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Determining predictive uncertainties and global sensitivities for large parameter systems: A case study for *n*-butane oxidation

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Abstract

A global sampling approach based on low discrepancy sequences has been applied in order to propose error bars on simulations performed using a detailed kinetic model for the oxidation of *n*-butane (including 1111 reactions). A two parameter uncertainty factor has been assigned to each considered rate constant. The cases of ignition and oxidation in a jet-stirred reactor (JSR) have both been considered. For the JSR, not only the reactant mole fraction has been considered, but also that of some representative products. A temperature range from 500 to 1250 K has been studied, including the negative temperature coefficient (NTC) region where the predictive error bars have been found to be the largest. It is this temperature region where the highest number of reactions play a role in contributing to the overall output errors. A global sensitivity approach based on high dimensional model representations (HDMR) has then been applied in order to identify those reactions which make the largest contributions to the overall uncertainty of the simulated results. The HDMR analysis has been restricted to the most important reactions based on a non-linear screening method, using Spearman Rank Correlation Coefficients at all studied temperatures. The final global sensitivity analysis for predicted ignition delays illustrates that the key reactions are mainly included in the primary mechanism, for temperatures from 700 to 900 K, and in the C_0—C_2 reaction base at higher temperatures. Interestingly, for predicted butane mole fractions in the JSR, the key reactions are almost exclusively from the reaction base, whatever the temperature. The individual contribution of some key reactions is also discussed.

Keywords: Uncertainties; Detailed kinetic model; Global sampling; Global sensitivity; Oxidation

1. Introduction

The effective use of combustion mechanisms to model and design practical devices requires robust models that can be used in a predictive way over wide ranges of temperatures, pressures and

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compositions. A measure of the robustness of a model can be obtained by estimating predictive error bars based on our knowledge about the uncertainties within the model parameterization and model structure [1]. However, model error bars are not commonly presented when comparing experimental data with equivalent model simulations. A single comparison is usually made using the best estimates of the model input parameters, with a local sensitivity analysis often applied afterwards to evaluate which parameters most strongly influence the selected target model outputs. Whilst local sensitivities have been used successfully to highlight important parameters within mechanisms for many combustion systems, the estimation of model error bars cannot generally rely on their use. This is particularly true for non-linear models, with input uncertainties that cover large ranges and high dimensional spaces. For such models, the propagation of uncertainties requires a sampling approach to ensure that all sensitive regions of the input space are covered. Sensitivity analysis is commonly used to explore discrepancies between model predictions and experimental data. However, even where good agreement is found, it should still form part of model validation since good agreement could occur due to cancellation of errors. Zsely et al. [2] highlighted the non-uniqueness of chemical kinetic models, suggesting that many different parameter sets may lead to similar target predictions. It is therefore useful to explore the combined influence of parameter variations on predicted output distributions requiring a global sensitivity approach.

Within such an approach, the uncertainties within the inputs are represented by a given distribution (uniform, log-normal etc.), which is then sampled and propagated through the model, providing distributions of the final model predictions. A large number of model runs may be required in order to obtain stable output statistics, such as the mean and variance of the predicted targets. The sampling approach used is critical, since we would like to obtain stable statistics using the lowest possible number of model runs in order to minimize computational costs [3,4]. Once stable output distributions are obtained, error bars may be calculated using variance based measures (e.g. 1σ or 2σ errors).

We may also wish to determine by how much each of the input parameter uncertainties contributes to the total output variance i.e. to perform an ANOVA (ANalysis Of VAriance) decomposition [5]. Such global sensitivity analyses are also usually based on sampling approaches and could be particularly challenging for large models where the input parameter space is highly multi-dimensional. Several approaches have been previously adopted for the global sensitivity analysis of kinetic models with those most commonly applied in combustion based on high dimensional model representations (HDMR) [6–8] and polynomial chaos expansions [9]. A review of available methods is given in [1].

In this paper we develop a methodology for estimating error bars for model simulations which incorporate high dimensional combustion mechanisms. Here we focus on uncertainties within the temperature dependent rate coefficients, but the approach could be applied to a wider range of inputs including thermodynamic parameters, transport properties, etc. We use a global sampling approach based on low discrepancy sequences with application to an *n*-butane oxidation model containing 1111 reactions [10,11]. A screening method is applied based on the calculation of Spearman Rank Correlation Coefficients (RCCs) of this input-output sample, in order to determine a subset of the main parameters which may affect the final errors over a wide range of conditions. A fully global sensitivity analysis is then performed for this parameter subset using HDMR. We demonstrate that it is possible to achieve an accurate variance decomposition of the output distributions using this two-step approach using reasonably small sample sizes. The work therefore provides a general method for estimating error bars for complex combustion models and obtaining a full ANOVA decomposition of these errors.

2. Methodology

Three types of experimental systems are mainly used to provide data for validating detailed low-temperature oxidation mechanisms: rapid compression machines (RCM), shock tubes (ST) and heated flow reactors, such as flow tubes or jet-stirred reactors (JSR). If models are able to reproduce such experimental data over wide ranges of temperatures and pressures, this suggests that the mechanisms may be appropriate for modeling practical combustion devices. However, discrepancies between model predictions and experimental data still exist for certain temperatures and it is therefore important to explore the impact of uncertainties in model input data on the model predictions.

The *n*-butane mechanism used in this study comprises 176 species and 1111 reactions. It is based on that proposed in [10] with updates by Bahrini et al. [11]. It is an automatically generated mechanism using the computer package EXGAS, previously used to produce oxidation mechanisms for many hydrocarbons and oxygenated fuels [12,13]. The system provides reaction mechanisms composed of three parts:

1. A comprehensive **primary mechanism** [13] including the following reactions: