

LOCAL AND GLOBAL SIMILARITY OF SENSITIVITY VECTORS OF COMBUSTION KINETIC MODELS

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Abstract

Local sensitivity functions $\partial Y_i / \partial p_k$ of many chemical kinetic models exhibit three types of similarity: (i) *local similarity*: ratio $\lambda_{ij} = (\partial Y_i / \partial p_k) / (\partial Y_j / \partial p_k)$ is equal for any parameter k ; (ii) *scaling relation*: ratio λ_{ij} is equal to $(dY_i / dz) / (dY_j / dz)$; (iii) *global similarity*: ratio $(\partial Y_i / \partial p_k) / (\partial Y_j / \partial p_m)$ is constant in a range of the independent variable z . Similarities can be detected by calculating the ratios above or, in a more efficient way, via the investigation of the correlations based on the scalar product of the corresponding sensitivity vectors. Local similarity may be a consequence of the existence of low-dimensional slow manifolds in chemical kinetic systems. Scaling relation may be present, if the dynamics of the system is controlled by a one-dimensional slow manifold. Global similarity emerges if local similarity is present and the sensitivity differential equations are pseudo-homogeneous. Global similarity means that the effect of the simultaneous change of several parameters can be fully compensated for all variables, in a wide range of the independent variable by changing a single parameter. The similarity relations are very important from a practical point of view in the fields of the 'validation' of complex reaction mechanisms and parameter estimation of chemical kinetic systems. Global similarity of models can be revealed by the principal component analysis of the sensitivity matrices. The statements are illustrated by numerical examples related to the homogeneous explosion and adiabatic laminar flames of stoichiometric methane-air mixtures.

Introduction

Sensitivity analysis is a widely used tool for the study of chemical kinetic and combustion models [1]. Most of the combustion simulation programs calculate local sensitivity coefficients $s_{ik} = \{\partial Y_i / \partial p_k\}$, which show the change of model result Y_i if parameter p_k has been slightly changed. In case of a general mathematical model, no relation is expected among the rows and/or the columns of the sensitivity matrix $S = \{s_{ik}\}$. However, in several chemical kinetic systems the following relations have been observed [2], [3]:

(i) *local similarity*: Value

$$\lambda_{ij}(z) = \frac{s_{ik}(z)}{s_{jk}(z)} \quad (1)$$

depends on the independent variable z (time or distance) and the model results Y_i and Y_j selected, but is independent of parameter p_k perturbed.

(ii) *scaling relation*: Equation

$$\left(\frac{dY_i}{dz} \right) = s_{ik}(z) \quad (2)$$
$$\left(\frac{dY_j}{dz} \right) = s_{jk}(z)$$

is valid for any parameter p_k . Existence of scaling relation includes the presence of local similarity.

(iii) *global similarity*: Value

$$U_{ym} = \frac{s_{ik}(z)}{s_{jm}(z)} \quad (3)$$

is independent of z (within an interval) and the model output studied.

Global similarity of sensitivity functions was first detected by Reuven *et al.* [4], who had created a computer code for the calculation of local sensitivities of stationary two-point boundary value problems and tested it on a symbolic model of 1D laminar flames. The original name was self-similarity, but it was changed [2] to global similarity, because 'self-similarity' is a widely used name for a different notion. Smooke *et al.* [5] used this program for the calculation of stationary adiabatic premixed laminar hydrogen-air flames and again global similarity of the sensitivity curves was found. Mishra *et al.* [6] applied the code for the calculation of sensitivities of adiabatic premixed burner-stabilized laminar CO/H₂/O₂ flames. They reported not only global similarity, but also scaling relations. Mishra *et al.* found that the similarity relations are not valid if the temperature profile is fixed. This observation suggested the importance of temperature as a variable that strongly couples the other variables in adiabatic combustion systems.

