Sensitivity of Reaction-Diffusion Manifolds (REDIMs) with respect to gradient estimates and boundary conditions

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1 Introduction

Combustion processes are still important in many engineering applications [1, 2]. In studies of reacting flows (and combustion in particular) mathematical modeling and simulation have become very valuable tools to study and to predict the behavior of combustion systems [3]. This, however, leads to computationally demanding if not prohibitive use of detailed chemical kinetics for the simulation general three-dimensional turbulent reacting flows [4, 5]. Model reduction for chemical kinetics based on the concept of low-dimensional manifolds and subsequent tabulation has become a pivotal tool to overcome and to cope with the problem of both high dimensionality and complexity, i.e., non-literality of mathematical models of mechanisms of chemical kinetics [4]. At present, many different methods have been developed aiming at construction and usage of these low-dimensional manifolds e.g., the Intrinsic Low-Dimensional Manifolds (ILDM) [6], the Flamelet Prolongation of ILDM (FPI) [7, 8], the flamelet model [9], the Reaction-Diffusion Manifolds (REDIM) [10] and Flamelet Generated Manifolds (FGMs) [11]. These methods, as well as many others, see e.g. [4] for reviews on model reduction for mathematical models of chemical kinetics in combustion, based on manifolds typically differ in the way manifolds are constructed and implemented. In this work, the Reaction-Diffusion Manifolds (REDIMs) method [10] is considered. The methodology has has been constantly improved and extended in the last decade see e.g., [12, 13]. The key element of this method concerns a gradient estimate, which describes the influence of the diffusion / molecular transport see e.g., [10, 14, 15] for empirical investigations on the dependence of the manifolds on the gradient estimates.

There is also a number of numerical investigations of this influence [12, 16]. It is preferable to develop a generic approach to identify and to quantify the influence / importance of the system gradient estimates as well as other parameters on the manifold based reduced chemistry. Hence the equation of the sensitivity of the low-dimensional manifolds on the system gradient estimate, the boundary condition and elementary reaction rates are studied, derived and implemented here. The focus, however, is made on the boundary conditions because these have significant influence on the diffusion term computed on the manifold. The sensitivities of the slow manifolds with respect to the perturbations have been already treated. For instance, in [17] a sensitivity analysis of the ILDM manifolds [6] with respect to the reaction rate coefficients was formulated. Now, an additional effort has been made to include the influence of the boundary conditions. The method is illustrated and verified by application to diffusion flames of diluted hydrogen and air combustion system.

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2 Mathematical model

A reaction/convection/diffusion system in the vector form is considered in the following. The detailed chemical source term and the matrix of transport coefficients are described by functions of the thermo-kinetic state only. The thermo-kinetic state vector $\boldsymbol{\psi}$ is an $(n = n_s+2)$-dimensional vector is introduced via e.g. the specific enthalpy h, the pressure p and the specific mole numbers ϕ_i ($\phi_i = w_i/M_i$, where w_i are the mass fractions and M_i the molar masses of the n_s chemical species - $\boldsymbol{\psi} = (h, p, \phi_1, \ldots, \phi_{n_s})^T$. In a general reacting flow this thermo-kinetic state is given as a function of both time and space: $\boldsymbol{\psi} = \boldsymbol{\psi}(t, \vec{r})$, with t the time and $\vec{r}$ the vector of spatial coordinates. As mentioned above to simplify notations the evolution equation for the state space vector is expressed as

$$\rho\frac{\partial\boldsymbol{\psi}}{\partial t} = G(\boldsymbol{\psi}) - \rho\vec{v}\,\mathrm{grad}\boldsymbol{\psi} + \mathrm{div}\,(D\,\mathrm{grad}\boldsymbol{\psi})\,, \qquad \boldsymbol{\psi} = \boldsymbol{\psi}(\vec{r}, t), \qquad \vec{r} \in \Omega, \qquad t \in [0, \infty]\,, \quad (1)$$

where $\vec{v}$ denotes the flow velocity vector, $\boldsymbol{G}(\boldsymbol{\psi})$ the n-dimensional vector of chemical source terms, D the $n \times n$-dimensional matrix of detailed transport coefficients (including diffusion, heat conduction, thermal diffusion, etc.). The equation system is closed by specifying boundary condition on the boundary $\partial\Omega$ of Ω.

2.1 Low-dimensional manifolds and Sensitivity

Following the main assumption, the accessed thermo-kinetic state space by the system solutions for both laminar and turbulent reacting flows remains "close" to low-dimensional manifolds [6, 18]. The assumption restricts the evolution of the state space vector within this manifold

$$\mathcal{M} = \left\{\boldsymbol{\psi} = \boldsymbol{\psi}(\boldsymbol{\theta}(\vec{r}, t)),\ \ \mathbb{R}^m \to \mathbb{R}^{n_s+2}\right\},$$

with $\boldsymbol{\theta}$ as the m-dimensional reduced coordinate vector. In the reacting flow calculation using manifold methods, the solution of Eq. (1) is replaced by the solution of an evolution equation for $\boldsymbol{\theta}(\vec{r}, t)$, which is obtained by projecting the governing equation system onto the manifold. For this the tangent space of the manifold can be used and described in matrix notation by $\boldsymbol{\psi_\theta}$, that denotes the $(n \times m)$-dimensional matrix of partial derivatives of $\boldsymbol{\psi}$ with respect to θ as $(\boldsymbol{\psi_\theta})_{ij} = \partial\psi_i/\partial\theta_j$.

2.2 Sensitivity of REDIMs

In the REDIM method [10] an evolution equation is solved in order to identify the low-dimensional manifolds (see e.g., [19] for more details). Below important notations and definitions are presented. The REDIM evolution equation towards an invariant manifold can be cast as

$$\rho\frac{\partial\boldsymbol{\psi}}{\partial t} = \mathcal{P}\ \left(G\left(\boldsymbol{\psi}\right) + \Xi(\boldsymbol{\psi}, \boldsymbol{\psi_\theta}, \boldsymbol{\psi_{\theta\theta}})\right), \quad (2)$$

where for the case of detailed transport

$$\Xi(\boldsymbol{\psi}, \boldsymbol{\psi_\theta}, \boldsymbol{\psi_{\theta\theta}}) = (D\boldsymbol{\psi_\theta}\chi)_{\boldsymbol{\theta}}\,\chi.$$

Once a simplified transport model is used with $D = dI$ and I being the identity matrix, the term Ξ can be simplified to $\Xi(\boldsymbol{\psi}, \boldsymbol{\psi_\theta}, \boldsymbol{\psi_{\theta\theta}}) = d(\boldsymbol{\psi}_{\theta\theta}\chi)\chi$. Here $\boldsymbol{\chi} = \nabla\boldsymbol{\theta}$ as the gradient estimate. The equation (2) is integrated for $t \to \infty$ with specified initial $\boldsymbol{\psi}(\theta) = \boldsymbol{\psi}_0(\theta)$ - and boundary conditions, for instance, in the case of Dirichlet boundary conditions become $\boldsymbol{\psi}|_{\partial\mathcal{M}} = \boldsymbol{\psi}_0(\theta)|_{\partial\mathcal{M}}$. The steady solution of Eq. (3) yields the REDIM.

The vector of sensitivities $\boldsymbol{s} = (s_1, s_2, \ldots, s_n)^T$ with respect to a parameter ($\boldsymbol{s} = \boldsymbol{\psi}_p$) or the matrix of sensitivities with respect to a parameter vector $\boldsymbol{p}$ defines how the manifold might change with changing parameters. Note that the definition of sensitivity - $\boldsymbol{s}$ in the context of manifolds based model reduction should account for a change of the manifold itself and not any change of $\boldsymbol{\psi}$ tangential to the manifold. Therefore, quite natural solution is to retain and to account for the components of the sensitivities perpendicular to the manifold leading to the condition $\boldsymbol{\psi_\theta s} = 0 : \boldsymbol{s} = \boldsymbol{\psi_{\theta\perp}\sigma}$. An alternative is to define the sensitivities by keeping several variables fixed (e.g. some major species) and allowing only the others to change. This choice yields the condition $C^T\boldsymbol{s} = 0 : \boldsymbol{s} = C_\perp\boldsymbol{\sigma}$. That is why it is advantageous to base the formulation of the sensitivity equation on the original invariance equation for the steady state at $t \to \infty$ (see e.g., [10, 19]), which is given by

$$Z\left[G\left(\boldsymbol{\psi}\right) + \Xi(\boldsymbol{\psi}, \boldsymbol{\psi_\theta}, \boldsymbol{\psi_{\theta\theta}})\right] = 0, \tag{3}$$

where $Z = S\boldsymbol{\psi}_{\theta\perp}^T$, $\boldsymbol{\psi}_{\theta\perp}$ is the orthogonal complement of $\boldsymbol{\psi_\theta}$ and S is an arbitrary $(n - m \times n - m)$-dimensional (regular) scaling matrix (see, e.g. [20] for suitable choices).

