



# Hierarchical generation of ILDMs of higher hydrocarbons

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## Abstract

Combustion mechanisms of higher hydrocarbons are governed by a hierarchical structure. This structure is reflected in reduced mechanisms based on intrinsic low dimensional manifolds (ILDM). Based on the mathematical analysis we present in this work a hierarchical concept where the ILDMs of higher hydrocarbons are generated on the basis of existing ILDMs of simpler reaction mechanisms. It is based on a numerical method which allows to estimate, how well an ILDM can be represented by an ILDM of a simpler subsystem. If the conditions for a good representation of the ILDM by the simpler subsystem are met, then the ILDM of the simpler system can be used directly. If not, then the ILDM of the simpler subsystem is used as a starting estimate for the generation of the ILDM which reduces the computational effort considerably. The example of a syngas ILDM which is used as a generic ILDM for n-heptane combustion validates the approach. © 2003 The Combustion Institute. All rights reserved.

*Keywords:* Reduced kinetics; Mechanism reduction; Combustion chemistry

## 1. Introduction

Detailed simulations of technical combustion systems (governed by 3D turbulent flows, complex geometries and complex fuels) are still computational prohibitive. In particular, due to the large range of chemical time scales ( $10^{-10}$  to  $10^0$  s) and the large number of chemical species, the description of the evolution of the chemical kinetics leads to very stiff and high-dimensional equation systems. To overcome this problem several methods have been devised for the simplification of detailed chemical reaction mechanisms (see e.g., [1–7]) reducing both, the stiffness and the dimension of the governing equations.

In this work the method of intrinsic low-dimensional manifolds (ILDM) [2,8] is used for the auto-

matic generation of reduced mechanisms. This method performs, locally in the state space, an analysis of the time scales of the detailed reaction mechanism. It is possible to separate the chemical processes of the system into slow and fast processes. The fast ones are, due to fast relaxation processes, assumed to be in steady states and can, therefore, be decoupled from the reaction system. Thus, the thermochemical state is parameterized by a small number of reaction progress variables (typically 2–4), only, which describe the slow movement along the low-dimensional manifold. The obtained ILDM-reduced kinetic schemes can be implemented in laminar [3,9,10] and turbulent [11–14] reacting flows.

For technical applications, reduced mechanisms of higher hydrocarbons (e.g., n-heptane, dodecane) are of increasing interest. Like reduced schemes for syngas or methane combustion these mechanisms can be generated by the use of the ILDM method [1]. Although ILDMs of higher hydrocarbons can be calculated in a straight-forward way, one major problem is the computational effort, which increases with the

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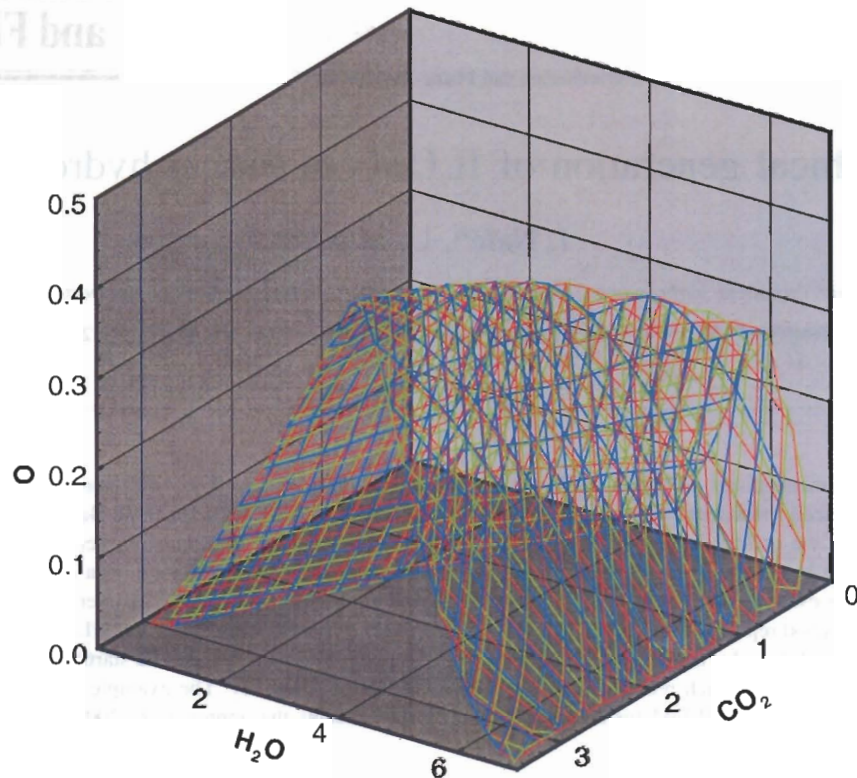


Fig. 1. 2-dimensional ILDMs for a syngas- (red mesh), a methane- (green mesh) and an n-heptane-air-mixture generated for a specific enthalpy of  $h = -25.69 \cdot 10^4 \text{ J/kg}$ , a pressure of  $p = 1 \text{ bar}$  and the molar element ratios  $C/H = 0.250$ ,  $C/O = 0.250$ , and  $C/N = 0.133$ . They are plotted as a projection of the state space onto  $\text{CO}_2$ - $\text{H}_2\text{O}$ - $\text{O}$  coordinates. Species concentrations are in units of  $[\omega/M_i] = \text{mol/kg}$ .