Rabitz and Smooke [3] claimed that the onset of scaling relations and global similarity could be explained by assuming that there is a single *dominant variable* in the system. According to their definition, a variable is called dominant, if changing the value of it changes the values of all other variables, but perturbation of the value of a non-dominant variable changes the values of other non-dominant variables most significantly through the perturbation of the value of the dominant variable. They assumed temperature to be the single dominant variable in adiabatic combustion models. Vajda *et al.* [7] investigated this subject by comparing sensitivity functions of models of adiabatic explosions and laminar flames of hydrogen-air mixtures. They concluded that the sensitivity functions of flames are much more similar than that of homogeneous explosions. Vajda and Rabitz [8] studied thermal explosions modelled by a single step, n th-order, exothermic reaction. Their model had two variables: temperature and fuel concentration. Global similarity was observed for parameter sets where the model simulated thermal runaway. In a dynamical system, the parameter perturbation causes a shift in the values of system variables, which induces further changes of these values. The system of sensitivity differential equations is called *pseudo-homogeneous*, if after some time (and/or distance) this indirect effect on system variables is much more significant than the direct effect of parameter perturbation. Vajda and Rabitz [8] stated that the onset of global similarity for explosions could be explained if the model has two properties: temperature is the single dominant variable and the sensitivity equations are pseudo-homogeneous in a time window.

In a series of recent papers [2], [9], [10], we have reinvestigated the similarity of sensitivities of various models of hydrogen-air combustion. The theoretical statements of our

previous papers are summarized here and the investigations are extended to another system, the combustion of methane-air mixtures.

The origin of local and global similarity

Changes of concentrations and temperature in a constant pressure spatially homogeneous reaction system can be described by the following initial value problem:

$$d\mathbf{w}/dt = \mathbf{f}_w(T, \mathbf{w}, \mathbf{p}), \quad \mathbf{w}(0) = \mathbf{w}^0 \quad (4a)$$

$$\dot{u}T/dt = f_T(T, \mathbf{w}, \mathbf{p}), \quad T(0) = T^0 \quad (4b)$$

where T is temperature, t is time, \mathbf{w} is the $(N-1)$ -vector of mass fractions, \mathbf{p} is the M -vector of parameters and T^0 and \mathbf{w}^0 are the initial values of temperature and mass fractions, respectively. Let \mathbf{Y} denote the vector of variables, that is $\mathbf{Y} = (\mathbf{w}, T)$ and accordingly $\mathbf{f} = (\mathbf{f}_w, f_T)$. Parameter vector \mathbf{p} includes the Arrhenius coefficients of reactions, the heat of formation of species, etc. Local sensitivity coefficient $s_{ik} = \partial Y_i / \partial p_k$ gives information on the effect of the small change of parameter p_k on model output Y_i . The sensitivity coefficients constitute the first order local sensitivity matrix $\mathbf{S} = \{\partial Y_i / \partial p_k\}$. This sensitivity matrix can be calculated by solving the following initial value problem:

$$\dot{\mathbf{S}} = \mathbf{J}\mathbf{S} + \mathbf{F}, \quad \mathbf{S}(0) = \mathbf{0} \quad (5)$$

where $\mathbf{J} = \partial \mathbf{f} / \partial \mathbf{Y}$ is the Jacobian and $\mathbf{F} = \partial \mathbf{f} / \partial \mathbf{p}$. If matrix \mathbf{F} were a zero matrix, equation (5) would be homogeneous. In some systems $\|\mathbf{F}\| \ll \|\mathbf{J}\mathbf{S}\|$, and then equation (5) is called pseudo-homogeneous [8]. Relation of the two terms on the right-hand-side of equation (5) has a physical meaning, which is explained here using the terms of a chemical kinetic model. Perturbation of an A -factor of a reaction changes the production rates of the species that are participating in this reaction, and therefore the concentrations of these species will be changed, which can be called the direct effect of parameter perturbation. After some time, the modified concentrations change also the rates of other reactions and thus modify all other calculated concentration values (indirect effect). Shortly after the parameter perturbation only the direct effect is significant ($\|\mathbf{F}\| \gg \|\mathbf{J}\mathbf{S}\|$). Having elapsed longer time, the direct and the indirect effects can be of comparable magnitude (equation (5) is inhomogeneous) or the direct effect becomes negligible compared to the indirect effect and equation (5) becomes pseudo-homogeneous.

