

Study of extinguishing limits of diluted hydrogen-air counter-flow diffusion flames with the REDIM method

A. Neagos, V. Bykov, U. Maas
Karlsruhe Institute of Technology
Karlsruhe, Germany

1 Introduction

Technological and environmental demands in using internal combustion engines (e.g. increasing the efficiency, reducing pollutants (CO_2 , NO_x etc.) emission) lead to an interest in developing a better understanding of the oxidation process of engine fuels [1]. In order to increase the efficiency of combustion facilities and reduce the emission of pollutants predictive models of chemical kinetics have to be developed over a wide range of the engines operating conditions (of pressure, temperature, and equivalence ratio, especially for lean fuel mixture composition).

Within this development the chemical kinetics of hydrogen plays a crucial role. Not only in case of direct use of hydrogen as a fuel but also in the combustion chemistry of all hydrocarbon fuels the hydrogen mechanism is of great importance. Therefore very detailed knowledge of both the chemical system and the transport properties of the contributing species is necessary [2]. Meanwhile many different mechanisms and transport models for hydrogen are available, leading to differences in the predictions and therefore to a range of uncertainty.

Combustion models are all facing the big challenge to predict the critical or transient regimes of combustion correctly, namely ignition, re-ignition and extinction processes. In addition to that, a very complicated non-monotonic pressure dependence needs to be addressed more accurately by the detailed kinetic mechanisms [2]. Still, the computational effort of detailed combustion models is very high. The numerical treatment of the system of governing equations describing chemically reacting flows is complicated enormously by the high dimensionality, non-linearity and stiffness of typical chemical reaction mechanisms. Consequently, there has been increasing effort in developing methods for automatic model reduction [3–5]. The reduction method of REaction DIffusion Manifolds (REDIM) is one of few which also take molecular transport into account [6].

In the present work the ability of the REDIM method to capture extinction phenomena in counterflow diffusion flames at elevated pressure is demonstrated. It is shown how three main difficulties of model reduction methodologies are overcome by the developed method. First of all, the system transient behavior is accounted for. Secondly, influence of molecular transport on the reduced model is taken into account explicitly. Thirdly, the case of the non-equal diffusivities is considered and pressure dependence is studied. All these aspects represent difficult problems for modeling of reacting flows and, as a consequence, for model reduction methodologies.

2 The REDIM model reduction method

For convenience the system of governing equations of a reacting flow is considered in the vector notations. One can show that the $(n = n_s + 2)$ -dimensional system state vector $\Psi = \left(h, p, \frac{w_1}{M_1}, \dots, \frac{w_{n_s}}{M_{n_s}} \right)^T$ evolves according to

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

where v represents the velocity field, ρ the density and D is the $(n \text{ by } n)$ -dimensional matrix of the transport coefficients [7] (note that this formulation includes heat conduction, ordinary diffusion and thermal diffusion), h denotes the enthalpy, p the pressure, $w_1, \dots, w_{n_s}$ the species mass fractions and $M_1, \dots, M_{n_s}$ the molar masses.

The main idea of the REDIM method is based on the assumption that the profile of a reacting system during the entire process does belong (remains confined) to the low dimensional manifold $M : \Psi = \Psi(\theta)$ embedded into the detailed thermo-chemical state of the system. Thus, in order to reduce the system a manifold needs to be found / constructed. It was suggested to look for an approximately invariant manifold of relatively slow motions with respect to the system (1).

Formulating the terms of the RHS of Eq. (1) in terms of the local parameters θ and applying the invariance condition yields the equation for the REDIM evolution in the form

$$\frac{\partial \Psi(\theta)}{\partial t} = (I - \Psi_\theta \Psi_\theta^+) \cdot \left(F(\Psi) + \frac{1}{\rho} \{ D \cdot \Psi_\theta \text{div}(\text{grad}(\theta)) + (D \Psi_\theta)_\theta \circ \text{grad}(\theta) \circ \text{grad}(\theta) \} \right). \quad (2)$$

The convection term $v \text{grad}(\Psi) = v \Psi_\theta \text{grad}(\theta)$ in Eq. (1) cancels out after applying the invariance condition [6]. In the case of equal diffusivity assumption employment (the diffusion matrix equals $D = d \cdot I$) the term $D \cdot \Psi_\theta \text{div}(\text{grad}(\theta))$ also cancels out because it belongs to the tangent subspace of the manifold. For the case of general transport matrix the term $D \cdot \Psi_\theta \text{div}(\text{grad}(\theta))$ complicates a lot the evolution equation because not only the first derivative $\text{grad}(\theta)$ of the parameter has to be estimated or specified for an approximation of the invariant manifold, but the second derivative ($\text{div}(\text{grad}(\theta))$) must be estimated as well.

In terms of the flamelet [8] terminology for a 1D spatial configuration for manifold construction not only a scalar dissipation rate in the second term of Eq. (2) should be taken into account, but the second derivative ($\text{div}(\text{grad}(\theta)) = \frac{\partial^2 \theta}{\partial x^2}$) has to be estimated. Thus, Eq. (2) can be rewritten in the form

$$\frac{\partial \Psi(\theta)}{\partial t} = (I - \Psi_\theta \Psi_\theta^+) \cdot \left(F(\Psi) + \frac{1}{\rho} \left\{ D \cdot \Psi_\theta \circ \frac{\partial^2 \theta}{\partial x^2} + (D \Psi_\theta)_\theta \circ \frac{\partial \theta}{\partial x} \circ \frac{\partial \theta}{\partial x} \right\} \right). \quad (3)$$

This equation is implemented to construct the REDIM. In a number of works the REDIM method was developed, improved and extended to yield a reliable reduction methodology. In [9] it was shown how the REDIM performs in the case of premixed and non-premixed flames. Then it was modified in [10] to describe the case of non-equal diffusivities in and making it more problem oriented. Finally, it was applied to study transient, non-stationary and non-premixed flames in [11].

3 Implementation

In this study a 1D counter-flow diffusion-flame of hydrogen diluted by nitrogen and air is considered. The quantities at the oxidizer boundary are denoted by $x_{O_2,o} = 0.21$ and $x_{N_2,o} = 0.79$ as the mole fractions of oxygen and nitrogen, respectively, $T_o = 298K$ as the temperature, v_o as the fuel velocity and

a_o as the strain rate. The quantities at the fuel boundary are denoted by $x_{H_2,f} = 0.14$ and $x_{N_2,f} = 0.86$ as the mole fractions of hydrogen and nitrogen, respectively, and $T_f = 298K$. The strain rate at the oxidizer boundary is given by $a_o = \frac{4v_o}{L}$, where $L = 0.01m$, and denotes the distance between the ducts [2]. The equation for a_o is valid, if the momenta at each boundary are kept equal to each other [2] and the Reynolds numbers at the boundaries are presumed to be large [12]. Note that in the present numerical calculations, the velocities at the boundaries arise by defining the mass flux on the oxidizer side and the tangential pressure gradient on the fuel side, respectively. Both were adapted to keep the flame in the center of the domain for each calculation. Hence, using the strain rate as a critical parameter is still reasonable, even if small differences in the boundaries momenta occur.

3.1 Detailed model

The Miller mechanism [13] was used to perform the detailed calculations. Figure 1 (a) illustrates temperature curves as a function of the axial coordinate for different pressure values at a strain rate $a_o = 100s^{-1}$. With increasing pressure the reaction zone is thinned out, showing the influence of the pressure on the molecular transport. The profiles show good agreement with the numerical results based on the San Diego hydrogen oxidation mechanism used in [2].

Figure 1 (b) shows the influence of the strain rate on the stationary profiles at a constant pressure of 2bar onto the H₂O – N₂-plane. a_o is varied from $300s^{-1}$ to $720s^{-1}$, which is the maximum strain rate before extinction. Obviously the range in which stationary solutions exist is very narrow, while the region in which transient extinction happens is much wider. The aim of the present work is to find out, whether the REDIM method is able to capture both the range of the stable and the unstable region.

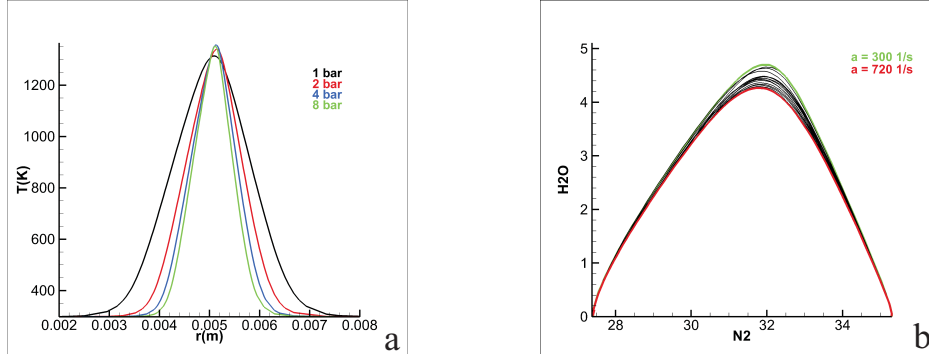


Figure 1: (a): Temperature profiles at different pressures. (b): Stationary solutions at 2bar for $300s^{-1} < a_o < 720s^{-1}$.

