Reaction-Diffusion Manifolds (REDIMs) method - gradient embedding approach

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1 Introduction

Modeling of combustion systems [1] has become an important tool for studying and predicting the behavior of combustion systems [2]. Detailed modeling of combustion systems requires huge computational resources [3], mainly because of the involved large chemical reaction mechanisms (with hundreds of reacting species and thousands of elementary reactions, e.g. gasoline surrogate for engine studies [4]).

Concepts based on low-dimensional manifolds in state space have been developed to reduce the large dimension of the system [3, 5, 6]. Tabulation approaches [5] to treat these manifolds have been developed, allowing to implement such reduced models for use in combustion simulations. Therefore, the problem of both high dimensionality and complexity, can be treated within such framework in a generic and automatic manner. Many different approaches based on manifolds were developed in the recent years dealing with construction and application of such low-dimensional manifolds e.g., the Intrinsic Low-Dimensional Manifolds (ILDM) [5], the Flamelet Prolongation of ILDM (FPI) [7], the flamelet model [8], the Reaction-Diffusion Manifolds (REDIM) [9] and Flamelet Generated Manifolds (FGMs) [10]. These methods above (as well as many others, see e.g., [3]) although similar, can be different in the way the manifold is determined and implemented. There are different assumptions made to single out certain low-dimensional subsets of the states in the system thermo-chemical state space with continuous / manifold structure. The latter is used then to redefine and to constrain the evolution of a reacting system solution profile onto this manifold. For instance, the REDIM methodology uses the hierarchical structure of the low-dimensional *invariant and slow* manifolds [11].

In this approach the manifold constructed describes a balance of molecular diffusion and reaction and decouples those chemical reaction processes, which have much smaller characteristic time scales as those of diffusion. In order to obtain and incorporate the information on the diffusion one should provide gradient estimates, which characterize and describe molecular diffusion characteristic time scales [9]. One can refer also to a number of numerical studies of this influence [12, 13]. This study further develops the implementation of Reaction-Diffusion Manifold (REDIMs) method [9, 14]. The methodology has already been developed and implemented to treat various combustion system configurations, see e.g., [14]. Here a generic approach to identify, quantify, and incorporate (integrate) the influence of the system gradient estimates is suggested.

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The REDIM equation for the low-dimensional manifold construction is briefly outlined first. The diffusion term of the REDIM equation is presented and discussed in detail. Different approaches to obtain and integrate the system gradient information are discussed. The embedding idea is formulated and presented with a test integration for 1D REDIM of freely propagating flames. The method is illustrated and verified by application to hydrogen premixed free flames configuration.

2 Mathematical model of reduced spaces - REDIM

The thermo-kinetic state vector of the system $\boldsymbol{\psi}$ is introduced as $\boldsymbol{\psi} = (h, p, \phi_1, \ldots, \phi_{n_s})^T$ having $n = n_s + 2$ dimensions given by 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. This thermo-kinetic state space vector for both laminar and turbulent reacting flows is considered to be "close" to and evolve on low-dimensional manifolds [5, 15]. The assumption restricts the evolution of the state space vector within this manifold

$$\mathcal{M} = \{\boldsymbol{\psi} = \boldsymbol{\psi}(\boldsymbol{\theta}),\ \mathbb{R}^m \to \mathbb{R}^n\},$$

here $\boldsymbol{\theta}$ is introduced as the m-dimensional reduced coordinate vector. This is simply a local coordinate of the manifold $\mathcal{M}$. The assumption of the invariance is employed at this stage assuming the system state vector - $\boldsymbol{\psi}$ depends on space and time only through the local coordinate - $\boldsymbol{\theta}(\vec{r}, t)$. Hence by using the manifold method, the solution of conservation equation system for the state $\boldsymbol{\psi}$ is replaced by the solution of an evolution equation for $\boldsymbol{\theta}(\vec{r}, t)$ and the whole system state is recovered from the manifold by $\boldsymbol{\psi}(\vec{r}, t) = \boldsymbol{\psi}(\boldsymbol{\theta}(\vec{r}, t))$. In the REDIM approach [9] an evolution equation is integrated, to find such a manifold $\mathcal{M}$.