3<sup>rd</sup> power of the number of species (particularly caused by the eigenvalue decomposition). This paper presents a solution to overcome this drawback by the use of a hierarchical concept.

It is a well known fact that the combustion chemistry of hydrocarbons is characterized by a hierarchical structure [15, 16]. The higher hydrocarbons decay fast by H-atom abstraction and  $\beta$ -decomposition. The resulting radicals  $\text{CH}_3$  and  $\text{C}_2\text{H}_5$  are oxidized slowly to the combustion products [16]. It has been shown recently that this hierarchical structure of the detailed reaction mechanisms is reflected in the ILDMs [1]. To visualize the hierarchical structure of ILDMs, Fig. 1 shows as an example three 2-dimensional ILDMs generated for a syngas- (red mesh), a methane- (green mesh), and an n-heptane-air-mixture (blue mesh). They are presented in a projection of the state space into the coordinates  $\text{CO}_2$ - $\text{H}_2\text{O}$ - $\text{O}$ . All three ILDMs are calculated for the same specific enthalpy, pressure and molar element ratios. It can be seen clearly, that the presented ILDMs are very similar. This fact can

be exploited for an efficient calculation of ILDMs of higher hydrocarbons.

In [1] a mathematical analysis has been presented, which gave the conditions for the equivalence of ILDMs of an original and an augmented reaction system. However, a numerical methodology to estimate how well the conditions are met and its application to a specific example for validation was missing. Such a numerical methodology is presented in this paper.

Based on an ILDM of an original system it provides an estimation whether the ILDM of an augmented system can be represented by the ILDM of the simpler system. If the ILDM of the augmented system can be described with a sufficient accuracy by an ILDM of a simpler (generic) system then the new ILDM is directly constructed based on the simple ILDM. If not, then the new ILDM is computed with the generic ILDM as starting estimate.

As an example the  $\text{CO}/\text{H}_2$  ILDM is used as a generic ILDM for n- $\text{C}_7\text{H}_{16}$  combustion. ILDMs ob-

tained using the strategy presented in this paper are compared with conventionally calculated ILDMs and results using detailed chemistry.

## 2. Mathematical model

In the following we shall describe the strategy for the application of the hierarchical concept outlined above. It is based on an analysis of two detailed reaction mechanisms, namely one for the CO/H<sub>2</sub> or the C<sub>1</sub>–C<sub>2</sub> subsystem, and one for a higher hydrocarbon, where it is assumed that the detailed mechanisms exhibit a hierarchical structure. This means that the higher hydrocarbon mechanism includes the mechanism of the small subsystem plus additional fuel specific chemical species and reactions. The analysis is performed for each point of an already existing ILDM of a smaller system. The result of this procedure yields information whether the investigated ILDM is dependent or independent of the used fuel.

More specifically, this analysis allows to decide

- whether the ILDM of the small subsystem can be used as a generic ILDM for the large system or
- whether the ILDM of the small system can at least be used as a starting estimate for the generation of an ILDM for the higher hydrocarbon.

Let the kinetics of the small chemical subsystem (e.g., syngas combustion) be governed by the equation system

$$\frac{\partial \psi}{\partial t} = F(\psi) \quad (1)$$

where  $\psi = (h, p, w_i/M_i)^T$  denotes the state vector (with the enthalpy  $h$ , the pressure  $p$ , the species mass fractions  $w_i$  and the molar masses  $M_i$ ),  $F(\psi)$  is the vector of the chemical source terms and  $t$  is the time. Now an additional number of  $n_s^a$  chemical species and  $n_r^a$  reactions shall be added to obtain the reaction scheme of a higher hydrocarbon (e.g., n-heptane combustion). The augmented system is then given by [1]

$$\begin{aligned} \frac{\partial \psi}{\partial t} &= F(\psi) + G(\psi, \eta) \\ \frac{\partial \eta}{\partial t} &= H(\psi, \eta) \end{aligned} \quad (2)$$

with the Jacobian  $J$  of the coupled system

$$J(\psi, \eta) = \begin{pmatrix} F_\psi + G_\psi & G_\eta \\ H_\psi & H_\eta \end{pmatrix}. \quad (3)$$

