

LAMINAR FLAME CALCULATIONS USING SIMPLIFIED CHEMICAL KINETICS BASED ON INTRINSIC LOW-DIMENSIONAL MANIFOLDS

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During the last 10 years, a great success has been achieved in the field of detailed mathematical modeling of combustion processes. However, most detailed models are restricted to the simulation of simple one-dimensional laminar flames, and the extension of detailed kinetic models to general reacting flows of practical importance (e.g., turbulent flow in internal combustion engines) is computationally prohibitive. Thus, simplified kinetic models have to be used. Recently, we presented a mathematical model, the method of intrinsic low-dimensional manifolds (ILDM), which reduces the chemical kinetics automatically. The only inputs to the procedure are the detailed reaction mechanism and the desired number of degrees of freedom. The reduction of the kinetics is performed using the assumption that the fastest timescales are in local equilibrium and can be decoupled. This paper discusses the implementation of the method in reacting flow calculations. The procedure is developed for general three-dimensional reacting flows, which are governed by the system of conservation equations, and the coupling of the reduced chemical kinetics with molecular transport processes and convection is discussed. Then, the mathematical model is verified by sample calculations of structures of laminar premixed flat flames, which provide a simple, but realistic test case. Examples, which verify the approach, are shown for H_2-O_2 and syngas-air flames. Even for these simple examples, a considerable speedup of the computations is observed. However, the method can also be used for other fuels and, thus, will allow an efficient treatment of combustion systems of practical importance.

Introduction

During the last 10 years, the interest in simplified descriptions of chemical reaction systems for the simulation of complex combustion processes has increased considerably, and a variety of different approaches can be found in the literature [1–7]. The main motivation for the use of reduced mechanism is the fact that due to the complexity of the combustion processes usually encountered in practical applications, methods have to be found to describe the chemical reaction by only a small number of species, while still retaining the essential dynamics of the system. Recently, we developed a method, the method of intrinsic low-dimensional manifolds (ILDM), which provides a simplification of the chemical kinetics starting from a detailed reaction mechanism. The method is based on the dynamic systems theory. Besides the detailed reaction mechanism, the only input to the scheme is the desired number of degrees of freedom (i.e., the number of reaction progress variables). Sample calculations for a perfectly stirred

reactor [9] have shown that the method yields good results and is able to describe the coupling of chemical reaction with physical processes like mixing or molecular transport.

A nice test case for the application of the ILDM method in reacting flow computations are laminar flames. Although being sufficiently complicated systems with various practical applications, they nevertheless allow a direct comparison with results of detailed calculations. Various investigations over the last few years have shown that up to two-dimensional laminar flame structures can be calculated using detailed reaction and transport models [10–12]. In most cases, the agreement with experimental results is very good. Typical one-dimensional laminar flame calculations take less than 1 h on modern work stations. However, structures of laminar flame calculations are tabulated quite often for use in flamelet models of turbulent flames [13,14]. In this case, many (sometimes more than 1000) flame structures are needed, and the use of detailed kinetics then causes an enormous computational effort. In the sim-

ulation of two-dimensional laminar flames, the computational effort is tremendous [11,12], and reduced mechanisms are desirable anyway. Thus, because laminar flames allow a good comparison between reduced and detailed reaction mechanisms and because their fast simulation is of practical importance, it shall be shown in the following that the concept of intrinsic low-dimensional manifolds can be applied successfully to these systems.

Mathematical Model

Governing Equations:

The dynamic behavior of a chemically reacting flow is governed by the system of conservation equations, namely, the conservation of mass, momentum, energy, and species masses [15,16]. These equations can be rewritten to yield equations for the scalar field of a reacting flow, which is described by the state vector $\underline{\psi} = (h, p, \phi_1, \phi_2, \dots, \phi_{n_s})^T$. Here, a formal separation into physical processes $\underline{\mathcal{E}}$ and chemical production terms \underline{F} shall be made. We obtain

$$\begin{aligned}\frac{\partial h}{\partial t} &= F_h + \underline{\mathcal{E}}_h \\ \frac{\partial p}{\partial t} &= F_p + \underline{\mathcal{E}}_p \\ \frac{\partial \phi_i}{\partial t} &= F_{\phi_i} + \underline{\mathcal{E}}_{\phi_i} \quad i = 1, 2, \dots, n_s\end{aligned}\quad (1)$$