$$\rho \frac{\partial \boldsymbol{\psi}(\boldsymbol{\theta})}{\partial t} = \mathcal{P}\ \left(F\left(\boldsymbol{\psi}(\boldsymbol{\theta})\right) + \Xi(\boldsymbol{\psi}, \boldsymbol{\psi}_{\boldsymbol{\theta}}, \boldsymbol{\psi}_{\boldsymbol{\theta}\boldsymbol{\theta}})\right), \tag{1}$$

Integrating the above equation to the steady state yields the slow invariant low-dimensional manifold, where ρ is density, $\mathcal{P}$ - projection onto the normal space the manifold [9], $\boldsymbol{F}(\boldsymbol{\psi})$ the n-dimensional vector of chemical source terms and $\Xi(\boldsymbol{\psi}, \boldsymbol{\psi}_{\boldsymbol{\theta}}, \boldsymbol{\psi}_{\boldsymbol{\theta}\boldsymbol{\theta}}) = \mathrm{div}\left(D\ \mathrm{grad}\boldsymbol{\psi}(\boldsymbol{\theta})\right)$, is the diffusion term computed on the manifolds. Here D the $n \times n$-dimensional matrix of detailed transport coefficients that might include diffusion, heat conduction, thermal diffusion, etc. For detailed transport the diffusion term becomes,

$$\Xi(\boldsymbol{\psi}, \boldsymbol{\psi}_{\boldsymbol{\theta}}, \boldsymbol{\psi}_{\boldsymbol{\theta}\boldsymbol{\theta}}) = (D\boldsymbol{\psi}_{\boldsymbol{\theta}}\chi)_{\boldsymbol{\theta}}\, \chi. \tag{2}$$

Here $\boldsymbol{\chi} = \mathrm{grad}\boldsymbol{\theta}$ is the gradient estimate of the local coordinate, which has to be provided as a function of the state variable, not as function of the spatial coordinate. Note that the REDIM method differs from most other manifold-based approaches in that it "adapts" the manifold to the reaction and diffusion properties of the modeled combustion system, and is not sampled based on detailed solution profiles computed under certain assumptions, which may or may not be valid. In order to close the REDIM evolution - relaxation equation Eq. (1), one needs to provide the function $\boldsymbol{\chi}(\boldsymbol{\theta})$, which governs the strength of the molecular diffusion.

2.1 Molecular diffusion - gradient estimates

The way the function $\boldsymbol{\chi} = \boldsymbol{\chi}(\boldsymbol{\theta})$ is obtained and provided is summarized below. First, one computes a detailed solution profile, say a steady solution profile given by $\boldsymbol{\psi} = \boldsymbol{\psi}^*(r)$. By differentiation of the steady solution one can compute the gradients $\mathrm{grad}\boldsymbol{\psi}^*$, which will be defined and available along the solution profile only. This will be the reference set that is used during the manifold evolution. This reference set is shown by 1D red line in the 2D projection of the system state space illustrated in

Figs. 1. Now, in order to integrate the REDIM equation and incorporate the information on $\mathrm{grad}\boldsymbol{\psi}^*$, the information available only on the red curve needs to be used while the REDIM evolves.