In many chemical kinetic simulations, the calculated sensitivity functions show some or all of the similarity relations above. Using equations (1) to (3), the similarity of sensitivity coefficient functions can be demonstrated. However, the similarity relations can be investigated also through the sensitivity vectors. Let $\mathbf{s}_i^T = \partial Y_i / \partial \mathbf{p}$ denote the vector of the i th row of the sensitivity matrix, where superscript T denotes transpose of a vector. The local similarity of sensitivity vectors implies relation

$$\mathbf{s}_i^T(t) = \lambda_{ij}(t) \mathbf{s}_j^T(t). \quad (6)$$

Equation (6) means that all other rows of the sensitivity matrix can be obtained from any non-zero row. Consequently, the rank of the sensitivity matrix is one. If rank r of the sensitivity matrix is greater than 1 but less than N (assuming that $N < M$), it might mean that there are

similarity relations among some of the rows of the sensitivity matrix, while the other rows are independent from the similar ones.

If scaling relation is also present, then scaling factor λ_{ij} is equal to the ratios of production rates (cf. equation (2)):

$$\mathbf{s}_i^T(t) = \frac{\frac{dY_i}{dt}(t)}{\frac{dY_j}{dt}(t)} \mathbf{s}_j^T(t) \quad (7)$$

Global similarity can be characterized by the following vector equation

$$\mathbf{s}_i^T(t') = \mu(t, t') \mathbf{s}_i^T(t), \quad (8)$$

if global similarity is valid in interval (t_1, t_2) and $t, t' \in (t_1, t_2)$.

We have shown [2] that the similarity of sensitivity functions is related to the existence of low-dimensional slow manifolds in chemical kinetic systems. Probably Roussel and Fraser [11] were the first who described the evolution of kinetic systems in connection with slow manifolds. They stated that the existence of very different time scales in chemical kinetic systems causes the trajectory of the solution to move on slow manifolds. In a dynamical system of N variables, the degree of freedom of the movement is originally $N_1 < N$, after deducing the exact and the approximately valid conservation relations. Examples for the former are the element conservations in closed systems. The trajectory originally moves on an N_1 dimensional manifold, but as time advances usually the dynamical dimension of the movement decreases and after some time the trajectory moves close to a two-dimensional surface (curved plate), then close to a one-dimensional curve, and finally arrives to the zero-dimensional equilibrium point if it exists. Maas and Pope [12] elaborated algorithms and computer codes for the approximate numerical calculation of slow manifolds. They also studied the existence of manifolds in several combustion models and generated reduced models having few variables only. In the functional description of slow manifolds, there are no distinguished independent variables; the manifolds can be locally parameterised by any non-constant variables.

The *scaling relation* can be explained [2] on the basis of two assumptions: (i) the dynamical behaviour of the system is controlled by a one-dimensional slow manifold in the space of the variables; (ii) an infinitesimal change of a parameter changes the velocity of the movement on the manifold, but negligibly dislocates the manifold.

Let the one-dimensional manifold be defined by vector function \mathbf{H} .

$$\mathbf{Y}(z, \mathbf{p}) = \mathbf{H}(Y_1(z, \mathbf{p})), \quad (9)$$

where function \mathbf{H} provides the values of all variables Y_i as a function of the arbitrarily selected parameterising variable Y_1 . Letter z denotes the single independent variable (time or distance). Assume that the kinetic system of differential equations are autonomous, therefore \mathbf{H} does not depend directly on z . Also, it can be assumed that a small change of parameters negligibly dislocates the manifold, therefore function \mathbf{H} does not depend directly on \mathbf{p} . Differentiating equation (9) with respect to z gives:

$$\frac{\partial \mathbf{H}(z, \mathbf{p})}{\partial z} = \frac{\partial \mathbf{H}}{\partial Y_1} \frac{\partial Y_1(z, \mathbf{p})}{\partial z}, \quad (10)$$

while differentiating equation (9) with respect to any parameter p_j yields

$$\frac{\partial Y_1(z, \mathbf{p})}{\partial p_j} = \frac{\partial \mathbf{H}}{\partial Y_1} \frac{\partial Y_1(z, \mathbf{p})}{\partial p_j}. \quad (11)$$

Combination of these two equations gives a scaling relation:

$$\frac{\partial Y_1(z)}{\partial p_j} = \frac{\partial Y_1(z)}{\partial p_j} \frac{\partial Y_1}{\partial z} \left(\frac{\partial Y_1}{\partial z} \right)^{-1} \quad (12)$$

This equation is valid for both temporal and 1D stationary systems and can be easily converted to equation (7), which does not contain variable Y_1 . This also supports the claim that in equation (9) selection of variable Y_1 is not unique. The reasoning above can be extended to multi-dimensional manifolds. In this case, existence of the manifold causes linear dependency of the rows of the sensitivity matrix, but the scaling relation is not valid.