The  $n_s^a$ -dimensional vector  $\eta$  comprises the additional species concentrations,  $H(\psi, \eta)$  denotes the source terms of the species  $\eta$  and  $G(\psi, \eta)$  represents the change of the source terms  $F(\psi)$  of the original system due to the added chemical processes. An analysis of the augmented reaction system and the criteria which are necessary for a hierarchical structure of the ILDMs have been presented in [1]. These conditions are given by

$$H(\psi, 0) = 0 \quad G(\psi, 0) = 0 \quad (4)$$

$$H_\psi(\psi, 0) = 0 \quad G_\psi(\psi, 0) = 0 \quad (5)$$

$$\lambda_s^a(H_\eta) < \min(\lambda_r^s(F_\psi), 0) \quad (6)$$

where  $\lambda_s^a(H_\eta)$  denotes the eigenvalue corresponding to the slowest time scale of the additional processes,  $\lambda_r^s(F_\psi)$  represents the eigenvalue corresponding to the fastest time scale of the slow processes, i.e., those not decoupled from the underlying smaller system during the generation of the ILDM of the subsystem represented by  $\psi$ . (Note that all eigenvalues in (6) are assumed to have negative real parts, that the eigenvalue largest in magnitude corresponds to the fastest chemical process, and furthermore that the introduction of positive eigenvalues automatically violates condition, Eq. 6.) If Eqs. (4) to (6) are fulfilled, the ILDM  $M^a$  of the augmented system will be simply defined by

$$M^a = \{(\psi, \eta) | \psi \in M^o, \eta = 0\} \quad (7)$$

where the manifold  $M^o$  denotes the ILDM of the small subsystem. Eq. (7)-states that the manifold  $M^a$  of the augmented system is the same as  $M^o$  embedded in the  $(n + n_s^a)$ -dimensional state space instead of the  $n$ -dimensional one of the simpler system.

Let us start with an analysis of Eq. (4). [Note that  $G(\psi, 0) = 0$  is automatically fulfilled, if we add only elementary reactions to the small subsystem which involve at least one new chemical species  $\eta$  as a reactand.] To investigate how well Eq. (4) is fulfilled, we analyse the Euclidian norm of  $H(\psi, 0)$  and  $G(\psi, 0)$  which is for an  $l$ -dimensional vector  $x$  defined by [17]:

$$\|x\| = \sqrt{\sum_{i=1}^l x_i^2}. \quad (8)$$

A suitable measure for the influence of the vectors  $G$  and  $H$  is given by the ratio of their norms with respect to the norm of the vector  $F$  of the chemical source terms of the small system:

$$\frac{\|H(\psi, 0)\|}{\|F(\psi)\|} < \epsilon_1 \quad \frac{\|G(\psi, 0)\|}{\|F(\psi)\|} < \epsilon_2 \quad (9)$$

where  $\epsilon_1$  and  $\epsilon_2$  denote error limits (see Eq. 9) To obtain a good accuracy. For the example given below, note that because a production of higher hydrocarbons from small fragments is not included in the detailed reaction mechanism, the conditions  $\mathbf{G}(\psi, 0) = 0$  and  $\mathbf{H}(\psi, 0) = 0$  are met automatically within a tolerance given by the numerical inaccuracies in the evaluation of the source terms  $\mathbf{G}$  and  $\mathbf{H}$ . Therefore,  $\epsilon_1$  and  $\epsilon_2$  are set to  $10^{-6}$  for the example given below.

In a similar way, we estimate how well Eq. (5) is fulfilled using the ratio of the norms of the matrices  $\mathbf{G}_\psi(\psi, 0)$  and  $\mathbf{H}_\psi(\psi, 0)$  to the norm of the Jacobian  $\mathbf{F}_\psi(\psi)$  of the original system (analogically to the criteria defined in Eq. (9)). The matrix norm used is the Frobenius norm [17] which for a  $(k \times l)$ -dimensional matrix  $\mathbf{A}$  is given by

$$\|\mathbf{A}\| = \sqrt{\sum_{i=1}^k \sum_{j=1}^l a_{ij}^2} \quad (10)$$

The resulting criteria for the conditions defined in Eq. (5) are then given by

$$\frac{\|\mathbf{H}_\psi(\psi, 0)\|}{\|\mathbf{F}_\psi(\psi)\|} < \epsilon_3 \quad \frac{\|\mathbf{G}_\psi(\psi, 0)\|}{\|\mathbf{F}_\psi(\psi)\|} < \epsilon_4 \quad (11)$$

where  $\epsilon_3$  and  $\epsilon_4$  are again error limits which are set to  $10^{-4}$  for the example given below.

