Model-to-model Bayesian calibration of a Chemical Reactor Network for pollutant emission predictions of an ammonia-fuelled multistage combustor

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Abstract
In this work, we propose a Chemical Reactor Network (CRN), composed of only five reactors, to describe the pollutant emissions of a purely NH₃-fuelled, micro gas turbine-like, multistage prototype combustor in varying operating conditions. We calibrate the CRN model by treating its most influential model parameters as random variables, and we infer their posterior probability densities in a Bayesian setting using higher-fidelity data from RANS computations and explicitly accounting for model discrepancy. The final goal is to employ the calibrated CRN model with quantified fidelity to determine the combustor performance map and discover a robust optimal operating point that minimizes pollutants emissions.

Introduction
Hydrogen represents undoubtedly a promising alternative to carbonaceous fuels. However, more efficient hydrogen storage means are needed to tackle its energy-intensive handling [1]. This has pushed many research efforts towards investigating alternative and more economically viable hydrogen storage means. Among possible solutions, ammonia (NH₃) shows considerable potential because of its thermophysical properties similar to traditional hydrocarbons, which make it a more efficient H₂ storage mean [2]. However, its direct use as fuel poses technical issues, such as difficulties in achieving stable flames (low reactivity) and increased production of NOₓ with respect to hydrocarbons. Therefore, developing ad-hoc combustion technologies suited for NH₃ and its mixtures has been a research challenge within the last few years.
Several experimental and numerical works [3]–[5] highlighted the potential of NH₃ combustion under slightly rich conditions in reducing global NO emissions. Pugh et
al. [6] showed experimentally that a multistage NH\textsubscript{3} combustion process, where rich and lean combustion are performed in sequence, resulted in lower NO emissions. However, the secondary injection of air should be handled carefully. Although practical devices have already been successfully tested [7], [8], the room for improvement is still considerable, and the exploration of new scenarios at a conceptual and design level remains open.

Investigating disparate configurations and operating regimes can be labor-intensive if detailed experiments or numerical simulations such as computational fluid dynamics (CFD) are used. To this end, simplified/reduced-order models can help alleviate the computational cost considerably in the conceptual and preliminary design stages. Chemical Reactor Networks (CRN) models have often been used in ammonia combustion to characterize emissions at a conceptual level [9], [10]. CRNs are particularly useful for performing detailed kinetic simulations of practical combustion devices by schematically representing complex flow fields as a series of interconnected ideal reactor models [11]. CRNs can be designed based on a priori knowledge of the system [12] or by manually observing the main flow field characteristics from CFD or detailed experiments [13]. Due to the low associated computational cost, they enable large design space exploration in a highly reduced amount of time. Therefore, the possibility of employing CRNs for outer-loop applications, such as optimization and Uncertainty Quantification (UQ) tasks, is extremely attractive [14]. As an example, Yousefian et al. coupled a highly schematic CRN model with Non-Intrusive Polynomial Chaos Expansion (NIPCE) [15], [16] and with Bayesian Inference [17] to perform UQ of NO and CO emissions for a practical swirl-stabilized premixed burner. Although the developed tool showed considerable potential, its application has been limited to a simplified test case.

In this work, a joint CRN-UQ approach is pursued for the investigation under uncertainty of multistage ammonia combustion in a practical combustion device, namely a prototype of a micro–Gas Turbine (mGT) combustion chamber. The objective is to calibrate the CRN model parameters in a Bayesian setting using higher-fidelity data from RANS computations, explicitly accounting for model discrepancy. The calibrated model, with quantified uncertainty, can be thereupon employed to investigate a wide spectrum of operating conditions and identify a robust optimal operating point that minimizes pollutant emissions.

**Test Case**

The test case is a prototype of a mGT combustor consisting of a first, rich, non-premixed combustion stage followed by a secondary lean combustion region, schematically represented in Figure 1. The fuel is injected co-axially to the primary air in the first combustion stage. The hot mixture of reactants proceeds through the sleeve and reaches the secondary lean combustion stage where it reacts with the remaining fraction of air. Globally, the system is designed to operate at large oxygen excess. The air in mGT systems is usually preheated to allow heat recovery from hot gases, thus increasing the thermal efficiency of the cycle. The main operating
parameters influencing the final overall emissions of NO and NH₃, which will be analyzed in this work, are the air inlet temperature and equivalence ratio of the fuel-rich mixture, listed in Table 1 along with their considered ranges. Instead, we fix the outlet temperature at a value close to 1200 K, typically encountered in practical devices installed in gas-turbine devices. Therefore, the lean stream equivalence ratio results from a thermal balance.