Vajda and Rabitz [8] suggested that a necessary condition of global similarity is the pseudo-homogeneity of the sensitivity differential equations and demonstrated it on the example of a single reaction-step model of thermal explosion having two variables. Their derivation was extended and developed further in article [2]. The conclusion of the reasoning there was that the pseudo-homogeneous property of the sensitivity differential equations and the presence of local similarity together imply global similarity.

Investigation of the correlation of sensitivity vectors

According to equation (6), local similarity means that a sensitivity vector can be obtained by multiplying any other non-zero sensitivity vector by a scalar. In other words, in the N -dimensional space of variables two locally similar sensitivity vectors point to the same direction. The level of local similarity can be defined [9] by the difference in the direction of the two vectors, which can be obtained by the calculation of the scalar product of the normalized vectors:

$$\hat{\mathbf{s}}_i^T \hat{\mathbf{s}}_j^T = \cos \theta_{ij}, \quad (13)$$

Here $\hat{\mathbf{s}}_i^T$ and $\hat{\mathbf{s}}_j^T$ are the normalized (unit length) sensitivity vectors and θ_{ij} is their angle. Local similarity can be characterized by $c_{ij} = \cos \theta_{ij}$, which is between -1 and $+1$. This value represents a kind of correlation measure, which is in accordance with the other correlation functions used in mathematical statistics and the geometric interpretation of the correlation of vector elements. A detailed comparison of the correlation functions and a discussion on their meaning is given in reference [9]. If c_{ij} is near to $+1$, it means that the sensitivity vectors are similar and there is positive correlation between the elements, that is increasing two parameters separately change the calculated values of all variables to the same direction. If c_{ij}

is near to -1 , then the sensitivity vectors are similar, there is negative correlation between the elements, and increasing one parameter has similar effect to decreasing the other parameter. If c_{ij} is close to zero, then the sensitivity vectors are not correlated and are not similar. In the next section, correlation function c_{ij} is always used in such a way that the correlation of several sensitivity vectors will be compared to a selected reference sensitivity vector.

Another tool for the investigation of the correlation of sensitivity vectors [9] is the application of cobweb plots. The elements of the examined vectors are plotted along vertical sections in a way that the greatest negative element is at the bottom, the greatest positive element is at the top, the zero is in the middle, and all other vector elements are linearly scaled in between these points. The sections belonging to the different vectors are arranged next to each other and the corresponding vector elements of the neighboring vectors are connected, *i.e.* $s_{i,1}$ is connected to $s_{i,2}$, $s_{i,2}$ is connected to $s_{i,3}$ *etc.* If the vectors are positively correlated, the lines are horizontal; if they are negatively correlated, the lines form an asterisk-like pattern, and no regular pattern can be observed if there is no correlation between the two vectors.

Similarity of the sensitivity vectors of methane-air combustion models

In the simulations presented here, the Leeds Methane Oxidation Mechanism [13], [14] was used, which contains 37 species and 350 (irreversible) reactions. The combustion of stoichiometric mixtures was investigated. Concentration-time and sensitivity-time curves of adiabatic explosion were calculated by program SENKIN [15]. The initial conditions were $p = 1$ atm, $T_0 = 1000$ K. Premixed laminar flames were simulated by program PREMIX [16]. The freely propagating laminar premixed flames had cold boundary conditions $p = 1$ atm, $T_0 = 298.15$ K. For the stationary simulation of freely propagating flames, the coordinate system moves with the flame front and it is fixed to a point of a given temperature of the flame. In our calculations, the reference temperature was always 400 K. In the case of burner-stabilized flames, the starting point of the coordinate system is the burner-surface and the shape of the flame is determined by the mass flow rate of the fuel-air mixture. If the mass flow rate corresponds to the velocity of the freely propagating flame, the flame becomes adiabatic and the temperature and concentration profiles become identical to that of the freely propagating flame.