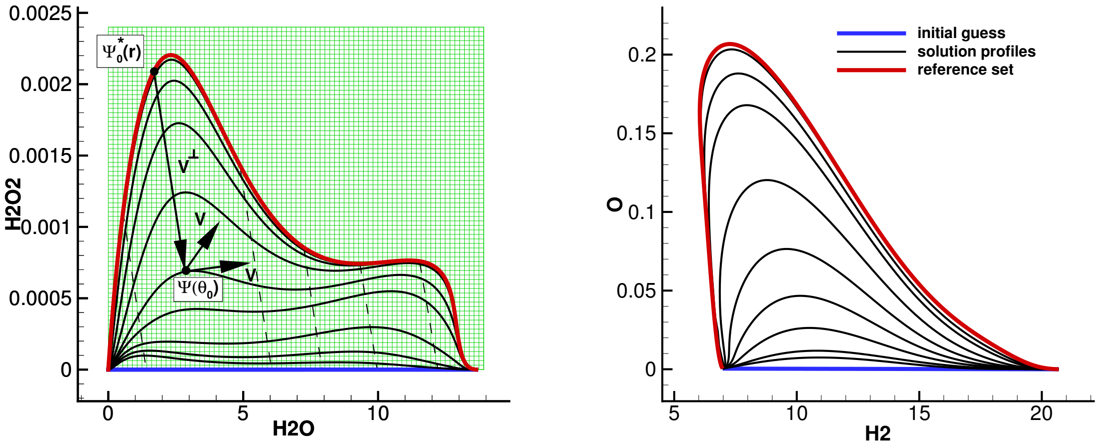

Figure 1: The figure shows the evolution of the one-dimensional solution profiles when solving Eq. (1). The system used here is a freely propagating flame configuration for a premixed hydrogen air mixture with equivalence ratio $\phi = 1.50$ at constant pressure.

These 1D REDIM evolving profiles as solution of Eq. (1) are shown also in Fig. 1 by black curves starting from an initial guess, which in this case is a linear interpolation between the unburnt composition and the chemical equilibrium. In the standard implementation of the REDIM, a so-called search vector V is specified and the gradient will be transferred from the reference set $\boldsymbol{\psi} = \boldsymbol{\psi}^*(r)$ to the point $\boldsymbol{\theta}_0$ on the evolving REDIM : $\boldsymbol{\psi} = \boldsymbol{\psi}(\boldsymbol{\theta}_0)$ by using this search vector - $\boldsymbol{\xi} = V\boldsymbol{\psi}$, namely,

$$\boldsymbol{\psi}_0^*(r) : V\boldsymbol{\psi}_0^*(r) = V\boldsymbol{\psi}(\boldsymbol{\theta}_0) = \boldsymbol{\xi}_0 \quad (3)$$

$$\boldsymbol{\chi}(\boldsymbol{\theta}_0) = \mathrm{grad}(\boldsymbol{\theta}_0) = \boldsymbol{\psi}_\theta^+(\boldsymbol{\theta}_0)\,\mathrm{grad}(\boldsymbol{\psi}_0^*(r)) \quad (4)$$

where the state $\boldsymbol{\psi}_0^*$, of the reference set $\boldsymbol{\psi} = \boldsymbol{\psi}^*(r)$, to retrieve the gradient information from is found along the line - $\boldsymbol{\psi} : V\boldsymbol{\psi} = V\boldsymbol{\psi}(\boldsymbol{\theta}_0) = \boldsymbol{\xi}_0$. The gradient information during REDIM evolution is obtained in two steps,

1. the minimum value of $\|V \cdot (\boldsymbol{\psi} - \boldsymbol{\psi}^*)\|$ over all the points in the reference set, is used to find the point $\boldsymbol{\psi}^*$ of the reference set that is closest to the point $\boldsymbol{\psi}(\boldsymbol{\theta}_o)$.

2. Then the gradient $\mathrm{grad}(\boldsymbol{\psi}_0^*(r))$ is determined by using simple interpolation on the reference set.

There are obvious drawbacks of this approach. First, an appropriate unique direction to pick the point with the minimum distance needs be used so that gradient information might be retrieved for all states on the REDIM and for all times of the evolution and relaxation. Secondly, there is a lot of post processing study needed to be verified, e.g., the properties of the system solution profile as well as relevance to the initial REDIM solution etc. For instance, in Fig. 1 on the right REDIM evolution process is shown in (H_2,O) projection. One can see that using H_2 as a search direction will not be appropriate because $H_2 = \mathrm{const}$ lines will not be suitable to retrieve gradient information during the 1D REDIM integration. Finally, the convergence of the REDIM is sensitive to the choice of the gradient search vector at least for low dimensional REDIMs. The suggested embedding procedure overcomes most of these difficulties and avoids demanding and extensive post-processing and REDIM convergence studies.

