Sensitivity of Reaction-Diffusion Manifolds (REDIM) for hydrogen counter-diffusion flames

Ulrich Maas, Viatcheslav Bykov
Karlsruhe Institute of Technology
Karlsruhe, Germany

1 Introduction

Model reduction for chemical kinetics based on the concept of low-dimensional manifolds represents a very attractive methodology to cope with the problem of high dimensionality in a very general manner [1]. Many different method exist e.g., the Intrinsic Low-Dimensional Manifolds (ILDM) [2], the Flamelet Prolongation of ILDM (FPI) [3, 4], the flamelet model [5], the Reaction-Diffusion Manifolds (REDIM) [6] and the Flamelet Generated Manifold (FGM) [7]. Although these methods are based on low-dimensional manifolds they differ in the way manifolds are identified and implemented.

In this work, we focus on the Reaction-Diffusion Manifolds (REDIMs) method [6]. In the past, much effort has been made to validate, improve and extend the methodology (see e.g. [8–11]). One of the crucial issues with the implementation concerns a gradient estimate, which is required in the generation of the REDIM [6]. These estimates are very important because they describe the influence of the diffusion / transport and related to the strength of the diffusion processes in particular system considered. Some empirical investigations on the dependence of the manifolds on the gradient estimate can be found e.g., in [6, 8, 12]. It has been shown numerically that the gradient estimate becomes less and less important with increasing dimension of the REDIM reduced chemistry. Despite of these numerical studies one would still like to have a general way to identify and quantify the influence / importance of the system gradient estimates for the model reduction on the manifold based reduced chemistry (e.g. REDIM and flamelet based model such as FGM [13]). Additionally, the question of sensitivity of the slow manifold with respect to elementary reactions can be crucial for further mechanism development. Knowledge about reactions that describe and influence the slow manifold can be used in mechanism improvement and it can also contribute to a better understanding of chemical kinetics.

To answer these questions, the equations for the sensitivity of the low-dimensional manifolds on the system gradient estimate and elementary reaction rates are derived and implemented. Note that the sensitivities of the slow manifolds with respect to the perturbations have been investigated before, for instance in [14], where a sensitivity analysis of the slow manifolds with respect to the reaction rate coefficients was formulated in the context of the Intrinsic Low-Dimensional Manifolds (ILDM) [2]. Now, it is also modified and implemented in the REDIM approach. Additionally, the influence of the system gradients on the simplified chemistry is studied by the sensitivity to the system gradient estimates. It is illustrated and verified for a diluted hydrogen / air diffusion flame.

Correspondence to: viatcheslav.bykov@kit.edu

2 Mathematical model

A general reaction/convection/transport system will be considered. The chemical source term and the matrix of transport coefficients are only functions of the thermo-kinetic state vector. The thermo-kinetic state vector $\boldsymbol{\psi}$ is an $(n = n_s + 2)$-dimensional vector represented by 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.

$$\boldsymbol{\psi} = (h, p, \phi_1, \ldots, \phi_{n_s})^T \,, \tag{1}$$

In a reacting flow this thermokinetic state is 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. To simplify notation we write the evolution equation in vector form according to

$$\rho \frac{\partial \boldsymbol{\psi}}{\partial t} = G(\boldsymbol{\psi}) - \rho v \,\mathrm{grad}\boldsymbol{\psi} + \mathrm{div}\,(D \,\mathrm{grad}\boldsymbol{\psi}) \,, \qquad \boldsymbol{\psi} = \boldsymbol{\psi}(\vec{r}, t), \qquad \vec{r} \in \Omega, \qquad t \in [0, \infty] \,, \tag{2}$$

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

The accessed thermo-kinetic state space for both laminar and turbulent reacting flows is typically close to low-dimensional manifolds [2, 15]. This can be expressed as:

$$\mathcal{M} = \left\{ \boldsymbol{\psi} = \boldsymbol{\psi}(\boldsymbol{\theta}(\vec{r}, t)), \;\; \mathbb{R}^m \to \mathbb{R}^{n_s+2} \right\} , \tag{3}$$

with $\boldsymbol{\theta}$ as the m-dimensional reduced coordinate vector. In the reacting flow calculation using manifold methods, the solution of Eq. (2) 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. The tangent space of the manifold is given in matrix notation by $\boldsymbol{\psi_\theta}$ and denotes the $(n \times m)$-dimensional matrix of partial derivaties of $\boldsymbol{\psi}$ with respect to θ as $(\boldsymbol{\psi_\theta})_{ij} = \partial\psi_i/\partial\theta_j$. The vector of sensitivities $s = (s_1, s_2, \ldots, s_n)^T$ 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 changes when the parameters are changed.

