



Numerical optimisation for model evaluation in combustion kinetics



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HIGHLIGHTS

- An overview of optimisation in combustion chemical kinetics is presented.
- Four optimisation methods were implemented and validated.
- Four mechanisms could not initially capture profiles related to ethane and methyl.
- GRI-mechanism 3.0 was optimised in that respect.

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ABSTRACT

Numerical optimisation related to the estimation of kinetic parameters and model evaluation is playing an increasing role in combustion as well as in other areas of applied energy research. The present work aims at presenting the current probability-based approaches along applications to real problems of combustion chemical kinetics. The main methods related to model and parameter evaluation have been explicated. An in-house program for the systematic adjustment of kinetic parameters to experimental measurements has been described and numerically validated. The GRI (Gas research institute) mechanism (version 3.0) has been shown to initially lead to results which are greatly at variance with experimental data concerning the combustion of CH_3 and C_2H_6 . A thorough optimisation of all parameters has been performed with respect to these profiles. A considerable improvement could be reached and the new predictions appear to be compatible with the measurement uncertainties. It was also found that neither GRI 3.0 nor three other reaction mechanisms considered during the present work should be employed (without prior far-reaching optimisation) for numerical simulations of combustors and engines where CH_3 and C_2H_6 play an important role. Overall, this study illustrates the link between optimisation methods and model evaluation in the field of combustion chemical kinetics.

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1. Introduction

The use of numerical methods for parameter estimation has become widespread in fields related to applied energy research. Amongst many other applications, they have been used for the development of a non-linear model describing a wind turbine [1] and modelling a fuel-cell [2], the heat dynamics of a building [3], and a hydraulic turbine [4], to name but a few of them. They are also increasingly employed for the development and evaluation of combustion models [5–8]. Computational Fluid Dynamics (CFD) simulation has become an essential ingredient for the optimal utilisation of complex power generation systems based on combustion [9–11]. Reliable computational predictions require the presence of an accurate chemical kinetic model which is

generated from the reduction of a complex reaction mechanism [12,13]. Since the reduction process lowers almost inevitably the accuracy of the kinetic description of the combustion [14], the detailed reaction mechanism must reach a high level of trustworthiness for allowing realistic CFD simulations of complex systems such as, say, internal combustion engines or gas turbines. The uncertainties of kinetic coefficients can lead to large prediction errors with respect to the release of pollutants and important combustion features such as ignition delay times or flame velocities because the parameter imprecisions get propagated towards all results [15,16]. Hence, a good understanding of combustion kinetics has become vital for the optimal and sustainable utilisation of fossil and renewable fuels [10,17,18] and the use of new fuels such as synthetic ones. It is universally recognised amongst researchers that the development of micro-kinetic detailed reaction mechanisms has been a great step forward for both the chemical understanding and the predictability of combustion processes.

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Such reaction mechanisms include all possible elementary reactions that can occur under a wide variety of conditions. They allow a detailed description of the chemical processes taking place on a molecular level. The determination of parameters corresponding to the real chemistry is a complex task. Traditionally, it has been achieved in three different manners.

- By designing experiments isolating some reactions in such a way that the model variables corresponding to the measurements can be expressed analytically as a function of parameters of interest. The optimal values can then be identified mathematically through a least-square regression.
- By using methods from theoretical chemistry such as Density Functional Theory calculations [19] coupled with Transition State Theory. Depending on the involved assumptions and simplifications, some methods, especially semi-empirical techniques, can lead to great uncertainties with respect to the evaluated parameters.
- By analogy with similar reactions with known rate coefficients. An unknown uncertainty is also introduced by this approach.

If the mechanism contains all possible reactions playing a role in the experiments at hand and enough profiles are available for an unambiguous estimation of all active parameters, the first method is the most promising one for obtaining values with great accuracy. Nevertheless, most chemical kinetic descriptions cannot be sufficiently simplified to provide analytical expressions precise enough for parameter estimation. In such a case, an optimisation method minimising the distance $d(p)$ between experimental results and model predictions must be utilised. The determination of optimal coefficients is itself closely related to model evaluation which consists of assessing how well a reaction mechanism can match a set of measurements.

