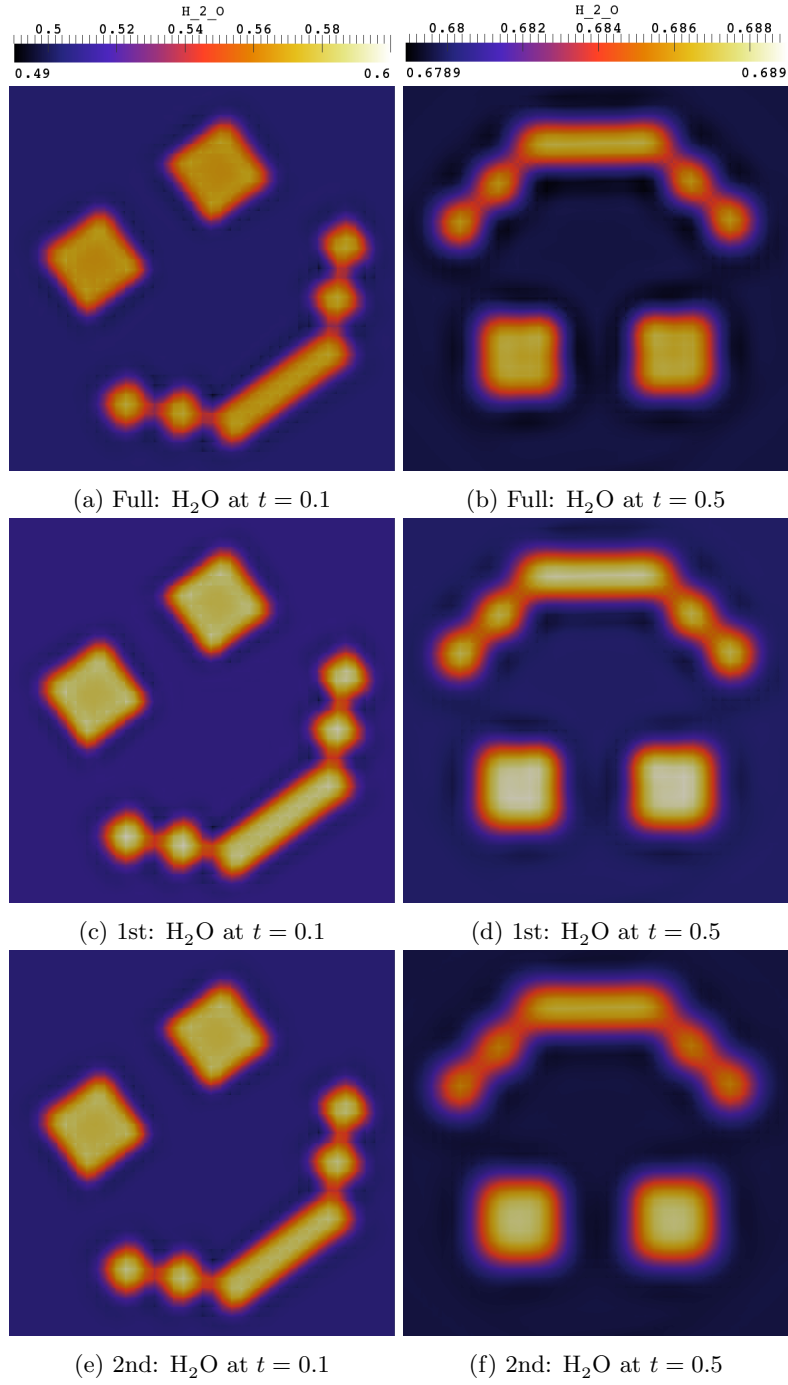
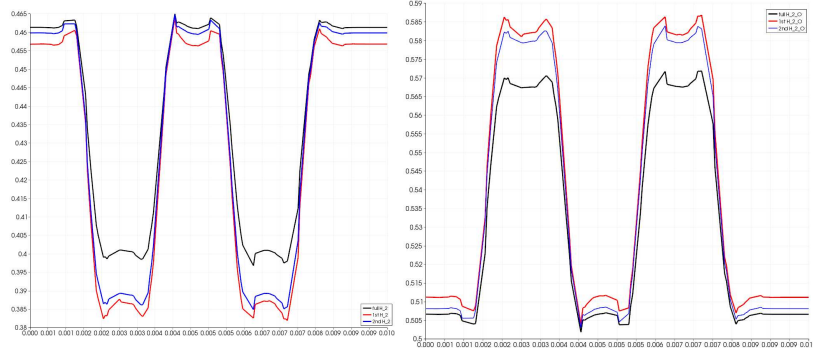
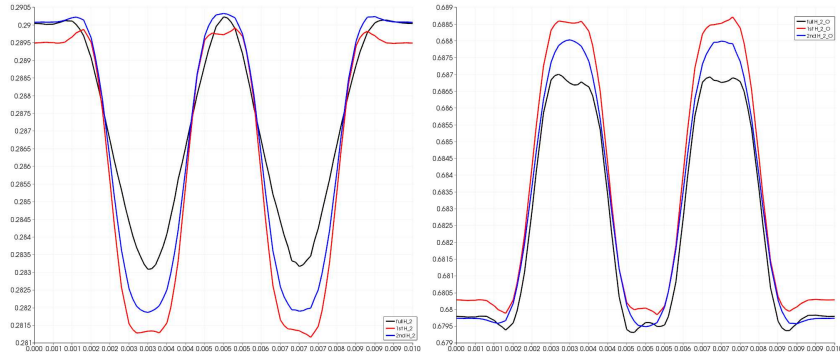