2.2 Sensitivity of REDIMs

In the REDIM method [6] an evolution equation is solved in order to identify the low-dimensional manifolds (see e.g., [16] for more details and notation)

$$\rho \frac{\partial \boldsymbol{\psi}}{\partial t} = \mathcal{P} \;\left(G\left(\boldsymbol{\psi}\right) + \Xi(\boldsymbol{\psi}, \boldsymbol{\psi_\theta}, \boldsymbol{\psi_{\theta\theta}}) \right) , \tag{4}$$

where

- For the case of detailed transport,

$$\Xi(\boldsymbol{\psi}, \boldsymbol{\psi_\theta}, \boldsymbol{\psi_{\theta\theta}}) = \left(D\boldsymbol{\psi_\theta}\chi\right)_{\boldsymbol{\theta}} \chi, \tag{5}$$

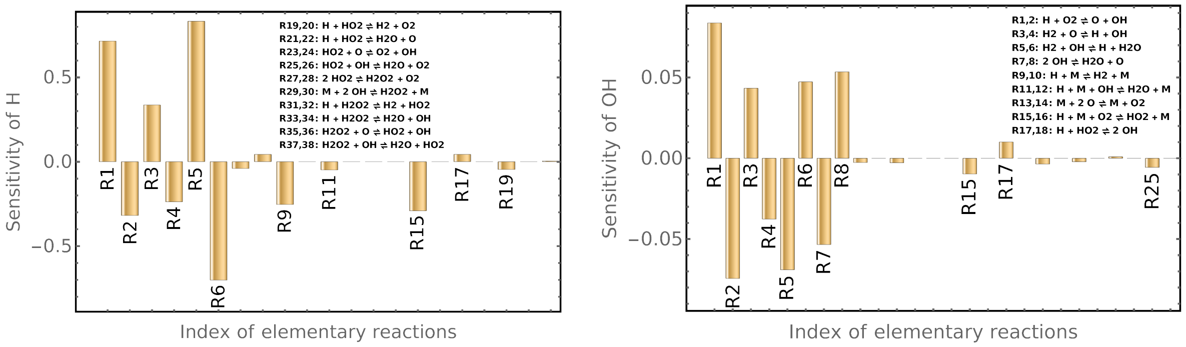

Figure 1: Maximal values (in magnitude) of sensitivity coefficients of H (left) and OH (right) radicals of the 2D REDIM with respect to the elementary reaction rates. The 10 most sensitive reactions are shown for each radical. List of all reactions is provided in the legends.

- For a simplified transport with $D = dI$ and I being the identity matrix, the term $\Xi(\boldsymbol{\psi}, \boldsymbol{\psi_\theta}, \boldsymbol{\psi_{\theta\theta}}) = d(\boldsymbol{\psi}_\theta \chi)_\theta \chi$ can be reduced to $\Xi(\boldsymbol{\psi}, \boldsymbol{\psi_\theta}, \boldsymbol{\psi_{\theta\theta}}) = d(\boldsymbol{\psi}_{\theta\theta} \chi)\chi$,

with $\boldsymbol{\chi} = \nabla \boldsymbol{\theta}$ as the gradient estimate. This equation is integrated for $t \to \infty$ and the steady solution yields the REDIM. However, for the formulation of the sensitivity equation it is advantageous to base it on the original invariance equation for the steady state at $t \to \infty$ (see e.g., [6, 16]), which is given by

$$Z\left[G\left(\boldsymbol{\psi}\right) + \Xi(\boldsymbol{\psi}, \boldsymbol{\psi_\theta}, \boldsymbol{\psi_{\theta\theta}})\right] = 0, \tag{6}$$

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. [17] for suitable choices).

2.3 Sensitivity equations

Starting from the invariance equation (6) simple calculus leads to the equation for the sensitivity $s = \frac{\partial \boldsymbol{\psi}}{\partial p}$

$$\frac{\mathrm{d}Z}{\mathrm{d}p}\left[G + \Xi\right] + Z\left[\frac{\mathrm{d}G}{\mathrm{d}p} + \frac{\mathrm{d}\Xi}{\mathrm{d}p}\right] = 0. \tag{7}$$