or in vector formulation

$$\frac{\partial \underline{\psi}}{\partial t} = \underline{F} + \underline{\mathcal{E}}\quad (2)$$

with $\underline{F} = (F_h, F_p, F_{\phi_1}, F_{\phi_2}, \dots, F_{\phi_{n_s}})^T$ and $\underline{\mathcal{E}} = (\underline{\mathcal{E}}_h, \underline{\mathcal{E}}_p, \underline{\mathcal{E}}_{\phi_1}, \dots, \underline{\mathcal{E}}_{\phi_{n_s}})^T$, and

$$\begin{aligned}F_h &= 0; \quad \underline{\mathcal{E}}_h = \frac{f_p}{\rho} - \mathbf{v} \cdot \text{grad } h - \frac{1}{\rho} \text{div } \mathbf{q} - \frac{1}{\rho} \\ &\quad \cdot \bar{p} : \text{grad } \mathbf{v} + \frac{1}{\rho} \text{div } \rho \mathbf{v} + \frac{\omega_h}{\rho} \\ F_p &= 0; \quad \underline{\mathcal{E}}_p = f_p \\ F_{\phi_i} &= \omega_i / \rho; \quad \underline{\mathcal{E}}_{\phi_i} = -\frac{1}{\rho M_i} \text{div } \mathbf{j}_i - \mathbf{v} \cdot \text{grad } \phi_i; \\ &\quad i = 1, 2, \dots, n_s\end{aligned}\quad (3)$$

where t = time, h = specific enthalpy, p = pressure, ρ = density, \mathbf{v} = velocity vector, \bar{p} = pressure tensor, \mathbf{q} = heat flux density, ω_h = source term for energy (e.g., radiation), n_s = number of species, ϕ_i

= specific mole number of species i ($\phi_i = W_i/M_i$), W_i = mass fraction of species i , M_i = molar mass of species i , \mathbf{j}_i = diffusion flux density of species i , and ω_i = molar rate of formation of species i . The symbol \otimes denotes the dyadic product of two tensors, and $:$ denotes the twofold contraction of two tensors. This choice of the dependent variables is quite arbitrary. Other formulations, like conservative, could be used, too. The term f_p describes the time behavior of the pressure. Because a constant pressure is used quite often in laminar flame calculations ($f_p = 0$), this term shall not be written down explicitly in this paper. Furthermore, note that the $\underline{\mathcal{E}}$ terms are complicated functions of the scalars, the velocity field, and the first and second spatial derivatives.

Coupling of Chemical Kinetics with Convection and Molecular Transport:

The basic assumption of manifold methods for the reduction of chemical kinetics is that the state ψ is (at any time and at any point in the flow) close to an attracting low-dimensional manifold (of dimension N). Then the dynamics of the reactive system can be approximated by assuming that the state is confined to those N -dimensional manifolds and is thus a function of N variables only, which describe the dynamics within the manifold. It has been shown in Refs. 8 and 9 that the method of intrinsic low-dimensional manifolds can be used to simplify chemical kinetics. This is done by projecting the governing equation system for the scalar field onto the low-dimensional manifolds. In this way, the $(2 + n_s)$ -dimensional equation system 2 is transformed into an equation system of much lower dimension (N) for the variables $\underline{\tau} = (\tau_1, \tau_2, \dots, \tau_N)^T$. The simplification of the detailed reaction mechanism yields $\underline{\psi}(\underline{\tau})$, which is the equation of the low-dimensional manifold, \underline{S} , and, in addition, provides information on how to project the physical processes $\underline{\mathcal{E}}$ onto the manifold. The variables $\underline{\tau}$ are, e.g., enthalpy, pressure, variables specifying the element composition, and reaction progress variables. Usually, the results of the mechanism reduction (i.e., the low-dimensional manifolds) are tabulated in terms of the variables $\underline{\tau} = (\tau_1, \tau_2, \dots, \tau_N)^T$ and stored for subsequent use in other reacting flow codes. This includes, e.g., the storage of the reduced reaction rates \underline{S} (rates of change of the reaction progress variables), the species mass fractions, and information about the local timescales. At this time, we want to emphasize the numerical issue that the reaction rates obtained directly from the reduction procedure are stored and used in the laminar flame calculation. Evaluating the reaction rates from the stored species mass fractions might lead to large errors, as can be shown by some simple examples [21]. Details about the mechanism reduction, the choice of the reaction progress variables, the tabulation pro-

cedure, and the subsequent table lookup and interpolation can be found in Refs. 8 and 9.

Physical perturbations tend to move the system off the intrinsic low-dimensional manifolds. However, if slow physical processes move the system off the manifold, the fast chemical processes cause a relaxation of the system back to the manifold. It can be shown that convection always leads to movements within the manifold. Thus, the only physical perturbation, which tends to move the system off the manifold in the equations above, is molecular transport.

Let the reacting system be described by the $(2 + n_e + n_c)$ parameters $h, p, \chi_1, \dots, \chi_{n_e}$, and $\tau_c = (\tau_{c,1}, \dots, \tau_{c,n_c})^T$ of the reduced scheme, where χ_j denote the n_e specific element mole numbers ($\chi_j = w_j/M_j$), w_j denote the mass fraction of element j , M_j denote the molar mass of element j , n_e denotes the number of elements in the reaction system, and $\tau_{c,i}$ denote the reaction progress variables. Then the state $\underline{\psi}$ is a known function of these parameters $\underline{\tau} = (h, \bar{p}, \chi_1, \dots, \chi_{n_e}, \tau_{c,1}, \dots, \tau_{c,n_c})^T$.

Using the results of Ref. 9, it can be shown (see appendix) that the governing equations of Eq. (1) can be rewritten as follows:

$$\begin{aligned} \frac{\partial h}{\partial t} &= \underline{\mathcal{E}}_h \\ \frac{\partial p}{\partial t} &= \underline{\mathcal{E}}_p \\ \frac{\partial \chi_i}{\partial t} &= \mu_i^T \underline{\mathcal{E}}_\phi \quad i = 1, 2, \dots, n_e \\ \frac{\partial \tau_c}{\partial t} &= \underline{S} + \underline{Z}\underline{\mathcal{E}} \end{aligned} \quad (4)$$

where $\underline{\mathcal{E}}_\phi = (\mathcal{E}_{\phi_1}, \mathcal{E}_{\phi_2}, \dots, \mathcal{E}_{\phi_n})^T$ and \underline{S} are rates of production of the reaction progress variables and \underline{Z} is an $n_c \times (n_s + 2)$ projection matrix. The variables \underline{S} and \underline{Z} are functions of the parameters $\underline{\tau}$, which are calculated in the mechanism reduction and tabulated.

Thus, we have equations for $2 + n_e + n_c$ dependent variables $\underline{\tau}$, namely, for the enthalpy, for the pressure, for each of the specific element mole numbers, and for each of the reaction progress variables. The implementation of the reduced mechanism is now very simple: At each point in the flow, the initial values of the dependent variables are known. Then we know the rates \underline{S} of the reaction progress variables and the thermochemical state $\underline{\psi}$ of the system, which are stored in the tabulation of the manifold. From this, we can compute the physical processes $\underline{\mathcal{E}}$ and, using tabulated values for the projection matrix \underline{Z} , calculate the total rates of change of the reduced variables. It shall be noted that the calculation can be further simplified by rewriting the molecular transport terms in terms of gradients of the variables

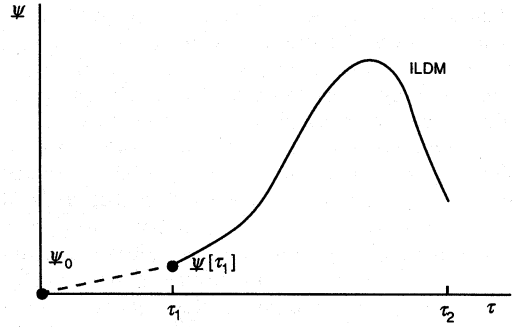


FIG. 1. Intrinsic low-dimensional manifold and its continuation into the domain of negligible chemistry.

of the reduced scheme (this can be done very easily, because the state $\underline{\psi}$ of the system is a known function of the variables of the reduced scheme). In this case, the molecular transport coefficients can be calculated during the mechanism reduction and do not need to be evaluated during the reacting flow calculation [20].

