

# Generation of reduced models by decoupling of chemical kinetics and convection/diffusion processes

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## Abstract

This work focuses on the construction of low dimensional manifolds representing the slow system dynamics in the state space. The technique is based on decoupling of the chemical and convection/diffusion processes. This idea is widely exploited in combustion theory in both analytical and numerical aspects due to the strongly non-linear and singular structure of combustion models. The method of Intrinsic Low Dimensional Manifolds (ILDM) is used for the decomposition of detailed chemical kinetics and for the approximation of the system dynamics on the intrinsic manifold in the domain of the fast chemistry. In the domain of the slow chemistry the low dimensional manifold of special type is used to approximate the system dynamics. The matching/projecting procedure in this domain can be treated as a projection of the system dynamics in composition space onto a manifold containing an initial point and an intersection of the boundary of the fast chemistry domain with the slow manifold.

Slightly modified to satisfy the general assumption the two-dimensional model (Davis and Skodje 1999) is presented to illustrate the approach. An extension to more complex systems is possible without principle difficulties and it is verified by analysis of premixed singas/air one-dimensional laminar flame structure with detailed chemistry. Preliminary results show that one and two-dimensional reduced models can be successfully applied for approximate the dynamics of the full model with detailed chemistry.

## Introduction

Mathematical models describing complex reactive flow are typically high dimensional systems of high order and complexity their exhibit behavior on widely differing time scales of the involved sub-processes. In combustion processes with detailed chemical kinetics these scales have orders of magnitude from 1 s to  $10^{-10}$  s and the number of species is some hundreds. This disparity in time scales leads to a stiffness of the mathematical models that makes their numerical treatment problematic from the point of view of amount of computational recourses needed for integration. Hence, in order to overcome this difficulty methods for reduction of both dimensionality and stiffness of such systems are frequently used.

At present there are a number of asymptotic or numeric tools able to treat the multi-scale system of equations. An incomplete list includes the computational singular perturbation (CSP) method (Lam and Goussis 1994), the method of integral manifolds (MIM) (Fenichel 1979, Gol'dshtein and Sobolev 1992) and the functional iteration method (Roussel and Fraser 2001). One of the reduction techniques is the ILDM manifolds method (Maas and Pope 1992), which allows exploring the hierarchical structure of the system by a spectral decomposition of the Jacobain field. It gives a good approximation for an invariant manifold of slow motions (up to the second order (Rhodes et al 1999)) when a gap condition of the

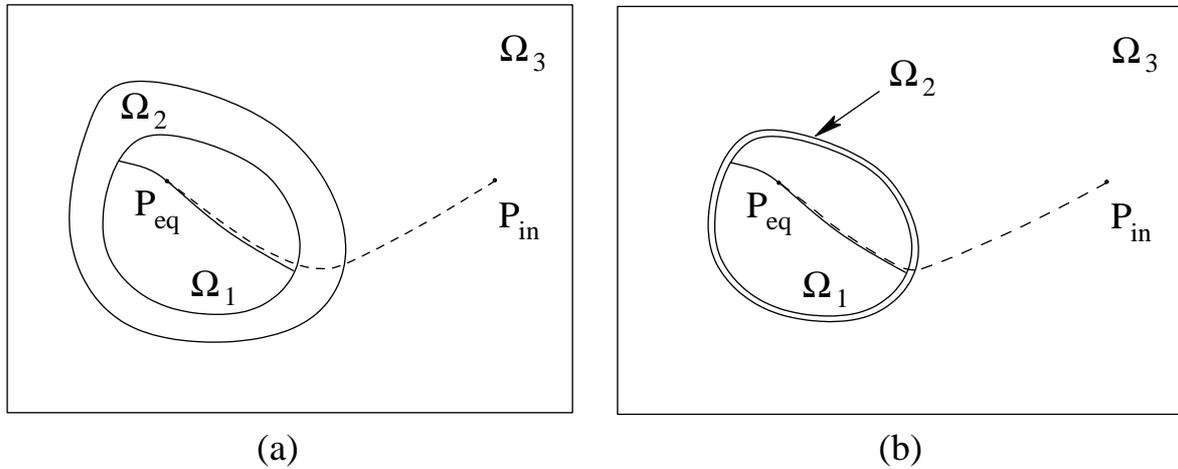
Jacobian eigenvalues is satisfied. For typical combustible mixtures and in most industrial applications dealing with reactive flows this condition is fulfilled and found to be natural.