2.3 Sensitivity equations

The invariance equation (3) with simple calculus and definition of the sensitivity vector yield the equation for the sensitivity $\boldsymbol{s} = \frac{\partial\boldsymbol{\psi}}{\partial p}$ as

$$\frac{\mathrm{d}Z}{\mathrm{d}p}\left[G + \Xi\right] + Z\Big[\frac{\mathrm{d}G}{\mathrm{d}p} + \frac{\mathrm{d}\Xi}{\mathrm{d}p}\Big] = 0. \tag{4}$$

All terms in Eq. (4) are introduced and discussed in [19], therefore, major conclusions drawn and the main results only shall be presented here. Hence, the final and general system of equations for the sensitivity vector reads

$$\begin{aligned} 0 = &- Z\boldsymbol{s_\theta}(C^T\boldsymbol{\psi_\theta})^{-1}C^T\left[G + (D\boldsymbol{\psi_\theta}\chi)_{\boldsymbol{\theta}}\,\chi\right] \\ &+ Z\left[G_{\boldsymbol{\psi}}\boldsymbol{s} + G_p + ((D_{\boldsymbol{\psi}}\boldsymbol{s})\boldsymbol{\psi_\theta}\chi)_{\boldsymbol{\theta}}\,\chi + (D\boldsymbol{s_\theta}\chi)_{\boldsymbol{\theta}}\,\chi + (D\boldsymbol{\psi_\theta}\chi)_{\boldsymbol{\theta}}\,\chi_p + (D\boldsymbol{\psi_\theta}\chi_p)_{\boldsymbol{\theta}}\,\chi\right], \end{aligned} \tag{5}$$

where the sensitivities are defined as changes of the vector $\boldsymbol{\psi}$ only perpendicular to the vectors in C. As an example assume that for a one-dimensional system C contains only one entry at the position of the variable H_2O. Then the sensitivity would give the net change of $\boldsymbol{\psi}$ resulting if H_2O is not allowed to change (see [19]).

Although the general sensitivity equation (5) can be integrated numerically, it is useful to restrict to the following simplified form. The following simplifying assumptions (1) $D = dI$, (2) d depends only weakly on the state vector ($d_{\boldsymbol{\psi}}s \approx 0$) and (3) C is a constant matrix introduced above are additionally applied.

The sensitivity equation covers three different sensitivities, namely

- with respect to gradient estimates: $G_p = 0$,
- with respect to chemical kinetics $\boldsymbol{\chi}_p = 0$,
- with respect to boundary conditions $\boldsymbol{\chi}_p = 0$, $G_p = 0$, with boundary values $\boldsymbol{s}(\theta)|_{\partial\mathcal{M}}$ for the sensitivities $\boldsymbol{s}$ specified.

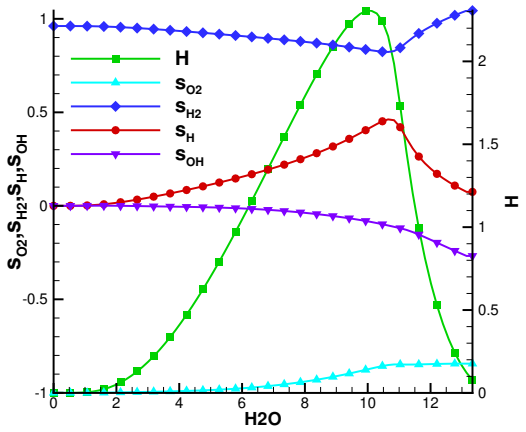

Figure 1: Sensitivities for a 1D REDIM for given value of $s_{\mathrm{H}_2} = 1$, $s_{\mathrm{O}_2} = -1$ at the boundary, all other sensitivities are set to 0.