Some complications can arise in the computations due to the fact that initial or boundary values might not correspond to states on the manifolds. A detailed discussion of this problem cannot be given here; however, there are methods to solve it [20]. Here, the basic idea in the case of laminar reacting flows shall be outlined briefly. Suppose that we have a one-dimensional manifold, which is defined in the parameter range $\tau^1 < \tau < \tau^2$ and where the boundary condition is given by ψ_0 (Fig. 1). For $\tau < \tau^1$, chemistry is very slow, and diffusion governs the dynamics of the process. Thus, we have a pure diffusion problem, which is characterized by the two conditions ψ_0 and $\psi[\tau^1]$. From this, it can be seen that the manifold, which is not defined in the interval $\tau < \tau^1$, can be approximated in this region by assuming a linear shape (corresponding to a linear mixing process). In this way, it is possible to define reasonable initial and boundary conditions for the system. If these conditions lie outside the domain of sufficiently fast chemistry, the dynamics are governed by molecular transport, and the overall process can be viewed as a movement into the domain of sufficiently fast chemistry, relaxation toward the manifold, and finally, a movement within the manifold.

Simulation of One-Dimensional Laminar Premixed Flames

Above, it was shown that the reduced kinetics based on intrinsic low-dimensional manifolds can be used to simulate laminar reacting flows. Although reduced mechanisms are of special interest in multi-dimensional problems, simple laminar flat premixed

flames shall be considered here. They provide a nice, simple test case for the verification of the model and are usually more sensitive with respect to the reaction mechanism than diffusion flames. The governing equations form a partial differential equation system with the time and one spatial coordinate as the independent variables. Transformation into a moving coordinate system allows the flame front to be fixed such that a stationary solution is obtained for $t \rightarrow \infty$. The governing equations form an initial-boundary value problem, which can be solved numerically. However, special properties of the equation system (like its stiffness) require special solution methods, and an implicit method, adaptive in space and time, has been chosen: Spatial discretization using finite differences on a statically adapted mesh leads to large systems of ordinary differential equations, which are solved numerically using an implicit extrapolation method with variable order- and stepsize control. Details on this method and references can be found in Refs. 17 and 19.

An existing program for detailed chemistry calculations of one-dimensional instationary flames [17] has been modified such that the tabulated reduced mechanism can be used in the simulation. The additional subroutines are written in a modular way and can be used in other computational fluid dynamics (CFD) codes, too.

Laminar Premixed H_2 - O_2 Flames:

The first example considered here are laminar stationary premixed flat H_2 - O_2 flames. The H_2 - O_2 system consists of ($n_s = 8$) chemical species (H_2O , H_2 , O_2 , H , O , OH , HO_2 , and H_2O_2). Furthermore, there are ($n_e = 2$) elements (H and O). Thus, a 10-dimensional state space, an 8-dimensional composition space, and a 6-dimensional reaction space are obtained. Two reaction progress variables shall be used, which (for simplicity) shall be given by the specific mole numbers of H_2O and H ($\theta_1 = \phi_{H_2O}$, $\theta_2 = \phi_H$). The notation θ is introduced here to denote those variables τ_i , which change in the reaction system. The other variables τ_j , which do not change, can be treated as constants of the system.

Three cases of different complexity are obtained:

1. Equal diffusivities, $Le = 1$. Although this approximation leads to large errors in the H_2 - O_2 system, it shall be considered here, because it is used quite often in the simulation of turbulent flames [18]. Starting from this assumption, the specific enthalpy, as well as the element composition, is constant ($\partial h/\partial t = 0$, $\partial p/\partial t = 0$, and $\partial \chi_i/\partial t = 0$). The system can be described completely by the reaction progress variables, and the scalar field is determined by two partial differential equations for the two variables $\theta_1 = \phi_{H_2O}$ and $\theta_2 = \phi_H$ of the two-dimensional manifold.

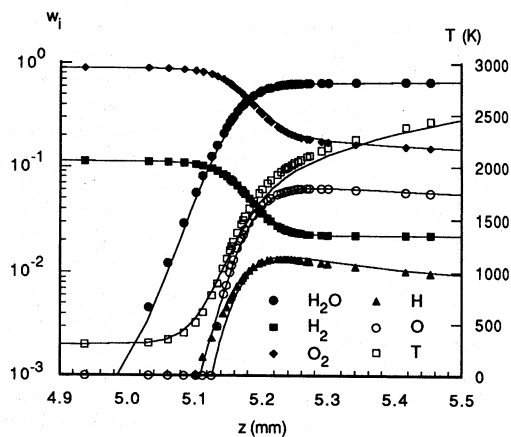


FIG. 2. Calculated structure of a stoichiometric laminar premixed hydrogen-oxygen flame (points: reduced mechanism; lines: detailed reaction mechanism).