In order to present mathematical and geometrical essence of the used technique (more detailed description can be found in (Maas 1994)) let us determine a vector  $Z = (Z_1, \dots, Z_n)$ , which defines the thermo-chemical state of the system in vector space, here  $Z_i$  represents such thermodynamic quantities as the pressure of mixture, the enthalpy, the mass fraction of chemical species etc. In these vector notations a system of governing equations for reacting flow can be written as

$$\frac{\partial Z}{\partial t} = F(Z) + G(Z, \nabla Z, \nabla^2 Z), \quad (1)$$

where the first term related to chemical kinetics and the second one describes convection and diffusion processes. Now, we outline general assumptions concerning properties of the considered model related to the reaction flows.

First, we assume that the domain of interest in the state or phase space can be separated into three different sub-domains such that in the first one  $\Omega_1$  (see sketch on Fig. 1(a)) the chemical kinetics governs the system dynamics, which means that the fast chemical processes always equilibrate towards some low dimensional manifold which contains the slow system dynamics and interaction of the slow sub-processes with convection diffusion ones. In the second domain  $\Omega_2$  the chemical and convection/diffusion processes are strongly coupled or in other words have the same order of magnitude. The third one  $\Omega_3$  is the domain of very (infinitely) slow chemistry where the chemical source term is negligible and the system dynamics is governed by convection/diffusion only.



**Figure 1.** Sketch of the phase space with asymptotical domains and. ILDM manifolds is sketched by solid lines, stationary system solutions – dashed lines.

The second assumption already mentioned above is the spectral gap condition (2). That is, for each point in the domain of strong chemistry  $\Omega_1$  the eigenvalues of the Jacobian are separated into two groups and the following inequalities are valid:

$$\begin{aligned}
& \text{(i): } \operatorname{Re}(\lambda_i(\mathbf{F}_Z)) \leq 0; \quad i = 1, \dots, n_f \\
& \text{(ii): } |\operatorname{Re}(\lambda_i(\mathbf{F}_Z))| \leq a \ll b \leq |\operatorname{Re}(\lambda_j(\mathbf{F}_Z))|; \quad a, b \geq 0. \\
& i = 1, \dots, n_s; \quad j = 1, \dots, n_f; \quad n_s + n_f = n
\end{aligned} \tag{2}$$

where  $\mathbf{F}_Z$  is the Jacobian of the chemical term. If this condition is satisfied then the locally invariant subspaces related to these eigenvalues are used to find corresponding projection operators:  $\tilde{\mathbf{Q}}_f, \tilde{\mathbf{Q}}_s$ . These projectors define both the system decomposition and reduced system dynamics. According to the ILDM method a manifold containing slow system dynamics is given as the manifold that annihilates the sub-processes spanned into the fast subspace:

$$\tilde{\mathbf{Q}}_f \cdot \mathbf{F} = 0. \tag{3}$$

Consequently, when the system trajectory reaches the first domain the system trajectory relaxes fast towards the slow manifold (3) and slow processes which are coupled with the physical processes govern the system dynamics.

### Simplified projection strategy

Unfortunately, only in pure homogeneous reactors and in some simple problems the whole system dynamics is contained only in the first asymptotical domain. Commonly, the system trajectory in the state space links the equilibrium point, which always belongs to the ILDM manifold as well as to the slow invariant manifold, with the initial point (typically the initial point corresponds to a given boundary condition) that can be located in any of three defined domains (see Fig. 1). Therefore, we have three different situations depending on the location of the initial point. Fortunately, in many practical models the second domain asymptotically shrinks into the boundary between first and third domains (see Fig. 1(b)) and can be ignored without essential loss of accuracy. This might be considered like an additional assumption, but in real combustible mixtures it is fulfilled automatically due to strong dependence of the chemical reaction term on system quantities (strongly non-linear dependence).