In all figures the results are plotted as a function of temperature instead of time (explosions) or distance (1D stationary flames). This is an equivalent representation of data, because temperature continuously increases with time and distance in adiabatic homogeneous explosions and 1D stationary flames, respectively. Also, presentation of the results as a function of temperature is more meaningful for the researchers of the combustion community. The combustion models contain large number of parameters, like Arrhenius parameters, thermodynamic data *etc.* For simplicity, in the subsequent calculations only the sensitivities of the calculated concentrations and temperature with respect to the preexponential factors A of the reactions will be investigated.

Figure 1 shows the sensitivity-temperature functions of the calculated CO_2 mass fractions in the case of adiabatic explosions. For plotting purposes, the semi-normalized $\partial y_{\text{CO}_2} / \partial \ln A_i$ sensitivity coefficient-temperature functions were drawn. The sensitivity curves are surprisingly well ordered. The next step is to check the existence of local similarity and scaling relations by calculating the appropriate ratios of sensitivity functions and production rates. Several parameters of the model are not effective, the corresponding sensitivity functions are near zero, and their value is calculated with large relative numerical error. For this reason, in Figure 2 the ratios of the sensitivity functions of only 90 effective parameters are plotted. This figure shows that all the ratios of the sensitivity functions of HO_2 and OH agree well above 1300 K. The dashed line in Figure 2 shows the ratio of the production rates

of HO_2 and OH . It does not coincide with the ratio of sensitivities, therefore scaling relation is not valid in this system, unlike in the adiabatic explosion of hydrogen-air mixtures [2].

Local similarity of sensitivity vectors can be investigated further by calculating the correlation of the vectors from the scalar products. Figure 3 compares the correlation of the sensitivity vector of CO_2 mass fraction with the sensitivity vectors of all other species at the same time (temperature). Above about 1300 K, these sensitivity vectors are always well correlated (positively or negatively) with that of CO_2 . This means that local similarity is valid in a wide range of time (temperature). If scaling relation were valid, the changes of correlation would coincide with the concentration extremes of the corresponding species [9]. In this case, the correlation changes are *not* at the location of the minimum or maximum of the concentration functions of the corresponding species.

One of the most sensitive parameters for the calculated CO_2 concentration is the preexponential factor of reaction $\text{O}_2 + \text{CH}_3 \rightarrow \text{CH}_2\text{O} + \text{OH}$. Ratios of the sensitivity coefficients of all other reactions and that of this reaction were calculated according to equation (3) and plotted in Figure 4. The ratios are constant above 1300 K indicating that global similarity is valid for the sensitivity functions of adiabatic methane-air explosions. Similar vertical lines were obtained when the sensitivity ratios belonging to other species were plotted. Presence of local similarity and pseudo-homogeneity of the sensitivity differential equations was stated [2] to be a necessary condition of global similarity. To check the latter condition, in Figure 5 the ratios of the norms of the inhomogeneous \mathbf{f}_j and the homogeneous $\mathbf{J}\mathbf{s}_j$ terms for each parameter j of the sensitivity differential equation (5) is plotted. The ratios are almost zero above a threshold time (temperature) for each parameter, showing the pseudo-homogeneity of the sensitivity ODE and supporting the statement.

It has been noted [2], that the presence of diffusion decreases the level of similarity. Figure 6 is similar to Figure 3, but in this case the correlation the sensitivity functions of a burner-stabilized flame was plotted. The sensitivity functions of about half of the species are well correlated with that of CO_2 , but in this case the correlation is poor for the rest of the sensitivity functions.

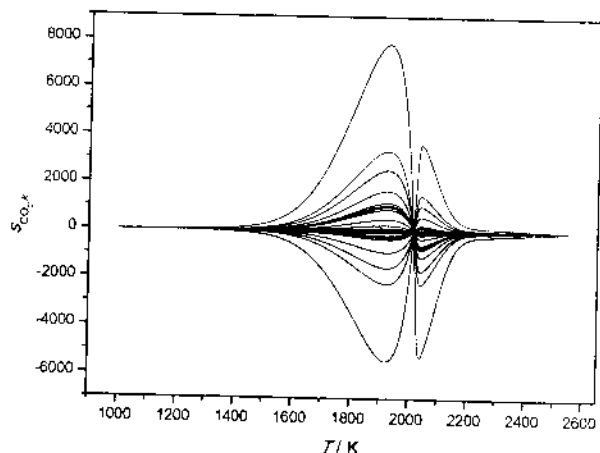


Figure 1. Semi-normalized CO_2 mass fraction sensitivity-temperature curves for the adiabatic explosion of a stoichiometric methane-air mixture. Each curve belongs to a different preexponential factor A_j .

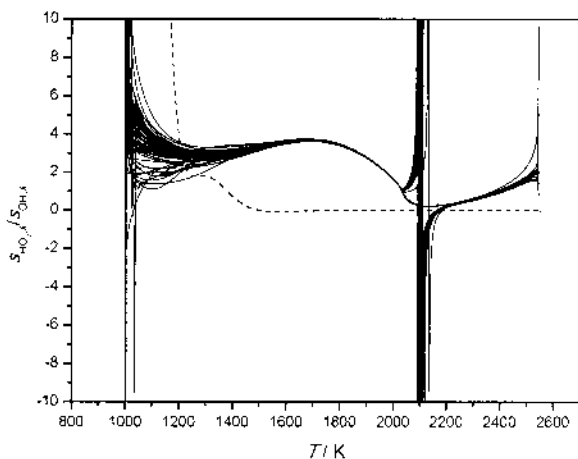


Figure 2. The solid lines show the ratio of sensitivity functions of HO₂ and OH for 90 reactions. The dashed line indicates the ratio of the corresponding production rates.

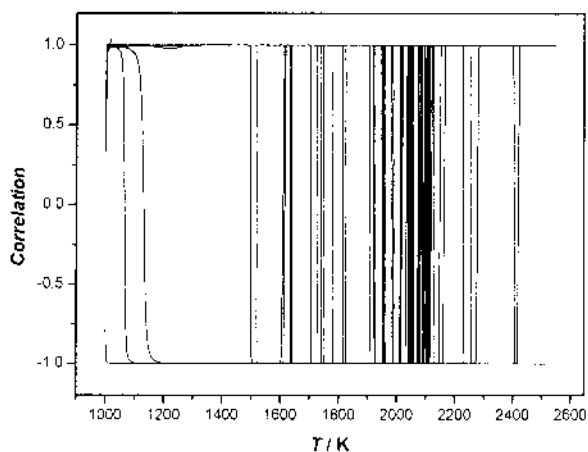


Figure 3. Correlation of all sensitivity vectors with the sensitivity vector of CO₂ as a function of temperature for a stoichiometric methane–air explosion. The figure shows the existence of good local similarity of sensitivity vectors above 1300 K.

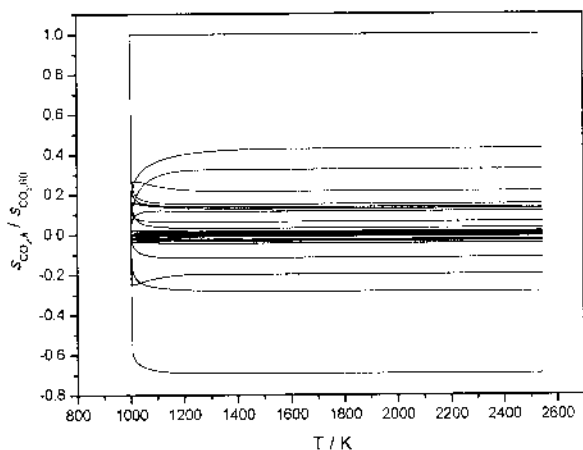


Figure 4. Considering the sensitivity of the calculated CO_2 concentration, the ratio of the sensitivity functions of the preexponential factors of each reaction and that of reaction 60 ($\text{O}_2 + \text{CH}_3 \rightarrow \text{CH}_2\text{O} + \text{OH}$) were calculated. The horizontal lines show that global similarity is present among the sensitivity vectors of methane-air stoichiometric explosions.