2. Equal diffusivities, $Le \neq 1$. In this system the element composition is constant, but the enthalpy changes. In addition to the two equations for the reaction progress variables, a conservation equation for the enthalpy has to be solved ($\partial h/\partial t \neq 0$). The system is described by the three variables $\theta_1 = \phi_{H_2O}$, $\theta_2 = \phi_H$, and $\theta_3 = h$ (corresponding to a three-dimensional manifold).
3. General case. In the general case, differential diffusion changes the element composition. Differential equations are obtained for the specific enthalpy, a specific element mole number (only two elements in the system), and the two reaction progress variables. Thus, the system is governed by the four variables $\theta_1 = \phi_{H_2O}$, $\theta_2 = \phi_H$, $\theta_3 = h$, and $\theta_4 = \chi_H$.

The specific example considered here is a stoichiometric flame at 1 bar and with a temperature of 298 K in the unburnt gas. As an example, Fig. 2 shows the calculated flame structure for equal diffusivities but with a nonunity Lewis number. Good agreement is not only obtained for the stable species but also for the radicals, although only two reaction progress variables are used to describe the whole system. The different transport models are compared in Fig. 3, which shows H-atom profiles for $Le = 1$ as well as for equal diffusivities but with $Le \neq 1$. Plotted is only the small region of the flame front, where the main changes in the profiles occur. It can be seen that the differences between the transport models are larger than the differences between reduced and detailed reaction mechanism. Furthermore, the results of a very detailed transport model (including thermal diffusion) are plotted in the figure. Calculations for this transport model, using the reduced mechanism, can be done too [20]. Other profiles show similar results but are not plotted due to space limitation. Compar-

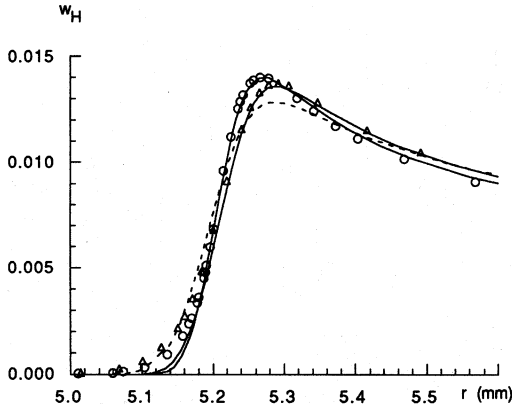


FIG. 3. Calculated profiles of the H-atom mass fractions in a stoichiometric laminar premixed hydrogen-oxygen flame for different transport models. ---: detailed transport model, including thermal diffusion; lines: detailed reaction mechanism; points: reduced mechanism; \circ : $Le = 1$; \triangle : $Le \neq 1$.

isons of the burning velocities show that the error of the reduced scheme is less than 3% for the specific example considered here. A detailed discussion of burning velocities and flame structures for different fuels (e.g., methane) and stoichiometries is beyond the scope of this work and will be given in a future paper [20].

Laminar Premixed $\text{CO-H}_2\text{-O}_2\text{-N}_2$ Flames:

The second example is a syngas (40 vol.% CO , 30 vol.% H_2 , 30 vol.% N_2)-air system at $p = 1$ bar and with a temperature of 290 K in the unburnt gas. The fuel/air ratio is 6/10. The low-dimensional manifold used for this calculation corresponds to the one used in Ref. 9 for homogeneous systems. The fact that the same manifold is used reflects that the method of intrinsic low-dimensional manifolds yields general reduced mechanisms. An adaptation to the specific problem is not necessary. (Furthermore, this reduced mechanism has been used for the simulation of turbulent jet diffusion flames [23], where the mixture fraction was used as an additional tabulation coordinate for the manifold.)

