

Validation of the Reaction-diffusion manifolds (REDIMs) reduced chemistry for the non-premixed CH₄ counter-flow diffusion flames under MILD condition

SUN Yuanxiang^{1*}, YU Chunkan², WU Chunwei² and Ulrich MAAS²

¹ State Key Laboratory of Explosion Science and Technology, Beijing Institute of Technology, Beijing, China

² Institute of Technical Thermodynamics, Karlsruhe Institute of Technology, Karlsruhe, Germany;

1 Introduction

Oxygen-enriched combustion is a well-known technology for high energy efficiency and low pollution emission [1]. This technology is an option for carbon capture and storage [2]. Together with internal gas recirculation this technology provides high energy efficiency and low NO_x emissions [3]. Furthermore, the oxy-fuel combustion technology combined with CO₂ capture is also developed to reduce emission of CO₂ significantly. Numerical simulations help to understand such phenomena.

For the numerical simulation of a general reacting system, one must solve a set of governing conservation equations for mass, species, momentum and energy. Suppose one has the thermo-kinetic state vector $\Psi = (h, p, \phi_1, \phi_2, \dots, \phi_{n_{sp}})^T$ with n_{sp} species, where h is the enthalpy, p the pressure and ϕ_i the specific mole number of i -th species defined as $\phi_i = w_i/M_i$ (w the species mass fraction and M the molar mass). Then the governing equation for the description of thermo-kinetic state vector can be formulated in the following PDEs in general symbolic vector notation [4]:

$$\frac{\partial \Psi}{\partial t} = F(\Psi) - \vec{v} \cdot \text{grad}(\Psi) - \frac{1}{\rho} \text{div}(D \cdot \text{grad}(\Psi)) = \Phi(\Psi) \quad (1)$$

where $F(\Psi)$ is the source term, \vec{v} the velocity field, ρ the density and D the $n \times n$ matrix of the transport coefficients. However, solving Eq.(1) is difficult due to two main reasons. First, chemical reactions involve hundreds of species and thousands of elementary reactions [4, 5]. Even for the smallest hydrocarbon fuel such as methane one should consider at least 30 species and more than 150 elementary reactions (e.g. Warnatz mechanism [6]). Increasing the number of involving reactive species leads to an increasing computational time. Second, and also the major difficulty, is that different chemical time-scales cause stiffness in PDEs [4, 5]. Usually chemical time-scales can cover a wide range of order of magnitude and in the numerical simulation one must set integration time step at most to be the smallest chemical time-scale to ensure a reliable and stable numerical solution. This is even more challenging for turbulent reacting flows. Therefore, model reduction for chemical kinetics is needed.

One possibility is the concept of low-dimensional slow manifolds, which uses the fact that trajectories are attracted under the influence of the fast time-scales [4]. As discussed in [7], the dimension of slow

manifolds becomes lower and lower if more fast time-scales become exhausted. Typically, the reduction method focuses on the source term of the system of governing equations and the physical transport such as molecular diffusion is not taken into account. The Intrinsic Low-dimensional Manifold (ILDM) [7], the laminar steady flamelet approach [8], flamelet generated manifold (FGM) [9], and reaction-diffusion manifolds (REDIM) [10] are several examples falling into this group.

In the present work, Reaction-diffusion manifolds (REDIMs) based reduced chemistry is used for modeling methane counter-flow diffusion flames under oxygen-enriched MILD condition. we focus on the REDIM method for the chemical kinetic simplification. In this method, the low-dimensional slow manifold is constructed by considering both chemical reaction and physical transports. The advantage of REDIM is that its generation can be achieved with least information about the combustion system.

2 Brief overview of Reaction-diffusion manifolds (REDIM)

In [10] Bykov and Maas suggested that the Reaction-diffusion manifolds (REDIM) can be obtained by solving the following partial differential equation system:

$$\frac{\partial \Psi(\theta)}{\partial \tau} = (I - \Psi_\theta \cdot \Psi_\theta^+) \cdot \Phi(\Psi(\theta)) \quad (2)$$

where $\Psi_\theta = \left(\frac{\partial \psi_i}{\partial \theta_j} \right)_{i,j}$ and Ψ_θ^+ is the Moore-Penrose pseudo-inverse of Ψ_θ . $(I - \Psi_\theta \cdot \Psi_\theta^+)$ is a projection operator that projects the thermo-kinetic state onto the normal space of the space. In this way, one restricts the thermo-kinetic state move tangential on the manifold. θ is the vector of the reduced coordinates that describes all thermo-kinetic state. Based on the Eq.(2), one obtain the REDIM simplified chemistry by integrating the Eq.(2) towards its stationary solution.