Most terms in Eq. (7) have been discussed in [16], therefore, we shall only summarize the results, and we shall adopt a general notation for matrix $C_\perp$ noting that it can also be given as $C_\perp = \boldsymbol{\psi_{\theta\perp}}$. The final general equations system for the sensitivity then reads

$$\begin{aligned} 0 = & - Z s_{\boldsymbol{\theta}} (C^T \boldsymbol{\psi_\theta})^{-1} C^T \left[G + (D\boldsymbol{\psi_\theta}\chi)_{\boldsymbol{\theta}}\, \chi\right] \\ & + Z\left[G_{\boldsymbol{\psi}} s + G_p + ((D_{\boldsymbol{\psi}} s)\boldsymbol{\psi_\theta}\chi)_{\boldsymbol{\theta}}\, \chi + (D s_{\boldsymbol{\theta}} \chi)_{\boldsymbol{\theta}}\, \chi\right]. \end{aligned} \tag{8}$$

Although the general sensitivity equation (8) can be integrated numerically, it is useful to restrict to a further simplified system. Because in this study we are interested in the sensitivity with respect to a change of the gradient estimate magnitude, e.g., $p:\ \boldsymbol{\chi}^*(p) = \boldsymbol{\chi}(1 + p)$ and the kinetics parameters, but not in the sensitivity on the applied transport model 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, are additionally applied.

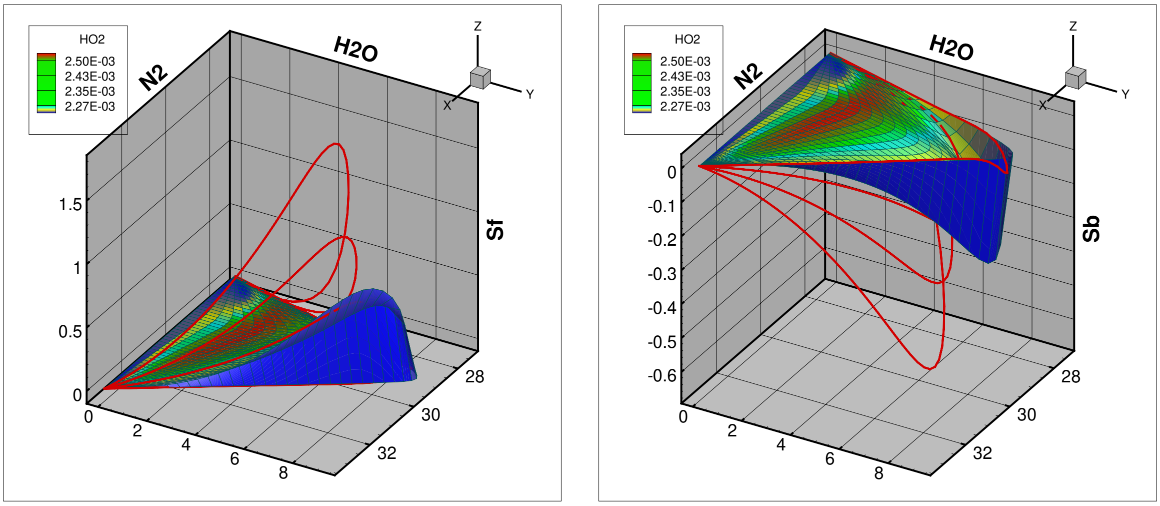

Figure 2: Sensitivity coefficients of H radical with respect to first (forward on the left) and second (backward on the right) elementary reactions, computed on 2D REDIM (shown by the mesh with H_2O_2 contours) and on 1D REDIMs - red lines, shown as 2D functions of H_2O and N_2 specific mole numbers.

3 Results

The 1D and 2D REDIMs equations are integrated and constructed for a counter-flow diffusion flame configuration of a $50 : 50$ mixture of H_2 : N_2 and air at a standard ambient pressure and temperature ($T_0 = 298$ K and $p_0 = 1$ bar). 1D REDIMs are constructed for different strain rates and results are shown in Figs. 2 and 3 by red lines. The following sensitivity matrices are computed for a number of n_r elementary reactions (A_j) and perturbation parameter p of the gradient estimate $(i = 1, ..., n_s;\ j = 1, .., n_r)$

$$s^r_{i,j} = \frac{\partial \psi_i}{\partial A_j},\ s^g_i = \frac{\partial \psi_i}{\partial p}.$$

Figure 1 summarizes the results of sensitivity study with respect to the rate coefficients of the elementary reactions, where forward and backward reactions are treated separately. Maximal (in absolute values) sensitivities of H and OH radicals with respect to the elementary reaction rates computed on the 2D REDIM are illustrated. The most important reactions for OH and H radicals are identified (only the 10 most important reactions are shown) depending on the local values of N_2 and H_2O. Figure 2 shows actual values of sensitivity of H with respect to reaction - H + O_2 = O + OH. The comparison shows that the 2D REDIM is less sensitive to the rate of this elementary reaction. Moreover, one can notice that for 1D REDIMs for different strain rates the larger the strain rate the higher the sensitivity.