The influence of simplified transport models has been described in the last section. Here, for the sake of simplicity, only systems with equal diffusivity shall be considered, because this section shall mainly discuss the influence of the tabulation on the results and computational aspects. Figure 4 shows profiles of CO_2 , H_2O , OH , and H in the flame both for reduced and detailed kinetics. Good agreement is obtained in the flame front as well as behind the flame front, where the slow equilibration process takes place. Differences are observed at the beginning of the flame

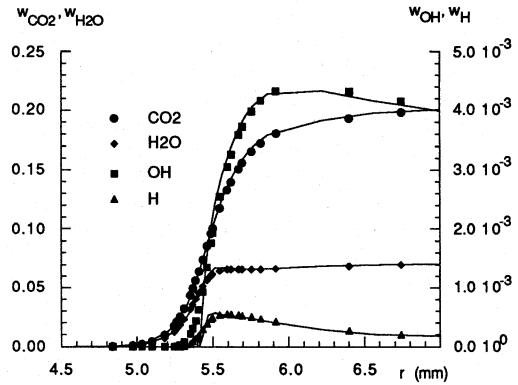


FIG. 4. Calculated structure of a syngas-air flame (points: reduced mechanism, lines: detailed mechanism).

front. This is caused by the fact that at the low temperatures ($T < 1000$ K), the decoupled timescales are of the order of the timescales of diffusion and heat conduction. However, if an additional reaction progress variable were used, the results would become much more accurate. The small deviations behind the flame front can be attributed to discretization errors in the numerical simulation (the points reflect directly the mesh used in the computations).

In order to allow a realistic comparison between central processing unit (CPU) times for reduced and detailed chemistry calculations, we performed computations where the initial profiles were assumed as step functions between burnt and unburnt gas composition. Of course, this is an extremely bad initial guess, and it turns out that very small time steps are needed in the detailed chemistry calculations. Surprisingly, those problems do not arise, if the reduced mechanism is used. Because 16 partial differential equations have to be solved for detailed chemistry and only five if reduced chemistry is used, the reduction of the computational effort can be estimated to be a factor of 6 (due to the small number of equations, there is not yet the quadratic dependence of the CPU time on the number of equations, which is typical for implicit methods). However, the comparison of the CPU time shows that the detailed chemistry calculations need 844 CPU seconds on a Silicon-Graphics Indigo, and the calculations with the reduced scheme only 86 CPU seconds, which corresponds to a speedup of a factor of 10. The main reason for this behavior is that much of the stiffness is removed by the mechanism reduction, and large step sizes are chosen by the integrator. Further research will show whether explicit methods can lead to a further speedup of the computation. It is an interesting and promising result that the mechanism generated by the ILDM method does not cause convergence problems in the numerical simulations, which are usually observed, if reduced mechanisms

are used [1]. The reason for this behavior is mainly that the fast timescales are really decoupled locally, whereas the global conditions of other reduced mechanisms do not guarantee this.

One further aspect, which has to be discussed, is the influence of the tabulation procedure on the accuracy of the results. It is evident that the reduced mechanisms are improved, if more reaction progress variables (degrees of freedom) are used. Nevertheless, tables of higher dimension are only possible if coarse grids are used. Thus, following a suggestion of Gran [22], we performed calculations with tables having different levels of refinement (cf. Ref. 9 for the tabulation procedure). The different tables are shown in Fig. 5, and the results obtained with those grids are shown in Fig. 6 for the OH profiles together with the results of the detailed reaction mechanism (the other profiles show a similar-trend, but they show less significant deviations). Only the coarsest grid leads to large errors, especially in the region of the equilibration zone. But even the use of the second coarsest table, which uses only about 100 kbyte of storage, yields good results. From this, it can be seen that even higher dimensional tables can be produced with a reasonable storage requirement. Better refinement strategies and better interpolation methods can further reduce the storage requirement.

Conclusions

It has been shown that the method of intrinsic low-dimensional manifolds can be used successfully in the simulation of laminar reacting flows. There is in principle no restriction to simplified transport models, but processes like nonunity Lewis number, differential diffusion, or thermal diffusion can be treated by the method very easily. The results are in very good agreement with results from detailed chemistry calculations, which verifies the approach and allows the conclusion that the method can be used in turbulent flame calculations for a reliable description of the chemistry. The computational effort in the laminar flame calculations is reduced considerably. Convergence problems, known from simulations with reduced reaction mechanisms, are not observed, but, to the contrary, the method of intrinsic low-dimensional manifolds yields reduced reaction mechanisms, which simplify the numerical integration to a great extent and might even allow the use of explicit integration methods. It can be estimated that the method can speed up calculations of flames with more complex fuels (like octane) by a factor of much more than 1000.