2.2 Gradient estimation - embedding

The goal is to construct a ”background field” from which the gradients required for the solution of the REDIM-equation can easily be retrieved. The sketch shows how the gradients $\mathrm{grad}(\boldsymbol{\psi}^*)$ could be chosen during the REDIM evolution process. As evolution progresses, it is not always clear what gradient has to be chosen for a given state. For this, information about the scalar-gradient correlations of a given combustion system is taken from a set of n_p empirical data points $(\boldsymbol{\xi}_i, \nabla\boldsymbol{\psi}_i)$, $i = 1, \dots, n_p$, similar to the reference set of the conventional method (as described in section 2.1). The new method is more generic than the conventional method in several aspects: First, the new method can use scattered sample points, while the conventional method requires that the sample points are ordered on a regular structure. Secondly, in the new method, the underlying sample space can —in principle— have any dimension $n_{\boldsymbol{\xi}}$, and so is not limited to dimension 1 or 2.

The field is obtained by constructing a smooth surface which runs through the set of empirically given data points, also fulfilling pre-defined boundary conditions. This method has been introduced in [16]; in that paper, it provided a smooth representation of a scalar function on a two-dimensional rectangular mesh. An extension that simultaneously provides multiple functions (i.e., a vector-valued function) on a rectangular mesh of —in principle— arbitrary dimension in $\boldsymbol{\xi}$-space is used now.

All physical information on scalar gradients of the state variables as a function of their values, i.e., of the function $\mathrm{grad}(\boldsymbol{\psi}) = f(\boldsymbol{\xi})$ in Eq. (4) with the parametrization function $\boldsymbol{\xi} = V\boldsymbol{\psi}$, where a k-dimensional parametrization matrix V, $(k \times n)$ is employed. Once this function is available at any manifold point $\boldsymbol{\theta}_0$ one can easily obtain the gradient estimates at this point $\boldsymbol{\theta}_0 : \ \boldsymbol{\psi}_0 = \boldsymbol{\psi}(\boldsymbol{\theta}_0)$ as

$$\begin{aligned} \boldsymbol{\xi}_0 &: \boldsymbol{\xi}_0 = V\boldsymbol{\psi}(\boldsymbol{\theta}_0) \\ \boldsymbol{\chi}(\boldsymbol{\theta}_0) &= \mathrm{grad}(\boldsymbol{\theta}_0) = \boldsymbol{\psi}_\theta^+(\boldsymbol{\theta}_0)\, f(\boldsymbol{\xi}_0). \end{aligned} \tag{5}$$

For a point $\boldsymbol{\xi}$ on the domain, $f(\boldsymbol{\xi})$ is retrieved by $n_{\boldsymbol{\xi}}$-dimensional interpolation on the rectangular, equidistant mesh on which the surface is given. For instance, in the case shown in Figure 2, the species H_2O and H_2O_2 care used for the vector $\boldsymbol{\xi}$. So the search matrix V will have the dimension $k = 2$, and the indices corresponding to the positions of the species H_2O and H_2O_2 in the state vector $\boldsymbol{\psi}$ will be provided by the matrix $V_{i,j} = \delta_{i,\ell(j)}$, where δ is the Kronecker-Delta, $i = 1, 2$ and $\ell(j) = 1$ for $j = H_2O$, $\ell(j) = 2$ for $j = H_2O_2$.

3 Results

In order to demonstrate the described method, we chose the simplest configuration of freely propagating Hydrogen air flames with a unity Lewis number assumption. The integration of process is started from an initial solution that is a mixing line and the system is then integrated.

Figure 1 shows the integration process leading to an invariant manifold. The integration process is started from initial states that are considerably far away from the invariant manifold and the gradient information is distributed on a two dimensional grid. During the evolution of the manifold, the gradients for a given point on the manifold are retrieved based on the location of this point $\boldsymbol{\xi}_0 = V\boldsymbol{\psi}(\boldsymbol{\theta}_0)$ on the extended grid.