3.2 REDIM implementation

In order to solve the equation of the REDIM evolution (Eq. (3)) an initial solution for the manifold is needed. The initial manifold is constructed based on a set of stationary flamelets which are theoretically close to the manifold in the state space [11]. The different stationary flamelets are achieved through a variation of the strain rate. The initial manifold is bounded by two characteristic stationary solutions. One is the mixing line and its opposite is the stationary solution linked to the lowest strain rate. As it was pointed out above the REDIM evolution equation is integrated until a stationary solution is obtained, thus yielding a tabulated form of the state space [6].

Note that the effect of differential diffusion becomes significant for the considered geometry configuration and highly diluted hydrogen flames. The (N₂, H₂O) plane is used to define the local coordinate frame, where specific mole number of N₂ is used to trace the mixing process (similarly to the mixture fraction), while H₂O is taken to reproduce reaction progress [11]. Calculations however are carried out using the generalized local parameters (θ_1, θ_2).

Figure 2 shows the REDIM manifolds for two different cases of equal (a) and non-equal (b) diffusivities albeit for two different flame configurations as well as mixture compositions. Although it is shown for different parameters one can see clearly the differences in the shape of the REDIM manifold of these cases. Manifolds are constructed for four different pressures, namely 1bar, 2bar, 4bar and 8bar. Calculations of the extinction limits are performed for each pressure and compared with the detailed results.

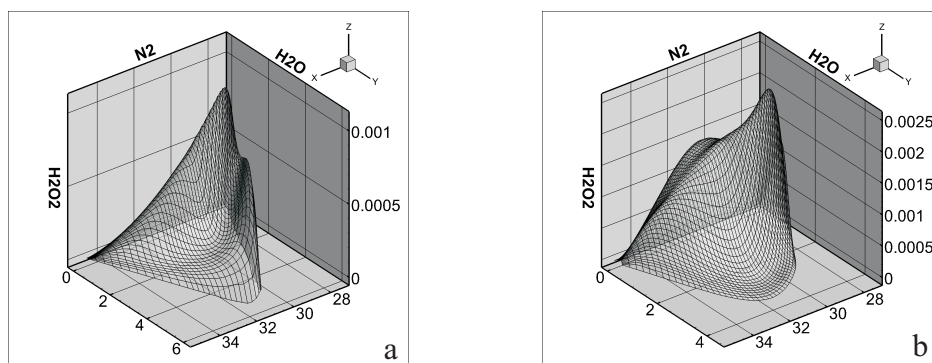


Figure 2: REDIM of equal (a) and non-equal diffusivities (b) in projections onto (N₂, H₂O, H₂O₂) specific mole numbers.

4 Results

The comparison of detailed and reduced calculations is carried out for four different pressures. Figure 3 depicts the constructed manifolds at each pressure in the N₂ – H₂O – OH projection of the state space from two different angles. With increasing pressure the specific mole numbers of the minor species OH are decreasing throughout the considered domain in state space. This is an indication for the thinner reaction zone in a high pressure atmosphere (see Fig. 1).

In particular, the lower specific mole numbers of the radical OH for increasing pressure indicate that the REDIM is able to address the crucial mechanisms of chain-branching and chain-termination in the hydrogen mechanism. With increasing pressure the chain-termination step $\text{H} + \text{O}_2 + \text{M} \rightarrow \text{HO}_2 + \text{M}$ is dominating because of the higher concentration of the third body M [14].

Additionally, the local analysis of the eigenvalues of the Jacobian matrix of the chemical source term restricted to the tangential subspace shows that one eigenvalue changes sign in a characteristic region within the manifold (contour plot Fig. 3). The existence of the positive eigenvalue means that the chemical kinetics exhibits an unstable behavior which is the reason for an absence of stationary flamelets in the corresponding domain [11].

In Fig. 4 the stationary solutions of the detailed and the reduced calculations at 1bar are compared for three different strain rates. The profiles show excellent agreement in the projection onto the N₂/H₂O-plane (a) as well as in 3D-projection (b). The profiles with the lowest specific mole numbers in OH represent stationary solutions near the extinction limit. In this case both the detailed and the reduced

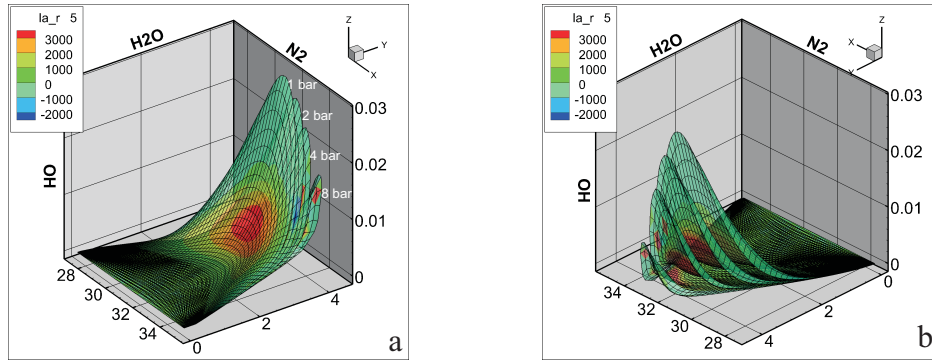
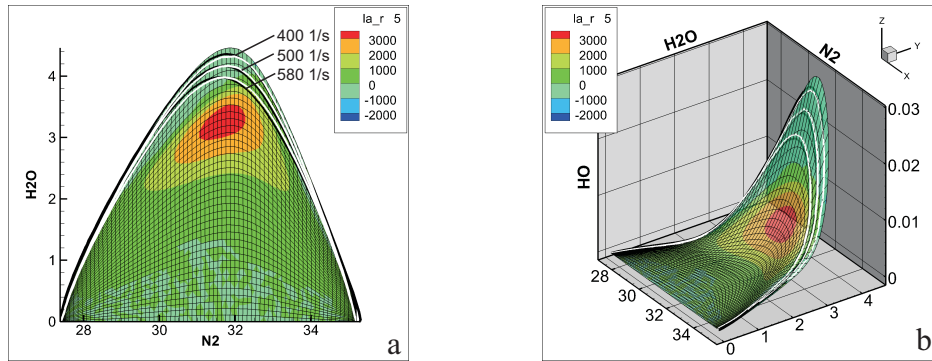


Figure 3: Manifolds at pressures of 2bar, 4bar, 6bar and 8bar.

profiles are located near the unstable region with positive eigenvalues. Thus, increasing the strain rate would push the profiles into the unstable region leading to an extinction of the flame (see also Fig. 1 (b)).

Figure 4: Comparison of the stationary solutions of detailed (black) and reduced solutions (white) at $a = 400s^{-1}$, $500s^{-1}$ and $580s^{-1}$.

The extinction limit strain rates are established for the detailed and the reduced model at each pressure by slowly increasing the strain rate at the boundaries and keeping the flame always in the center of the considered domain. Corresponding to [2] a non monotonic relation between the pressure and the critical strain rate was reported. As it can be seen in Tab. 1 both the detailed and the reduced calculations are able to capture this behaviour.

	detailed	reduced
1bar	$560s^{-1}$	$600s^{-1}$
2bar	$720s^{-1}$	$760s^{-1}$
4bar	$580s^{-1}$	$680s^{-1}$
8bar	$320s^{-1}$	$300s^{-1}$

Table 1: Strain rates at extinction for different pressures.

5 Conclusion

The main outcome of the current study is the fact that the REDIM method of model reduction is able to address near limit counter-flow diffusion flames at different pressures. It was shown that two dimensions

(two parameters) are enough to reproduce the non-monotonic function of critical strain rate and system pressure in the case of a detailed transport model. Although the simple model example of hydrogen combustion is considered, it is nevertheless very important as a basic building block of any detailed model of hydrocarbon combustion mechanisms and contains all main features of more complex mechanisms. An additional analysis of the time scales of the chemical processes shows that the REDIM captures the time scale separation introduced by the chemical kinetics. Thus, not only a manifold of relatively slow motions is constructed, but it is also shown that the REDIM is able to describe the transient behavior within this slow manifold. Estimations of critical system parameters show a potential of the REDIM to address adequately critical and non-stationary system behavior.

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