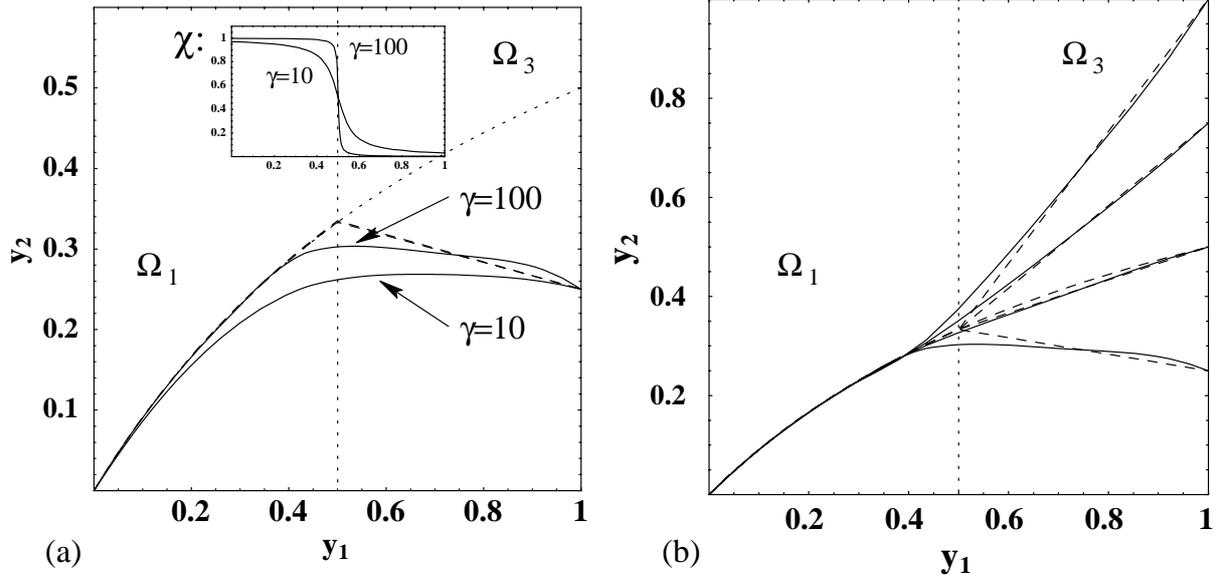
The basic idea is now to exploit this behavior of the different domains and to construct a matching procedure between the different domains. To illustrate the idea let us analyze a test case similar to the well-studied by Davis and Skodje (Davis and Skodje 1999). Two modifications are applied: the addition of a diffusion term and the assumption that the chemical source term becomes negligible in part of the composition space. The system of governing equations is

$$\begin{aligned}
\frac{\partial y_1}{\partial t} &= F_1(y_1, y_2) + \varepsilon \frac{\partial^2 y_1}{\partial x^2} \\
\frac{\partial y_2}{\partial t} &= F_2(y_1, y_2) + \varepsilon \frac{\partial^2 y_2}{\partial x^2}
\end{aligned} \tag{4}$$

where chemical term is multiplied by  $\chi(y_1) = \arctan(-(1-2y_1)/\gamma)/\pi + 1/2$  which is in fact a truncation function and used here to model/satisfy the general assumption of shrinking the second asymptotical domain

$$F_1(y_1, y_2) = -y_1 \chi(y_1), \quad F_2(y_1, y_2) = \left( -\gamma y_2 + \frac{(\gamma-1)y_1 + \gamma y_1^2}{(1+y_1)^2} \right) \chi(y_1),$$

here both  $\varepsilon$  and  $1/\gamma$  are small system parameters. The system (4) is considered with the following initial and boundary conditions:  $y_1(0, x) = \alpha x$ ,  $y_2(0, x) = \beta x$ ,  $y_1(t, 0) = 0$ ,  $y_2(t, 0) = 0$ ,  $y_1(t, 1) = \alpha$ ,  $y_2(t, 1) = \beta$ . Initial conditions are chosen to be straight lines, they satisfy the general assumption – join initial and equilibrium values on boundaries.



**Figure 2.** (a) - Extended ILDM curve – dashed line, stationary solutions – solid lines for  $\gamma = 10, 100$ ,  $\alpha = 1; \beta = 0.25$ ,  $\varepsilon = 0.1$  and original ILDM curve – dotted line; (b) - extended ILDM curves – dashed lines, full system stationary solution – solid lines for  $\gamma = 100$ ,  $\alpha = 1$ ,  $\beta = 0.25, 0.5, 0.75, 1$  and truncation boundary – dotted line.

Let us start from analysis of the modified reaction term. First of all, in this model system both eigenvalues of the Jacobian of the chemical reaction term have constant values:  $\lambda_f = -\gamma$  and  $\lambda_s = -1$  so that a gap condition is automatically satisfied and the ILDM curve has a simple analytical form:

$$y_2 = \frac{y_1}{1+y_1} + \frac{2y_1^2}{\gamma(\gamma-1)(1-y_1)^3}. \quad (5)$$

In Figure 2(a) this curve is shown by dotted line. The boundary of domain of the fast chemical process is asymptotically presented by curve:  $y_1 - 1/2 = 0$  (see definition of the function  $\chi$ ). One can see that if the value of the parameter  $\gamma$  increases then along this line the phase plane will be sharply subdivided into the fast and no chemistry domains.