Figure 9: Simulations of  $\text{H}_2\text{O}$ . From top to down: full, first and second approximation. Left column: Snapshot at  $t = 0.1$  Right column: Snapshot at  $t = 0.5$ .



(a) Plot over the line connecting the points  $(0, 0.004)$  and  $(0.008, 0.01)$  for  $H_2$  at  $t = 0.1$

(b) Plot over the line connecting the points  $(0, 0.004)$  and  $(0.008, 0.01)$  for  $H_2O$  at  $t = 0.1$



(c) Plot over the line  $[0, 0.01] \times \{0.003\}$  for  $H_2$  at  $t = 0.5$

(d) Plot over the line  $[0, 0.01] \times \{0.003\}$  for  $H_2O$  at  $t = 0.5$

Figure 10: Cross-sectional results to show the quality of the particular approximation approaches w.r.t. the full model at two distinct time points  $t = 0.1$  and  $t = 0.5$ . Left column:  $H_2$ . Right column:  $H_2O$

## 8. Discussion and conclusion

In this treatise, we make the following contributions:

We present a novel *weak formulation for discontinuous Galerkin methods* to cope with *reduced problems*, especially the CPA formulation. To our knowledge, this is the first time that the CPA method is applied in conjunction with higher dimensional SIMs for nontrivial mechanisms in several space dimensions.

Moreover, we introduce a new method for the *adaptive tabulation* of manifold points.

We also present the *application* of a relative novel method for chemistry-based model reduction based on nonlinear optimization to a 2D chemically reacting flow scenario and demonstrate its limitations. This optimization approach can be seen as an adaption of the methods proposed in [11, 10]. It is the very first time that the MR approach (4.3) has been applied to nontrivial PDE systems. Our implementation of the method requires relatively little knowledge of chemical issues and is intended to provide a rather automatized way of dealing with dimension reductions of chemically reaction flows: the user has only to select those variables out of the chemical species vector that are to serve as reaction progress variables, the method of approximation (none, 1st or 2nd) as well as thermochemical and transport data.

Besides the simplicity of the nonlinear programming approach, this method still exhibits a few drawbacks:

First of all, problem (4.3) is ill-posed in the sense that the objective functional (4.3a) is usually non-convex. This means, that any minimal point of (4.3) is not unique. Hence it is difficult to judge whether, amongst other things, a local solution depicts an actual SIM point and how, in dependence on initial values, “better” solutions can be determined. A possible convexification could circumvent this shortcoming. Moreover, up to now it seems uncertain how to devise a reasonable adaptive strategy in order to control errors introduced by a dimension reduction.

A second issue concerns computational resources. In this paper we were interested in a qualitative comparison of the different methods (full, 1st, and 2nd) only and thus we have not spent much efforts to make the reduced approaches very efficient. Indeed, the current implementation demands slightly more computational resources in comparison to the full model, even with the adaptive tabulation approach used. For the simple hydrogen combustion, the 1st and 2nd approximations are still around 1/5 and 1/3, respectively, slower than the simulation of the full system. Future work on this topic will concentrate on optimizing the implementation from a software technical point of view. Owing to the small dimension of the combustion mechanism, a speed-up should most likely occur for (much) more complex chemical compositions, i.e. much more chemical species and possible 3d scenarios. Future research will treat these issues.

Moreover, for the sake of simplicity, we dropped any dependence on temperature. Including thermal energy definitely blows up the optimization problem, thus resulting in additional computational expense.

Further investigations are supposed to deal with non-isothermal settings in order to incorporate thermal energy as well as momentum, thus describing a full set of Navier-Stokes equations. This will be laid out in a future paper and we expect our approach to get a viable alternative to existing model reduction methods.

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