Another useful analysis of the perturbation of  $\mathbf{F}_\psi$  by the added system is based on an investigation of the influence of the perturbation matrix  $\mathbf{G}_\psi$  on the “fast” eigenvalues of the Jacobian  $\mathbf{F}_\psi$  of the original system. In order to estimate the perturbation, the vector  $\Lambda_{\mathbf{F}_\psi}$  containing the eigenvalues of the Jacobian  $\mathbf{F}_\psi$  and the vector  $\Lambda_{(\mathbf{F}_\psi + \mathbf{G}_\psi)}$  containing the eigenvalues of the matrix  $(\mathbf{F}_\psi + \mathbf{G}_\psi)$  are computed. The eigenvalues of these vectors are ordered according to decreasing real parts. Differences between the two vectors are determined as follows

$$\sqrt{\sum_{i=n_s+1}^n \left( \frac{\Lambda_{\mathbf{F}_\psi}^i - \Lambda_{(\mathbf{F}_\psi + \mathbf{G}_\psi)}^i}{\max(|\Lambda_{\mathbf{F}_\psi}^i|, |\Lambda_{(\mathbf{F}_\psi + \mathbf{G}_\psi)}^i|)} \right)^2} = \|\Lambda\| < \epsilon_5 \quad (12)$$

where  $n$  is the number of species of the augmented system,  $n_s$  is the number of slow processes and  $\epsilon_5 = 10^{-2}$  is used as error limit for the example given below (note that only the differences of the “fast” eigenvalues are accounted for).

According to Eq. (6), the time scales, introduced by the additional chemical species and reactions, have to be smaller than the slow time scales of the original system given in Eq. 1 [1]. This condition refers to eq. (6) and leads to the following criterion

$$\frac{\lambda_s^a(\mathbf{H}_\eta)}{\lambda_f^s(\mathbf{F}_\psi)} > 1. \quad (13)$$

If all criteria are fulfilled, then the ILDM of the small system is a generic representation of the ILDM of the large system. Thus, the ILDM of the smaller hydrocarbon can directly be used for the higher hydrocarbon using Eq. (7). For the case that one or more assumptions are not met for a point on the ILDM, this point can nevertheless be calculated using the value  $(\psi, 0)$  as a starting guess. This is still an advantage compared to the conventional generation of ILDMs. Because, it is possible to use the ILDM point of the smaller hydrocarbon as an initial guess, this leads to a significant reduction of the computational effort.

The mathematical model presented above has been developed assuming that the added chemical processes mainly introduce time scales which are faster than the ones of the reaction progress variables of the underlying system. If one adds a reaction system containing slow chemical time scales (e.g., NO-formation), these processes can very often be decoupled from the reaction system, too. An analysis of such systems is presented in [18].

### 3. Comparison of CO/H<sub>2</sub>- and n-C<sub>7</sub>H<sub>16</sub>-ILDMs

In this section, the analysis presented above is performed for n-heptane combustion, on the basis of the syngas system being the small subsystem. Two- and three-dimensional ILDMs have been calculated for a specific enthalpy of  $h = -25.69 \cdot 10^4$  J/kg, a pressure of  $p = 1$  bar and the molar element ratios  $C/H = 0.250$ ,  $C/O = 0.250$ , and  $C/N = 0.133$  (note that the combustion products CO<sub>2</sub> and H<sub>2</sub>O are added to the n-heptane-air-mixture to guarantee the same element composition). The detailed syngas mechanism includes 13 chemical species and 72 reactions and the n-heptane mechanism consists of 68 chemical species reacting in 799 reactions [16]. In addition for the evaluation of the quality of the reduced mechanisms a laminar flat flame using detailed n-heptane chemistry is calculated. The computation is performed assuming Lewis number  $Le = 1$  and equal diffusivities, although it has been shown [3] that this is not a necessary assumption in the context of ILDM.

The analysis procedure is realized in several steps. At first the ILDM of the smaller system, in this example the syngas system, is calculated. Then the additional  $68 - 13 = 55$  species and  $799 - 72 = 727$  reactions are added. The concentrations of the new

species are set to zero, according to Eq. (7). Now the criteria defined in the mathematical model are checked for each grid point of the underlying ILDM.

Figure 2 presents the results obtained from the analysis procedure for the 2-dimensional ILDM. The ILDM is plotted in a projection of the state space into the coordinates  $\text{CO}_2\text{-H}_2\text{O}$  and the results of the analysis of the 3-dimensional ILDM, the analysis is performed for points along a flame trajectory (see Fig. 3). The criteria are plotted against the coordinate  $\phi_{\text{CO}_2} = w_{\text{CO}_2}/M_{\text{CO}_2}$ , which can be regarded as a reaction progress variable. It can be seen that the norms  $\|\mathbf{H}(\psi,0)\|$ ,  $\|\mathbf{G}(\psi,0)\|$ ,  $\|\mathbf{H}\psi(\psi,0)\|$ ,  $\|\mathbf{G}\psi(\psi,0)\|$  and  $2\|\mathbf{A}\|$  are, again very small. The ratio  $\lambda_s^a(\mathbf{H}_\eta)/\lambda_r^s(\mathbf{F}_\psi)$  is smaller than in the case of the 2-dimensional ILDM, but, is still greater than one in the accessed domain. Again, this example shows that all criteria are fulfilled and, thus, this reduced mechanism is not fuel specific, either.