It is evident that the system solution in stationary limit and without chemical term has a linear structure because of resulting equation is pure linear. Moreover, any linear dependence of the variables in phase/composition space is conserved in this domain. Additionally, the presence of the asymptotically large parameter  $\gamma$  gives a decomposition of the system dynamics into fast and slow and the slow manifold may be used to reduce the dimension in the fast chemistry domain. Hence, in the considered example the one-dimensional invariant system manifold in phase space may be approximated by straight line in the no chemistry domain and by ILDM manifold in the fast chemistry domain.

The problem of low dimensional manifold construction is reduced to finding the boundary between the domains. Let us assume that the boundary is defined then the natural way to match the different asymptotical manifolds is join the ends of these curves at the boundary. In other words, the intersection of this boundary with the ILDM curve yields the point, where the curves are matched. A basis for this matching is a continuity assumption of the reduced manifold. It has to be continuous at stationary limit, which is of interest of many applications, as an approximation of the system solution.

As an illustration of the proposed approach the results of the integration are presented in Figure 2 (b). It is not hard to see that with increasing the large system parameter  $\gamma$  the system solution follows the constructed manifold which is consisting of two parts: pure linear curve in the no chemistry domain and the ILDM curve in the fast chemistry domain. Figure 2 shows that the full system solutions and the reduced solutions obtained by matching the different domains are in a good agreement as far as the large asymptotic system parameter goes to infinity. In addition, the proposed strategy of matching the reduced dynamics in both asymptotical domains to produce low dimensional manifolds approximating the system dynamics can be applied in high dimensional cases too.

To generalize the idea let us consider the complex situation when it is needed to increase the accuracy of the reduced model and therefore one has to add an extra dimension to the slow manifold in the fast chemistry domain. It may happen also that in the case when the boundary manifold separating the dynamics is not negligible then one has to increase the dimension in order to improve the situation on this boundary domain. Of course, the system dimension is no longer limited by two and the system might be of any dimension. In this case the same one-dimensional structure is preserved in the no chemistry domain -  $\Omega_2$ , but the slow manifold in the fast chemistry domain has its increased dimension. In fact this case does not differ much from the mentioned above. In particular, intersection of the  $m$ -dimensional slow manifold with the boundary becomes to be  $(m-1)$ -dimensional manifold boundary manifold, but the system dynamics outside of the fast chemistry domain is still having a linear structure. Therefore, to extend the  $m$ -dimensional slow manifold onto the no chemistry domain one can take a manifold consisting of lines and joining the boundary manifold with the initial point. This will allow the reduced system to approach the system dynamic more accurately near the boundary manifold.

It should be mentioned here that in the case of strong interaction of some chemical modes with convection/diffusion processes close to the boundary domain that can be considered as boundary manifold, it is possible to reduce the problem to current formulation by increasing the dimension of the slow subsystem or by applying refinement procedure to the ILDM manifold with additional terms that takes into account the interaction.

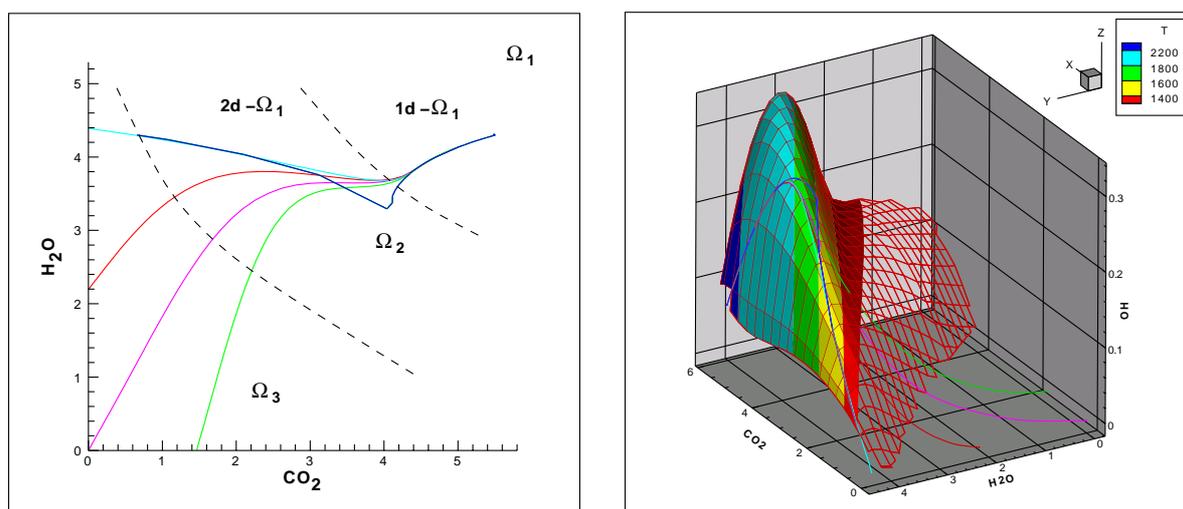
Finally, in complex systems the behavior might be complicated by the fact that there exist multiple timescales and the domains of fast and slow chemistry may have different dimension. Nevertheless, the method presented above can also be applied to these more complex systems, if the points of intersection are generalized to hyper-surfaces of