Acknowledgments

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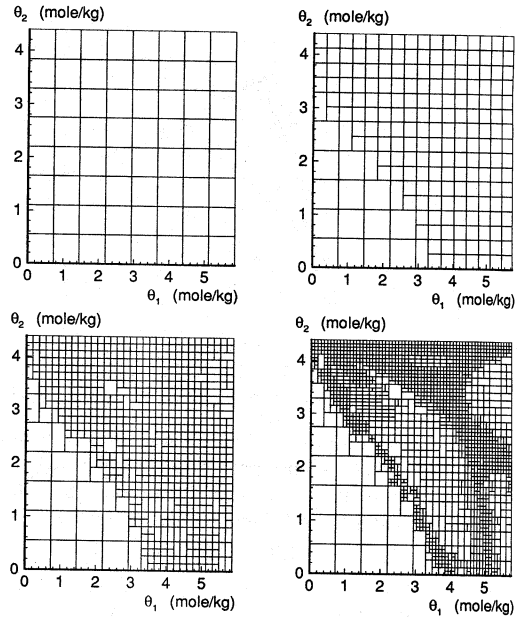


FIG. 5. Structures of the different tables. Upper part: Table 1 (left) and Table 2 (right); lower part: Table 3 (left) and Table 4 (right).

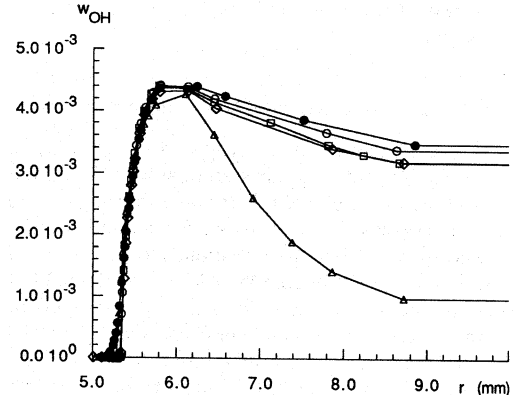


FIG. 6. Comparison of tables with different refinement levels; ●: detailed mechanism; △: Table 1; ◇: Table 2; □: Table 3; ○: Table 4.

Appendix

Within the concept of intrinsic low-dimensional manifolds, a reacting system can be characterized by the $(2 + n_e + n_c)$ parameters $\tau = (h, p, \chi, \tau_c)^T$; i.e., the system is described by its specific enthalpy, pressure, element composition, and n_c reaction progress variables. Then, the state $\underline{\psi}$ is a known function of the parameters

$$\underline{\psi} = \underline{\psi}(\underline{\tau}[\mathbf{r}, t]). \tag{5}$$

Let the parametrization be given by the matrix C,

$$d\underline{\tau} = C d\underline{\psi}. \tag{6}$$

Then, projection of the ($n = n_s + 2$)-dimensional equation system (2) yields [9]

$$\frac{\partial \underline{\tau}}{\partial t} = C \underline{F} + C P \underline{E}. \tag{7}$$

The projection matrix P is given by [9]

$$P = I - V_f \tilde{V}_f \tag{8}$$

where V_f denotes the ($n \times n_f$)-dimensional matrix of the "fast" right eigenvectors and \tilde{V}_f denotes the corresponding ($n_f \times n$) matrix of the left eigenvectors (see Ref. 9).

The parametrization matrix can be split into two components, namely, one describing the specific enthalpy, the pressure, and the element composition and another describing the reaction progress variables. Then we obtain

$$C = \begin{pmatrix} C_e \\ C_c \end{pmatrix} \tag{9}$$

where C_c is the ($n_c \times n$)-dimensional matrix for the reaction progress variables and C_e is the ($n_e + 2 \times n$)-dimensional matrix for the remaining variables, which is given by

$$C_e = \begin{pmatrix} 1 & 0 & 0 & \dots & 0 \\ 0 & 1 & 0 & \dots & 0 \\ 0 & 0 & \text{---} & \underline{\mu}_1^T & \text{---} \\ \vdots & \vdots & & \vdots & \\ 0 & 0 & \text{---} & \underline{\mu}_{n_e}^T & \text{---} \end{pmatrix} \tag{10}$$

where $\underline{\mu}_i = (\mu_{1,i}, \mu_{2,i}, \dots, \mu_{n_s,i})^T$ are the element composition vectors and $\mu_{j,i}$ denotes the number of atoms of element i in a species j . It has the property that it allows the direct calculation of the specific element mole numbers from the state vector:

$$C_e \underline{\psi} = (h, p, \chi_1, \chi_2, \dots, \chi_{n_e})^T. \tag{11}$$

It can be shown that the element vectors are orthogonal to the fast eigenvectors, and we can obtain very easily the relation

$$C_e P = C_e - C_e V_f \tilde{V}_f = C_e. \tag{12}$$

This means that physical processes, which change

specific enthalpy, pressure, or element composition, do not have to be projected. The physical reason for this is that processes, which perturb in direction of eigenvectors with zero eigenvalue (i.e., which change variables conserved in chemical reaction), have no relaxing component.