In summary, it can be concluded that 2-dimensional ILDMs and 3-dimensional ILDMs for the example considered here are, for a given enthalpy, pressure and element composition, independent of the used fuel. Similar results are obtained for methane and dodecane combustion (not shown here). It can also be seen that the ratio  $\lambda_s^a(\mathbf{H}_\eta)/\lambda_r^s(\mathbf{F}_\psi)$  decreases with a higher number of reaction progress variables. This means, that for more than three progress variables, the ILDMs can probably not be assumed to be independent of the underlying fuel. In this case the ILDM of the higher hydrocarbon can nevertheless be generated hierarchically using the ILDM of the smaller system (in this example syngas) as initial guess.

To check the hierarchical concept detailed (lines) and reduced (symbols) mechanisms are compared (see Figs. 4 and 5). Detailed results correspond to results obtained from a calculation of a laminar free

flat flame using the detailed n-heptane mechanism (presented above) and the results of the reduced chemistry correspond to species concentrations predicted by the different ILDMs. According to the analysis procedure the reduced syngas mechanism (symbols  $\square$ ) can be used directly as a generic representation of the n-heptane system. The symbols  $\nabla$

Even in the case that the criteria above for a use of generic ILDMs are not met, nevertheless, the ILDMs can be generated hierarchically. This means that ILDMs of higher hydrocarbons are calculated using existing ILDMs of smaller hydrocarbons as an initial guess. This has been done for the example considered in this paper, and as a result, exact ILDMs (symbols  $\Delta$  in Figs. 4 and 5) are obtained, but the computational effort is reduced considerably. Results obtained with ILDMs generated by this method are also plotted in Figs. 4 and 5, but one can see that of course they do not differ from the results obtained using ILDMs which have been generated by the standard straightforward method. An alternative to the hierarchical generation of ILDMs consists in the use of perturbation theory for invariant subspaces [17]. This yields an estimation of the influence of the added chemical processes on the original system. Assuming that the eigenvalues of the underlying system are good approximations this leads to a reduction of the computational effort, too. Such an analysis is, however, beyond the scope of this paper.

It was shown (see Fig. 4 and 5) that the different ILDMs exhibit no visible differences and, therefore, yield the same accuracy if they are implemented in flame calculations. This causes the interesting question, how much CPU time is saved using the hierarchical concept? The most drastic reduction of the computational effort is obtained, if only the analysis procedure is needed. This leads to a reduction of the