intersection, and if “linearity” in the domain of slow chemistry is generalized to “geodesic surfaces on non-linear hyper-surfaces”.

### Application to syngas/air premixed laminar flame

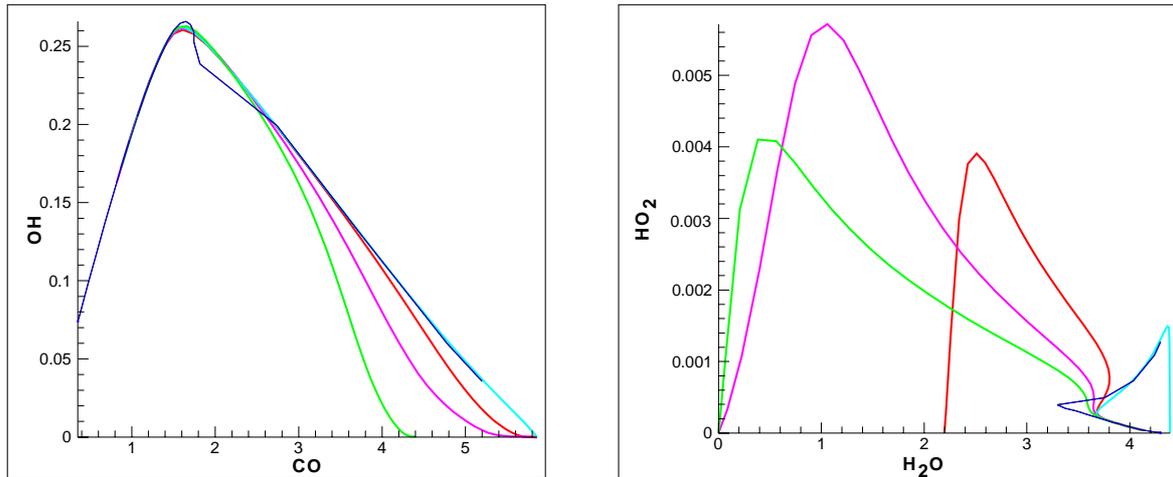
To verify the approach let us show the results of one-dimensional premixed adiabatic free syngas/air flame with detailed chemical mechanism. In order to simplify the presentation, the main physical assumptions are as the following: constant pressure, equal diffusivity, Lewis number equals to unity.

On the figures below, the one-dimensional ILDM manifold is shown together with the stationary solutions for different boundary conditions. In Figure 3 (left) phase space is projected onto  $\text{CO}_2 - \text{H}_2\text{O}$  plane. These variables are often used to parameterize the ILDM manifold and are relatively slow ones. As one can see the projected part of the phase plane might be roughly subdivided according to the suggested line. Near the boundary part all the system trajectories are follow the straight lines, then near the boundary manifold they start to deviate in both directions - the system trajectory leaves the one-dimensional ILDM manifold and the system trajectory is not longer a straight line from the other hand. Since the chosen variables are relatively slow their modes are strongly coupled with convection-diffusion processes in the boundary manifold which becomes to be no longer negligible.



**Figure 3.** Projections of the state space onto  $\text{CO}_2 - \text{H}_2\text{O}$  plane and  $\text{CO}_2 - \text{H}_2\text{O} - \text{OH}$  space. Color lines are stationary solutions, blue line is the one-dimensional ILDM manifold, and red mesh represents the two-dimensional ILDM manifold.