The generated REDIM simplified chemistry has an explicit formulation for the m_s -dimensional slow manifold defining as:

$$\mathcal{M} = \{ \Psi: \Psi = \Psi(\theta), \quad \mathbb{R}^{m_s} \rightarrow \mathbb{R}^n \} \quad (3)$$

For the numerical simulation based on REDIM reduced chemistry, only m_s conservation equations for θ are solved. The Eq.(1) can then be re-written into the form of reduced coordinate system as follows:

$$\frac{\partial \theta}{\partial t} = \Psi_\theta^+ \cdot \left(F(\Psi) - \frac{1}{\rho} \text{div}(D \cdot \Psi_\theta \cdot \text{grad}(\theta)) \right) - \vec{v} \cdot \text{grad}(\theta) \quad (4)$$

Since the REDIM reduced chemistry is described by an explicit formulation, one can use a tabulation strategy to store the thermo-kinetic states (θ), reduced reaction rates $\Psi_\theta^+ \cdot F(\Psi)$, and $D \cdot \Psi_\theta$ in the REDIM table. During the integration of Eq.(4), these stored quantities can be obtained via a table look-up algorithm.

3 Flame configuration and numerical set-up

The configuration of a counter-flow diffusion flame studied in the present work is consistent with the one studied in [11]. The fuel side is a mixture of CH₄/CO₂ with different fuel mole fractions X_F . The oxidizer side is a mixture of O₂/CO₂ with the mole fraction ratio O₂:CO₂=0.4:0.6. The temperatures of both sides are set to be 300 K and the system is working under atmospheric pressure.

The numerical simulation in this work is performed by using in-house code INSFLA [12]. The strain rate is defined as:

$$a = \sqrt{\frac{J}{\rho_u}} \quad (5)$$

where $J = 1/r \partial p / \partial r$ means the tangential pressure gradient, and ρ_u is the density of fuel side. In [11] it is also shown that for this flame configuration radiative heat loss has no significant effect on the extinction limits and numerical simulations based on adiabatic flame assumption are reasonable. Therefore, hereafter the computational results are all calculated using the adiabatic flame assumption. It is further discussed in [11] that the equal-diffusivity assumption ($Le=1$) is not a good assumption and the detailed transport model considering e.g. thermal diffusion must be considered. Therefore, in the following, a detailed transport model based on Curtiss-Hirschfelder approximation with thermal diffusion included [13] is applied for both detailed chemistry simulation and generation and application of REDIM reduced chemistry.

4 Generation of REDIM reduced chemistry

In this section, we will provide the procedures generating REDIM reduced chemistry. The CH₄/CO₂ with fuel mole fraction $X_F = 0.4$ versus O₂/CO₂ counter-flow diffusion flame will be used as example. For other fuel mole fractions, the same procedure is performed to generate the corresponding 2D REDIM reduced chemistry. For the generation of REDIM reduced chemistry, Eq.(2) can be reformulated into the following PDEs (derivation see [10]):

$$\frac{\partial \Psi(\theta)}{\partial \tau} = (I - \Psi_\theta \cdot \Psi_\theta^+) \cdot \left(F(\Psi) - \frac{1}{\rho} (D \cdot \Psi_\theta \cdot \xi(\theta))_\theta \cdot \xi(\theta) \right) \quad (6)$$

where $\xi(\theta) = \text{grad}(\theta)$. $\xi(\theta)$ is the gradient estimation, which has to be provided for the integration of Eq.(6) (see below).

For reduced chemistry that is able to capture both steady and extinction behavior, a 2D REDIM reduced chemistry is generated. To integrate the Eq.(6), we must first specify the boundary condition, corresponding to the application range for the REDIM reduced chemistry, as shown in Fig.2. The upper boundary in Fig.2 is obtained from the flamelet with strain rate $a = 100 \text{ s}^{-1}$ using detailed chemistry, and the low boundary from the pure mixing of fuel and oxidizer. Both upper and boundary conditions (Solid lines in (a) in Fig.2) encloses the whole application domain.