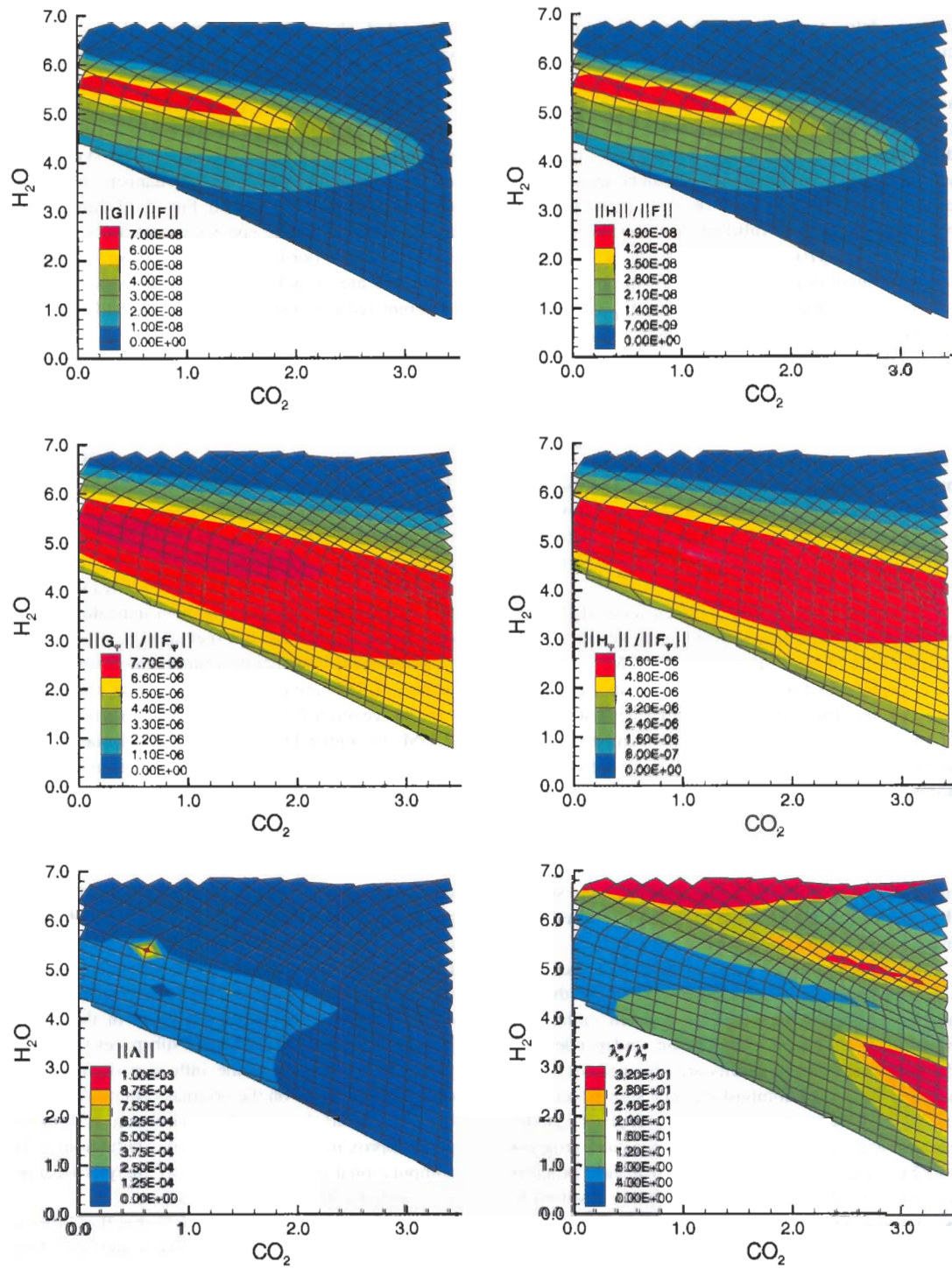


Fig. 2. Contour plots of the results obtained from the analysis procedure for an n-heptane system as a function of the reaction progress variables. Species concentrations are in units of  $[\nu_i/M_i] = \text{mol/kg}$ .