The present article concerns the use of numerical optimisation methods for the evaluation of models used to describe combustion chemical kinetics such as those applied to the CFD simulation of power generation. It has been organised in such a way to remain relevant for model evaluation in other fields of applied energy. In Section 2, different approaches to the evaluation of kinetic models are presented and examined. In Section 3, the optimisation program Kinefit [14] is presented and validated. In Section 4, Kinefit is applied to the combustion of ethane and the methyl free radical. Finally, in Section 5, the conclusion of this work and future outlook are given.

2. Methodologies for model evaluation in chemical kinetics

2.1. Frequentist and Bayesian approaches

Frequentism and Bayesianism [20] are currently the main approaches utilised for the evaluation of predictive models. While confronted with the problem of estimating parameters out of a set of experimental data, both frequentists and Bayesians view a measurement (which might be a concentration, an ignition delay time, a flame velocity and so on.) as composed of three terms:

$$m = m_t + m_s + m' \quad (1)$$

where m_t is its true value, m_s its systematic error and m' a random fluctuation around $m_t + m_s$. The systemic error is always an unknown term which would be otherwise corrected. For the sake of parameter estimation, one has to suppose it is negligible and set it to zero. It is an assumption which might be well founded in cases where the measurement method has been independently validated. It can be more problematic otherwise, in situations where the measurement technique has not been well assessed. Let N be

the number of measured profiles, n_i the number of measurements for the i -th profile with $i \in \{1 \dots N\}$ whereas m_{ij} and e_{ij} designate the model and experimental values, respectively. Let σ_{ij} denote the standard deviations corresponding to the experimental values. The chi-squared norm is defined as

$$d = X^2 = \sum_{i=1}^N \sum_{j=1}^{n_i} \left(\frac{m_{ij} - e_{ij}}{\sigma_{ij}} \right)^2. \quad (2)$$

Since the standard deviations are frequently unknown, they are often pragmatically approximated as being proportional to the measurements $\sigma_{ij} = \epsilon e_{ij}$ whereby measurements equal to zero are not considered in the sum of Eq. (2). Following this rule, the relative distance or relative least-square can be defined as

$$d = X^2 = \sum_{i=1}^N \sum_{j=1}^{n_i} \left(\frac{m_{ij} - e_{ij}}{e_{ij}} \right)^2. \quad (3)$$

Under the assumption that the measurement errors are normally distributed, the probability that the experimental values would obtain if model M is correct $p(E|M)$ can be computed [8]. If $\nu = N_{data} - N_{parameters}$ is large then

$$p(E|M(k)) = p(X^2) = \frac{\Gamma\left(\frac{\nu}{2}, \frac{X^2}{2}\right)}{\frac{\Gamma}{2}} \quad (4)$$

whereby k represents the ensemble of uncertain parameters taking on some value ranges [8].

It is at the next stage that Bayesians and frequentists part ways. While the former use this value and a prior probability distribution of the parameter space for computing a posterior distribution via Bayes' theorem (with problems discussed in Section 2.2), the latter do not consider that model M has a probability of being true. Instead, they judge it according to its agreement with the experimental data represented by $p(E|M(k))$ which should be below a given threshold (there are no universal rules for determining such limits). If the model cannot reproduce the measurements within their uncertainties, it is rejected, otherwise it is deemed worthy of further considerations. It is worth noting that the truth of a model is indicated solely by its ability to correctly reproduce the set of experimental data while *all other possible alternatives* fail to do so [21]. The remainder of this subsection focuses on the frequentist approach to model evaluation.

If the parameters of a reaction mechanism could only take on one set of precise values known beforehand, the whole modelling enterprise in chemical kinetics would merely consist of comparing the performances of different models. In practice, this is never the case. A priori, parameters can have any of the values included within so-called feasible sets accounting for theoretical and experimental constraints. The parameters must then be optimised within this domain so as to minimise $X^2(k)$ and hence also maximise $p(E|M(k))$ according to the spirit of Maximum Likelihood Estimation [22]. The most widespread form of such feasible sets consists of hypercubes where all parameters are included between a lower and an upper bounds, that is $l_i \leq p_i \leq u_i$. The problem of this representation lies in the fact that parameters are very often tightly correlated with respect to their accounting for experimental data. Consequently, the feasible set is frequently spanned up by a complex geometry (which may not even be continuous). Frenklach [23] emphasised the need to consider the *whole* ensemble of available measurements for fitting kinetic parameters while determining a feasible set defined as all physically possible parameter values compatible with the experimental data and their uncertainties. In this way, the size of the feasible set (i.e. the imprecision of the parameter set) diminishes as new measurements come in.

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