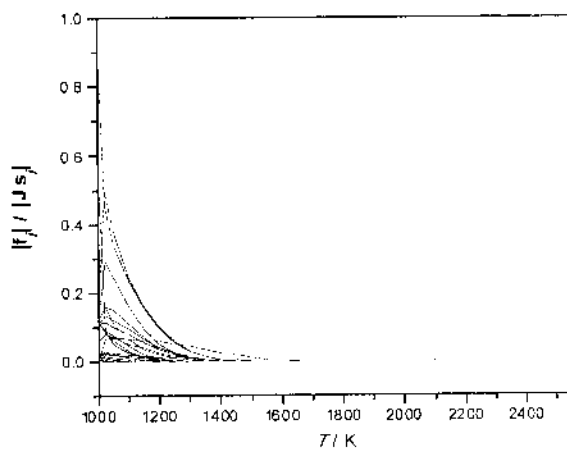


Figure 5. Ratio of the norms of the inhomogeneous \mathbf{f}_j and the homogeneous $\mathbf{J}\mathbf{s}_j$ terms for each parameter j of the sensitivity differential equation in the case of adiabatic homogeneous explosion of methane-air mixtures. The near-zero ratios show that the sensitivity ODE is pseudo-homogeneous above a threshold temperature.

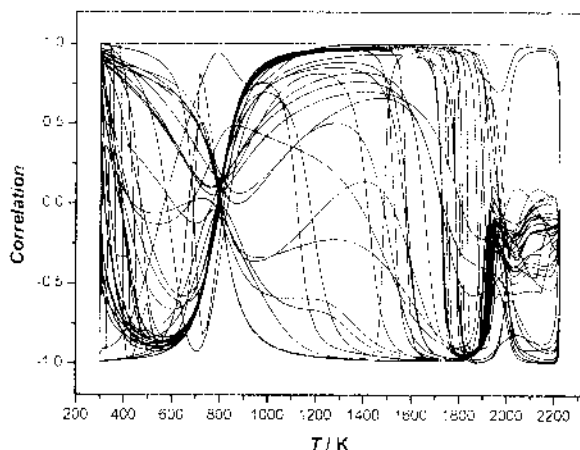


Figure 6. Correlation of all sensitivity vectors with the sensitivity vector of CO_2 as a function of temperature for a methane-air stoichiometric 1D stationary burner-stabilized flame. The figure shows that local similarity is valid for about half of the sensitivity vectors.

The importance of the similarity of sensitivity functions in chemical kinetics

Global similarity means that if several parameters are changed in a model, its effect can be fully compensated by changing a single effective parameter. This way the values of *all* variables can be restored to the original value in a *wide range* of time or distance. If only local similarity is present, the values of *all* variables are restored simultaneously, but only at a point of the independent variable.

In the case of empirical models, the only task of the model is to provide a good description of the observations. Presence of global similarity means that different parameter sets can provide the same simulation results. In case of physical models, all parameters are assumed to have a 'true' value, which can be unambiguously determined in independent experiments. Perfect agreement between the bulk experimental and the simulation results for all variables in a wide range of time (distance) is usually considered to be a proof that all of the used parameters are correct. Existence of global similarity means that if the values of some of the parameters are wrong, it can be fully masked by other parameters being also incorrect. If a physical parameter is determined in such a system by fitting to experimental data, error in the fixed parameter values cause the determined parameter to become erroneous. However, the fitted model perfectly reproduces all the experimental data, even if the values of several variables are measured at several time points (distances).

A powerful tool for the investigation of the efficiency and interaction of parameters for the calculated results is the principal component analysis [17] of the sensitivity matrices. The method includes the eigenvalue-eigenvector decomposition of matrix $\tilde{S}^T \tilde{S}$, where matrix \tilde{S} is composed of several normalized sensitivity matrices belonging to a series of points in time or distance. The number of large eigenvalues is equal to the number of effective parameter groups and these parameter groups are defined by the eigenvectors. It has been shown [10] that if all the component sensitivity matrices are selected from the region of global similarity, then matrix $\tilde{S}^T \tilde{S}$ has a single large eigenvalue.

Similarity relations of the sensitivity vectors is related to the observation that matrix $\tilde{S}^T\tilde{S}$ generated from chemical kinetic simulations has usually few large eigenvalues. The consequence is that complex reaction mechanisms can frequently reproduce all available bulk experimental data by tuning only few rate parameters. Probably this is the reason why very different reaction mechanisms exist in the literature that describe the same set of experimental data with similar level of accuracy [13].

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