To demonstrate the results, we compare the manifold obtained using the modified methodology with the one that uses the conventional method (where a one dimensional reference set is used). Fig. 2 (right) compares the manifold obtained against a steady state solution from a detailed calculation. The differences in the profiles of the major species can hardly be observed but for a minor species such as HO_2, the differences are more discernible. These differences observed in the manifold obtained using the modified approach is due to the fact that the gradients that are estimated from the modified approach will be different compared to the gradients that are provided from the steady state solutions due to differences in interpolation. Nonetheless, the method results in a manifold that is close to the steady state solution. This embedding idea that uses a set of higher dimension to provide fields for gradient estimates, along with solving the problem with proper location to obtain gradient estimates from, also improves the performance and robustness of the REDIM approach. By using this modified approach, a

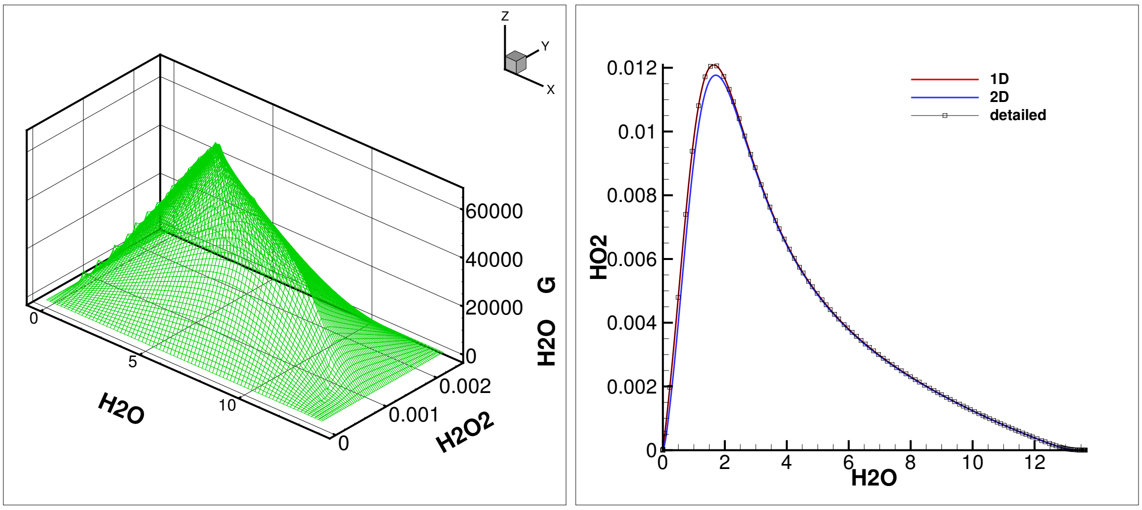

Figure 2: The figures show the extended gradient field and the comparison of the REDIMs obtained to the detailed solution. The legend entries refer to the method used to store the gradient information and the steady state solution from a detailed calculation respectively.

reference set of higher dimensions can be used to store the gradient information. The implementation does not require large modifications to existing methodology and can be extended to higher dimensions enabling storage and retrieval of more complex gradient data.

4 Conclusions

The method of gradient estimation for REDIMs with respect to the detailed integration and detailed solution profiles provided was briefly introduced and outlined. The method was illustrated by application to 1D REDIMs constructed for hydrogen premixed freely propagating flames. Several important conclusions were drawn, namely,

- A new approach to gradient retrieval during manifold evolution has been introduced and demonstrated;
- The method demonstrates how information from complex gradient fields can be incorporated into the manifold integration process and implemented in a very generic manner;
- The REDIM relaxation process can be performed by using gradient fields in dimensions higher than the manifold itself.
- The suggested modified approach improves the gradient retrieval procedure and makes it independent of the dimension of the manifold, and in addition increases the robustness considerably.

A simple configuration was chosen to demonstrate the results. There are, however, no limitations to particular combustion system, dimension of manifold and / or configuration.

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