Using this property, we obtain from Eq. (1) for the scalar field of a reacting flow

$$\begin{aligned} \frac{\partial h}{\partial t} &= \underline{E}_h \\ \frac{\partial p}{\partial t} &= \underline{E}_p \\ \frac{\partial \chi_i}{\partial t} &= \mu_i^T \underline{E}_\phi \quad i = 1, 2, \dots, n_e \\ \frac{\partial \tau_c}{\partial t} &= C_c \underline{F} + C_c P \underline{E} \end{aligned} \tag{13}$$

with $\underline{E}_\phi = (\underline{E}_{\phi_1}, \underline{E}_{\phi_2}, \dots, \underline{E}_{\phi_{n_e}})^T$.

Thus, we have obtained equations for specific enthalpy and pressure, n_e equations for the specific element mole numbers, and n_c for the reaction progress variables. Because $\underline{\psi}$, and thus also \underline{F} , \underline{E} , and P are defined by the low-dimensional manifold in terms of h, p, χ , and τ_c , the equation system can be solved numerically.

REFERENCES

1. Smooke, M. D. (Ed.), "Reduced Kinetic Mechanisms and Asymptotic Approximations for Methane-Air Flames." *Lecture Notes in Physics*, Springer, Berlin, 1991, Vol. 384.
2. Peters, N., and Rogg, B., *Reduced Kinetics Mechanisms for Applications in Combustion Systems*. Springer, Berlin, 1993.
3. Chen, J.-Y., *Combust. Sci. Technol.* 57:89 (1988).
4. Frenklach, M., in *Numerical Approaches to Combustion* (E. S. Oran and J. P. Boris, Eds.), AIAA, Washington, DC, 1991, p. 129.
5. Keck, J. C., *Twenty-Second Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, 1989, p. 1705.
6. Tomlin, A. S., Pilling, M. J., Turanyi, T., Merkin, J. H., and Brindley, J., *Combust. Flame* 91:107 (1992).
7. Oppenheim, A. K., and Maxson, J. A., *Prog. Astron. Aero.* 152:365 (1993).
8. Maas, U., and Pope, S. B., *Combust. Flame* 88:239 (1992).
9. Maas, U., and Pope, S. B., *Twenty-Fourth Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, 1992, p. 103.
10. Warnatz, J., *Twenty-Fourth Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, 1992, p. 553.

11. Smooke, M. D., Mitchell, R. E., and Keyes, D. E., *Combust. Sci. Technol.* 67:85 (1989).
12. Maas, U., and Warnatz, J., *IMPACT Comput. Sci. Eng.* 1(4):394 (1989).
13. Bilger, R. W., in: "Turbulent Flows with Nonpremixed Reactants" P. A. Libby and F. A. Williams, (Hrsg.) Eds., *Turbulent Reactive Flows*, Springer, New York, 1980.
14. Peters, N., *Twenty-first Symposium (International) on Combustion*. The Combustion Institute, Pittsburgh, 1988, p. 1231.
15. Hirschfelder, J. O., and Curtiss, C. F., *Third Symposium on Combustion Flame and Explosion Phenomena*, Williams and Wilkins, Baltimore, 1949.
16. Bird, R. E., Stewart, W. E., and Lightfoot, E. N., *Transport Phenomena*, Wiley Interscience, New York, 1960.
17. Maas, U., and Warnatz, J., *Combust. Flame* 74:53 (1988).
18. Libby, P. A., and Williams, F. A. (ed.), *Turbulent Reactive Flows*, Springer, New York, 1980.
19. Goyal, G., Maas, U., and Warnatz, J., *Proc. 14th ICEDERS*, Coimbra, Portugal, 1993.
20. Schmidt, D., Maas, U., Pope, S. B., and Warnatz, J., in preparation.
21. Lam, S. H., private communication, 1992.
22. Gran, I, private communication, 1993.
23. Norris, A., "The Application of PDF Methods to Turbulent Diffusion Flames." Dissertation, Cornell University, Ithaca, NY, 1993.