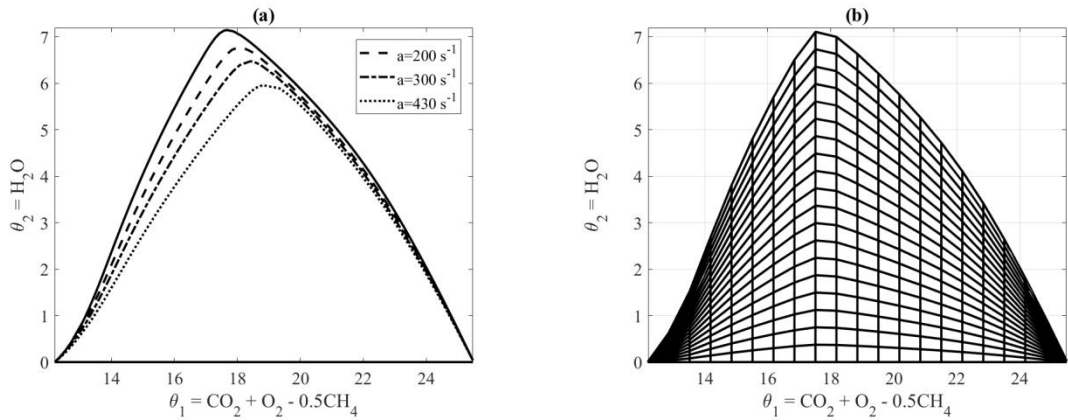


Fig.2. Illustration of generation of 2D REDIM reduced chemistry.

The next step is to provide the gradient estimation $\xi(\theta)$. Note that with increasing dimension of REDIM, the gradient estimation becomes less and less important [10]. However, for a 2D REDIM reduced chemistry, a reasonable gradient estimation should be provided. There are several possibilities to find appropriate gradient estimation, and one possibility (and also used in present work) is to obtain $\xi(\theta)$ from simulation of combustion scenarios based on detailed chemistry. For example, information about the detailed system gradients (flamelets with $a = 200 \text{ s}^{-1}$, 300 s^{-1} and 430 s^{-1}) is used on the system rough gradient estimation $\xi(\theta)$, as illustrated in (a) in Fig.2 (dashed lines). The gradient estimation for the whole domain is obtained by linear interpolation.

The third step is to provide the initial profile for the integration of Eq. (6). The initial profile is the least important point for the generation of REDIM reduced chemistry, because we are only interested in the steady solution of Eq.(6). However, numerical experiment shows that a “good” initial profile speeds up the convergence of Eq.(6). Therefore, we use the initial profile consists of flame scenarios with $a = 200 \text{ s}^{-1}$, 300 s^{-1} and 430 s^{-1} as used in gradient estimation. And all states between boundary conditions and these flamelets are linearly interpolated.

Another important numerical point of view is the grid discretization. In order to have a good grid resolution, we select $\theta_1 = \text{CO}_2 + \text{O}_2 - 0.5 \cdot \text{CH}_4$ and $\theta_2 = \text{H}_2\text{O}$, as shown in (b) in Fig.2. We discretize 100 grids in θ_1 and 100 grids in θ_2 (due to optical reason we only show 20×20 grids in (b) in Fig.3). This grid resolution is high enough so that the results of steady laminar flame do not change with increasing grid numbers. Note that here such choice of θ aims only at having a good grid discretization. The REDIM reduced chemistry itself does not depend on the choice of θ [14].

5 Results and discussion

In this section, we validate our 2D REDIM reduced chemistry for CH₄/CO₂ with fuel mole fraction $X_F = 0.4$, as proposed in section 4. In Fig.3, we plot the maximum of temperatures and H-radical against increasing strain rates until the quenching limit. Symbols represent the solution based on detailed chemistry, and solid lines the solution based on 2D REDIM reduced chemistry. It can be seen clearly that the 2D REDIM reduced chemistry can represent the thermo-kinetic quantities very well, although minor deviations can be observed. The only major errors occur in the quenching domain. The 2D REDIM reduced chemistry predicts the quenching limit with $a_E = 412 \text{ s}^{-1}$, while detailed chemistry with $a_E = 431 \text{ s}^{-1}$. This error is relatively small and still represents an acceptable accuracy (in particular if one compares this difference with the differences that are obtained when using different detailed mechanisms).