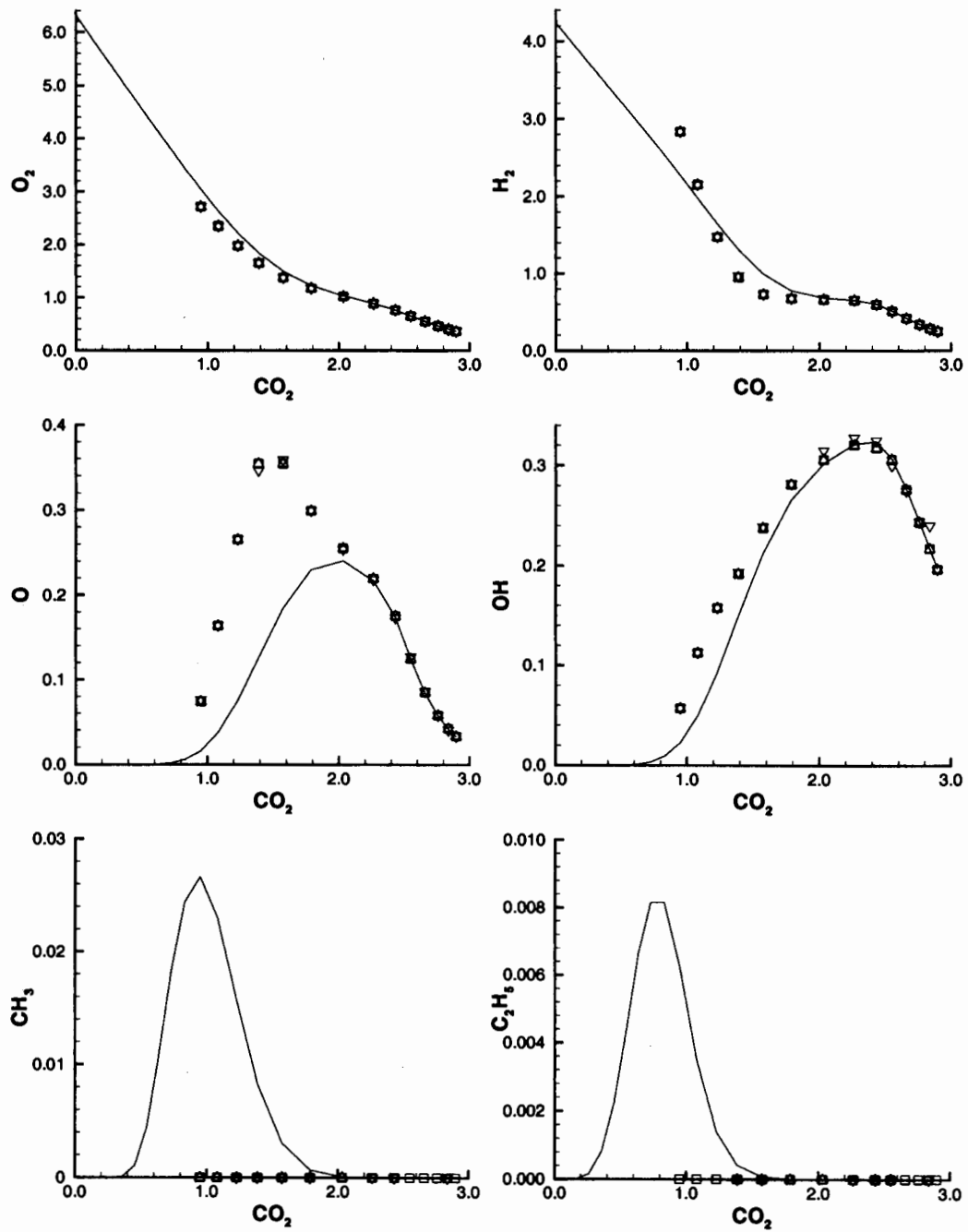


Fig. 4. Comparison of the results of a flame calculation using 2-dimensional ILDMs and a detailed mechanism. CO<sub>2</sub> is used to describe the reaction progress. Lines: detailed n-C<sub>7</sub>H<sub>16</sub> mechanism, □: CO/H<sub>2</sub> ILDM, Δ: hierarchically generated n-C<sub>7</sub>H<sub>16</sub> ILDM, ∇: conventionally generated n-C<sub>7</sub>H<sub>16</sub> ILDM. Species concentrations are in units of [w<sub>i</sub>/M<sub>i</sub>] = mol/kg.

computing time by a factor of  $\approx 27$  for the 2-dimensional ILDM and  $\approx 25$  for the 3-dimensional ILDM. For the ILDMs generated using ILDMs of the sim-

pler fuel as starting guess (symbols Δ in Fig. 4 and 5) a factor of  $\approx 9$  for the 2-dimensional ILDM and  $\approx 5$  for the 3-dimensional ILDM is obtained. The differ-

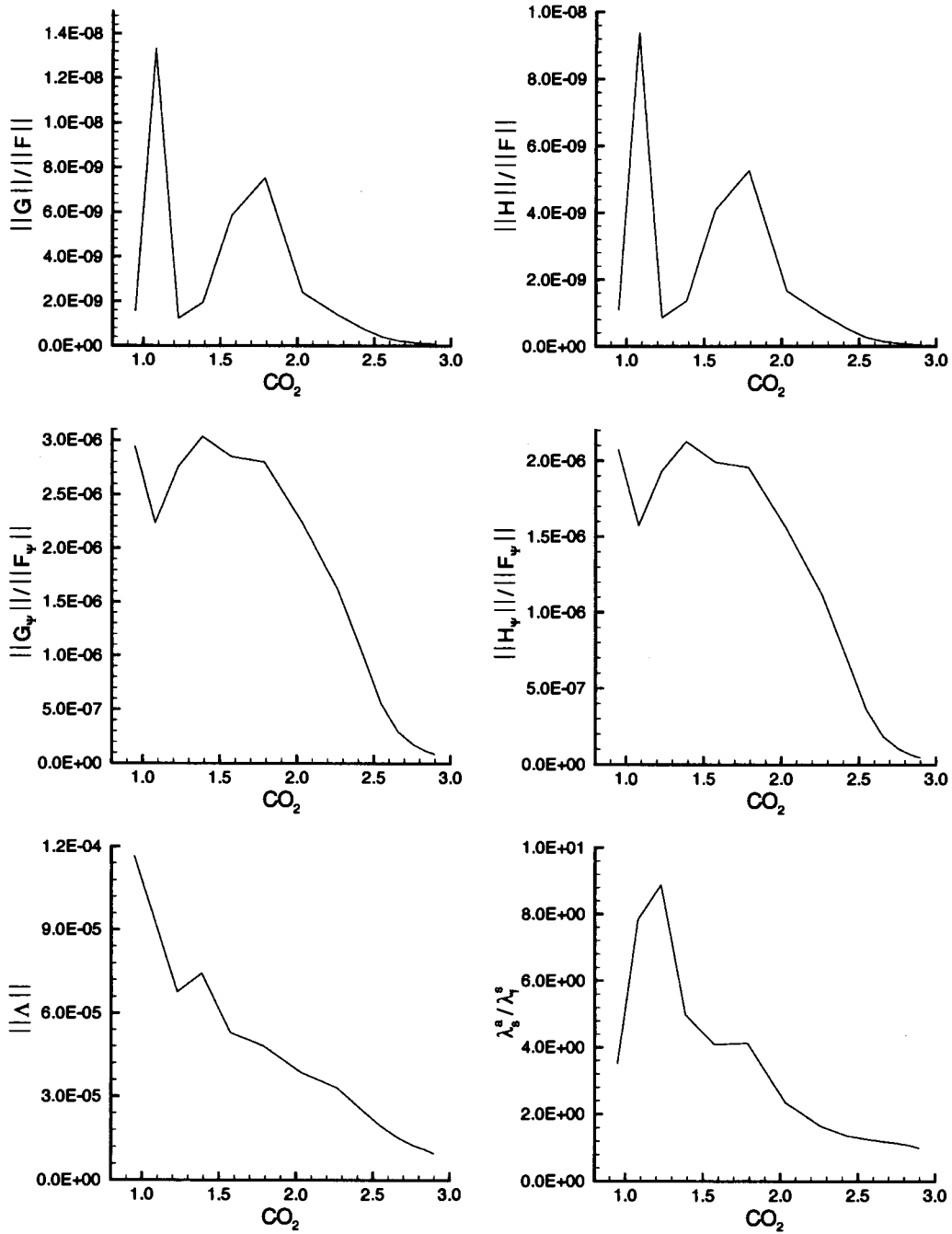


Fig. 3. Criteria of the analysis procedure plotted against the species concentration of  $\text{CO}_2$ . Presented are the results of an analysis of the n-heptane system based on a 3-dimensional syngas ILDM. Species concentrations are in units of  $[w/M_i] = \text{mol/kg}$ .

ence between these two factors is caused by the fact, that with an increasing number of reaction progress variables the ILDMs contain more and more infor-

mation about the used fuel. This means, the initial guess of the 3-dimensional ILDM is typically not as good as for the 2-dimensional ILDM.