It does not shrink to the boundary, therefore, in this case the use of two-dimensional manifold may improve the accuracy of the reduced model, and it can be extended to the new level. On the right figure the projection onto 3-dimensional space is given, here two-dimensional ILDM manifold is shown with color contour lines related to different mixture temperatures. To indicate the boundary contour lines are truncated at the temperature equals to  $1300^\circ\text{K}$ . This example clearly shows how the additional dimension improves the low dimensional manifold. Figure 4 provides further qualitative information about the structure of the system stationary solution for some minor species.



**Figure 4.** Projections of the state space onto different planes of minor species. Color lines are stationary solutions starting at different initial points and having the same element composition and enthalpy. Blue line is the one-dimensional ILDM manifold.

Although on the left figure the one-dimensional structure looks as a reasonable approximation on the right one the use of one-dimensional manifold gives very rough approximation for molar fraction of  $\text{HO}_2$ . Nevertheless, all the stationary trajectories at initial stage are linear and near the maximum their approaches the higher dimensional slow manifold which can be successfully approximated by two-dimensional ILDM manifold. Hence the questions of definition of the boundary manifold as well as determination of the minimal dimension of the slow manifold in the fast chemistry domain is crucial point of the suggested method and must be investigated before generating the matched manifold. Complete answers on these questions can not be given in general case; they strictly depend on local property of considered model and on accepted level of reduced model accuracy. Yet the results of many applications of the ILDM strategy in CFD modeling and simulations show that application of two-dimensional manifolds only produce rather accurate solutions and are in a good agreement qualitatively with detailed calculations.

## Conclusions

A method for constructing of the approximation for the manifold of slow motions has been discussed. It is based on natural assumptions widely used in combustion theory. Namely, it is assumed that there exists a sharp subdivision (splitting) of the state space into sub-domains with predomination of chemical kinetics or convection/diffusion terms. Accordingly, in the domain of fast chemistry we use the ILDM method to approximate invariant manifold of slow motions whereas in no chemistry or sometimes called as low temperature domain some projection and matching procedure on the ILDM manifold can be used to describe the system dynamics. In this domain the choice of the manifold (curve, hyper-surface etc.) with the linear structure (consisting of lines) is motivated by the fact that in the limiting case, when chemical term is neglected, the system is pure linear and therefore its dimension can be reduced automatically to one dimension. Fortunately, this property may be preserved in more complicated problems with additional conditions made. The two-dimensional model example

has been used with some additional diffusion and truncated condition terms, which is chosen to illustrate and verify of the proposed approach. Simple calculations show a good asymptotical agreement of the constructed manifold with full system solution. In addition the one-dimensional premixed adiabatic syngas/air free flame with detailed chemical mechanism was used to verify the proposed approach on real combustion problem. Preliminary analysis of the full model shows a potential for suggested method.

### **Acknowledgments**

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### **References**

Davis, M. J., and Skodje, R. T., 1999, Geometrical Investigation of Low-Dimensional Manifolds in Systems Approaching Equilibri, *Journal of Chemical Physics*, Vol. 111, pp. 859-874.

Fenichel, N., 1979, Geometric singular perturbation theory for ordinary differential equations, *J Differential Equations*, Vol. 31, pp. 53-98.

Gol'dshtein, V. and Sobolev, V., 1992, Singularity Theory and Some Problems of Functional Analysis, *AMS Translations*, Series 2, Vol. 153, pp. 73-92.

Lam, S.H. and Goussis, D.M., 1994, The GSP method for simplifying kinetics, *International Journal of Chemical Kinetics*, Vol. 26, pp. 461-486.

Maas, U., and Pope, S.B., 1992, Simplifying Chemical Kinetics: Low-Dimensional Manifolds in Combustion Code, *Combustion and Flame*, Vol. 88, pp. 239-264.

Maas, U., 1994, Automatische Rduktion von Reaktions mechanismen zur Zimulation reaktiver snomungen, *Hasilitation Thesis*, Universitat Stuttgart.

Rhodes, C., Morari, M., Wiggins, S., 1999, Identification of the Low Order Manifolds: Validating the Algorithm of Maas and Pope, *Chaos*, Vol. 9(1), pp. 108-123.

Roussel, M.R. and Fraser, S.J., 2001, Invariant Manifold Methods for Methabolic Model Reduction, *Chaos*, Vol 11(1), pp. 196-206.