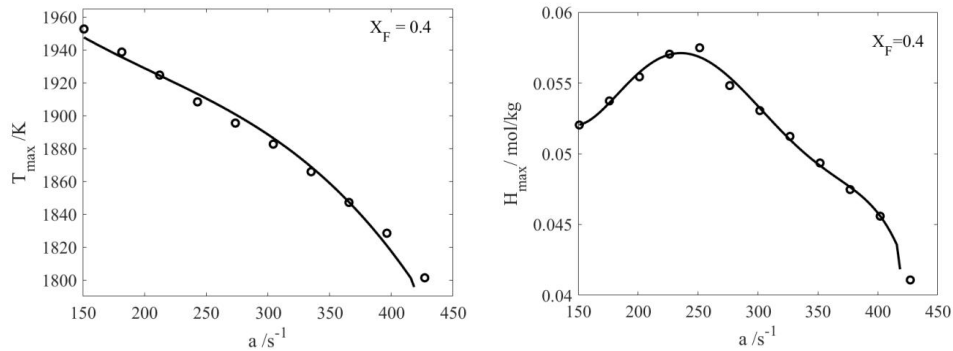


Fig.3. Maximum of temperature and H-radical against increasing strain rates. Symbols: detailed chemistry; Solid lines: 2D REDIM reduced chemistry.

In Fig. 4 the extinction process for the considered counter-flow diffusion flame is shown. The system has initially a strain rate $a = 200 \text{ s}^{-1}$, and the strain rate is suddenly increases to $a = 500 \text{ s}^{-1}$ as Heaviside step function in time. The quantities shown are in the position where the maximal temperatures occur. We observe that although both profiles do not overlap with each other, 2D REDIM reduced chemistry predict qualitatively very well with the detailed chemistry. We want to recall that during the generation of 2D REDIM reduced chemistry, no information about the extinction regime is needed. Using the REDIM reduced model, we can re-produce the extinction process.

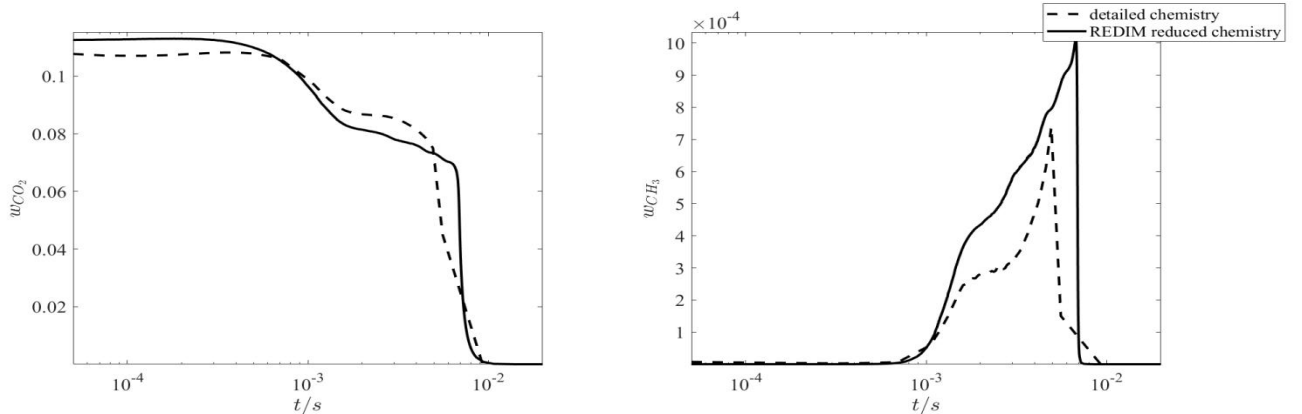


Fig.4. Extinction process based on detailed chemistry (dashed line) and 2D REDIM reduced chemistry (solid line). Left figure: CO₂. Right figure: CH₃.

6 Conclusions

In this work, Reaction-diffusion manifolds (REDIMs) based simplified chemistry is applied for the numerical simulation of a CH₄/CO₂ versus O₂/CO₂ counter-flow diffusion flame. The REDIM simplified chemistry can be generated easily with little information on the considered flame system. It is shown that the thermo-kinetic quantities can be well predicted by REDIM reduced chemistry for different strain rates until the extinction limit, and although no information for extinction regime needs to be known for the generation of REDIM simplified chemistry, the generated REDIM chemistry can re-produce the extinction limit very well. The extinction strain rate and time histories during extinction processes can be further improved by increasing the dimension of REDIM simplified chemistry to three dimensions, which is the subject of future research.

Acknowledgments

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