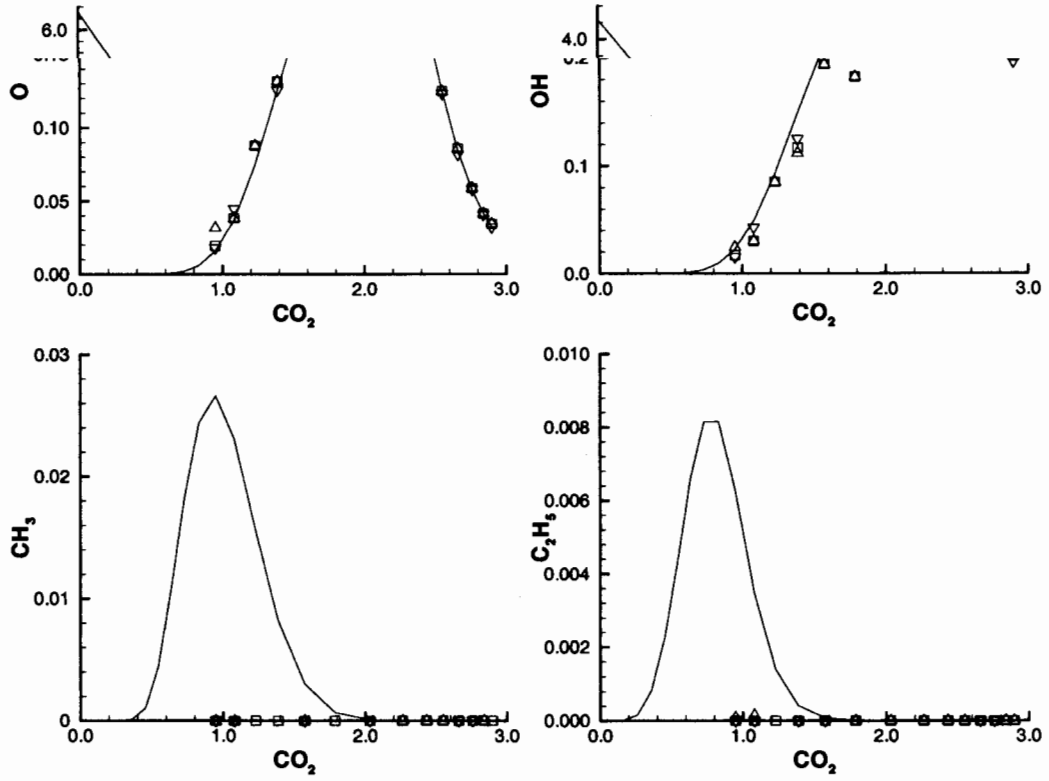


Fig. 5. Comparison of the results of a flame calculation using 3-dimensional ILDMs and a detailed mechanism. CO<sub>2</sub> is used to describe the reaction progress. Lines: detailed n-C<sub>7</sub>H<sub>16</sub> mechanism, □: CO/H<sub>2</sub> ILDM, Δ: hierarchically generated n-C<sub>7</sub>H<sub>16</sub> ILDM, ▽: conventionally generated n-C<sub>7</sub>H<sub>16</sub> ILDM. Species concentrations are in units of [w<sub>i</sub>/M<sub>i</sub>] = mol/kg.

#### 4. Conclusions

In a previous paper [1] it had been shown, that the hierarchical structure of detailed chemical mecha-

nisms is reflected in the ILDMs. This behavior can be used for an efficient calculation of ILDMs and, therefore, overcomes the costly computation of ILDMs of complex fuels. The hierarchical concept, presented in

this paper, consists of an analysis procedure which decides whether the investigated ILDM is fuel specific or not. If the ILDM is independent of the used fuel, it can be used directly for the higher hydrocarbon. Otherwise the ILDM of the smaller fuel can at least be used as initial guess for the ILDM of the higher hydrocarbon and, thus, leads to a reduction of the required CPU time. The hierarchical concept provides a procedure for an efficient calculation of ILDMs of complex fuels. The presented examples show, that 2- and 3-dimensional ILDMs of hydrocarbons for a given enthalpy, pressure, and element composition are very often not fuel specific, which allows the generation of “generic ILDMs” which are not fuel specific and depend only on enthalpy, pressure, and element composition.

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