Figures 3 show sensitivity vectors with respect to the gradient estimates. On the left a 2D projection is shown of the sensitivity vectors of (N_2, H_2O) projected onto 2D plane of (N_2, H_2O). Red curves show different 1D REDIMs, while red vectors (arrows) show the sensitivity of (N_2, H_2O) respectively. One can notice that the sensitivity grows with the increase of the strain rate. On the right figure 3D projections of sensitivity vectors (N_2, H_2O, H) are shown both along the 2D REDIM (mesh with H_2O_2 contours) denoted by cyan arrows and along 1D REDIMs presented by red lines and depicted by red arrows. It has to be noted that only after the 2D REDIM sensitivity vectors were amplified by a factor of 10^3 they show up within the same order of magnitude as the red arrows. This means the 2D manifold is by far less sensitive to the gradient estimates as expected. Additionally, one can easily see that the sensitivity vectors of 1D REDIMs point in the direction of the tangent space of the 2D REDIM.

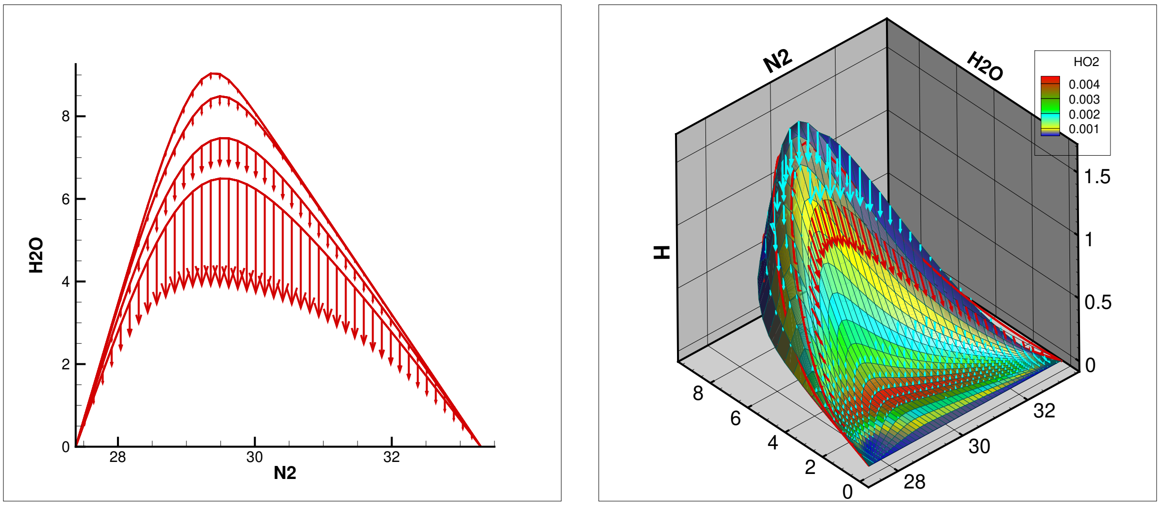

Figure 3: Sensitivity vectors projected onto 2D plane of specific mole numbers with 1D REDIMs (on the left) and 3D projections (right). 2D REDIM is shown by the mesh with H_2O_2 contours, while 1D REDIMs shown by red lines.

4 Conclusions

In this study the sensitivity analysis for REDIMs with respect to elementary reaction rate coefficients and gradient estimates was outlined and verified. The method to calculate the sensitivity was implemented in the REDIM evolution equation and integrated in a coupled way such that the converged REDIM solution provides the sensitivity automatically. The method was shown for 1D and 2D REDIMs, which were constructed for diluted hydrogen and air counter-flow diffusion flame. Several important observations were made, namely,

- the sensitivity of the slow manifold can be computed in a very generic manner providing information on the manifold sensitivity to the system parameters and gradient estimates;
- the sensitivity to the elementary reaction rates can be studied and used similarly as for the detailed solution such that crucial reactions which influence the form and the structure of the manifold can be singled out;
- with the increase of the manifold dimension the sensitivity with respect to the gradient estimates is reduced significantly, moreover, the sensitivity vectors for the gradients estimates of less dimensional manifolds (1D in this case) belong or remains close to the tangent space of manifolds of higher dimension (2D). The latter is a natural consequence of the hierarchical structure of the REDIMs.

Acknowledgments

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