As an example for the sensitivities with respect to the boundary conditions assume that the sensitivity with respect to one species mole fraction is of interest. Then all entries in the sensitivity vectors $\boldsymbol{s}$ are zero, except for that corresponding to the chosen species, which is 1. Note that by specifying other boundary sensitivities one could test how the manifold changes if, say, hydrogen is replaced by oxygen. The cases of the gradients estimates and chemical kinetics parameters were presented e.g. in [19]. In this study the question of sensitivity of the REDIM with respect to boundary conditions is addressed.

3 Results

Here we show examples for both a 1D and a 2D manifold. In the case of 1D the chosen example is a premixed flat stoichiometric hydrogen/air flame at 1 bar and and unburnt temperature of 298 K. Figures 1 shows an example in composition space for the sensitivity based on a sensitivity at the left boundary of $s_{\mathrm{H}_2} = 1$, $s_{\mathrm{O}_2} = -1$ at the boundary, all other sensitivities at the left boundary are set to 0. At the right boundary, Neumann conditions are specified. Note that this describes the sensitivity based on a replacement of oxygen by hydrogen at the boundary. The sensitivity for this replacement of oxygen by hydrogen is shown, and, as can be expected, such a change of the boundary conditions changes the manifold considerable (mixture composition is changed).

As a 2D example 2D REDIMs equations are integrated and constructed for a counter-flow diffusion flame configuration of a $50 : 50$ mixture of $\mathrm{H}_2 : \mathrm{N}_2$ and air at a standard ambient pressure and temperature ($T_0 = 298$ K and $p_0 = 1$ bar). Figures 2 show 3D projections of the sensitivity of the 2D REDIM with respect to the boundary conditions specified for OH radical: $s_{\mathrm{OH}} = 1$. This is shown by the mesh. On the left figure one can see the sensitivity of the OH radical, which has been specified for the analysis. One notices that it equals to one on the boundary and then vanishes rapidly towards the interior of the REDIM. This means the 2D manifold with respect to same OH radical is only sensitive near the boundary as expected. In the right figure the sensitivity of the H radical to the boundary values of OH shows also to be sensitive only in the vicinity of the boundary of the manifold.

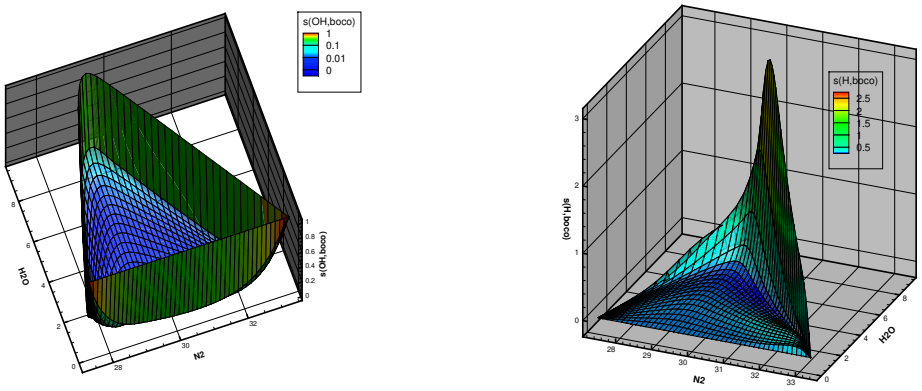

Figure 2: Sensitivities of OH (left) and H (right) radicals with respect to the value of OH at the boundary ($s_{\mathrm{OH}} = 1$ all other sensitivities are 0) computed for the 2D REDIM shown by the mesh with contours as 2D function of H_2O and N_2 specific mole numbers.

4 Conclusions

The sensitivity analysis for REDIMs with respect to gradient estimates, elementary reaction rate coefficients and boundary conditions was briefly introduced and outlined. The focus was made on the sensitivities with respect to boundary conditions. The method to compute the sensitivities was implemented in the REDIM evolution equation and integrated in a coupled way such that the converged REDIM solution provides both the manifold and the sensitivity at once. The method was illustrated by application to 1D and 2D REDIMs constructed for diluted hydrogen and air counter-flow diffusion flames. Several important conclusion were drawn

- the sensitivity of the slow manifold can be computed in a very generic manner providing information on the manifold sensitivity to important system and manifold parameters;
- the sensitivity obtained can be used to assist the manifold construction process improving boundary conditions. These can be studied and used to improve the convergence and stability of the REDIM manifold while integration.

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