**Table 1**: Operating parameters and range of conditions considered in this work.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Min value</th>
<th>Max value</th>
</tr>
</thead>
<tbody>
<tr>
<td>T_air</td>
<td>800 [K]</td>
<td>1000 [K]</td>
</tr>
<tr>
<td>φ</td>
<td>1.0 [-]</td>
<td>1.6 [-]</td>
</tr>
</tbody>
</table>

**CFD Model**

We first obtain six Reynolds-Averaged Numerical Simulations (RANS) of the combustion chamber to be later employed as high-fidelity data for the calibration of the CRN's model parameters. The RANS of the system were carried out using an axisymmetric, 2-dimensional grid. The commercial code ANSYS Fluent v19.5 was used to solve the RANS equations on a structured grid of 52k cells. The realizable $k-\varepsilon$ model was used for turbulence, while the Discrete Ordinate model was used to take radiation into account. A finite-rate chemistry approach with the chemical mechanism from Otomo et al. [18] (33 species, 213 reactions) was used to model chemistry, in conjunction with the Eddy Dissipation Concept (EDC) to account for turbulence-chemistry interactions effects. Regarding the boundary conditions, two air mass flow inlets are used so that the amount of air in the two zones can be adjusted by the user. A total of 15 simulations was performed, for five equivalence ratios, namely 1.0, 1.15, 1.3, 1.45 and 1.6, for three different air inlet temperatures, namely 800.0, 900.0 and 1000.0 K.

**Chemical Reactor Network model**

A Chemical Reactor Network (CRN) model is the simplification of a more complex flow field as a network of interconnected canonical reactor models, such as Perfectly Stirred Reactors (PSR) or Plug Flow Reactors (PFR). The first model is a 0D model, which implies perfectly mixed reactants within the control volume. In contrast, the second is a 1D model with the hypothesis of inviscid flow and perfect mixing along
the radial direction. The model employed in this work to schematically represent the system is displayed in Figure 2.

![Figure 2: Proposed CRN of the prototype mGT multistage combustor](image)

Model parameters characterize each reactor, such as residence time, volume, or heat transfer coefficient. Those parameters are typically estimated considering the real geometry (e.g., the volumes of the reactors) or by trial-and-error procedures, trying to obtain CRN predictions in agreement with more sophisticated modelling approaches, namely CFD simulations. This procedure is a parameter estimation problem in which prior knowledge and observations are employed to estimate/fit parameter values such that the model response fairly approximates the truth (e.g., measurements or higher-fidelity data).

The main parameters that characterize the proposed CRN are listed in Table 2. The rich combustion zone is modelled as an ignition adiabatic PSR connected with a first post-flame PFR with intermediate quenching. The parameter $U_q$ represents the global heat transfer coefficient of the rich zone. The parameters $V_i$ and $F_{ai}$ are the volumes of the i-th reactor and the fraction of the total air fed to the i-th reactor, respectively. CRN simulations were performed with the solver NetSMOKE++ [19] with the same kinetic mechanism employed in the CFD simulations.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Units</th>
<th>Nominal</th>
<th>Range</th>
<th>Distribution</th>
</tr>
</thead>
<tbody>
<tr>
<td>$U_q$</td>
<td>W/m²K</td>
<td>10.0</td>
<td>0.0 – 25.0</td>
<td>Uniform</td>
</tr>
<tr>
<td>$V_1$</td>
<td>cm³</td>
<td>30.0</td>
<td>30.0 – 100.0</td>
<td>Uniform</td>
</tr>
<tr>
<td>$F_{a1}$</td>
<td>n.a.</td>
<td>0.976</td>
<td>0.95 – 1.0</td>
<td>Uniform</td>
</tr>
<tr>
<td>$F_{a3}$</td>
<td>n.a.</td>
<td>0.10</td>
<td>0.05 – 0.15</td>
<td>Uniform</td>
</tr>
<tr>
<td>$F_{a4}$</td>
<td>n.a.</td>
<td>0.15</td>
<td>0.05 – 0.15</td>
<td>Uniform</td>
</tr>
<tr>
<td>$V_3$</td>
<td>cm³</td>
<td>710.0</td>
<td>600.0 – 1000.0</td>
<td>Uniform</td>
</tr>
<tr>
<td>$V_4$</td>
<td>cm³</td>
<td>388.0</td>
<td>300.0 – 500.0</td>
<td>Uniform</td>
</tr>
</tbody>
</table>

**Global Sensitivity Analysis**

To identify the most influential parameters of the CRN and calibrate them in a Bayesian framework, a surrogate model of the CRN was constructed using Polynomial Chaos Expansion:
\[ f(y) \approx f_s(y) = \sum_{j=1}^{P+1} \alpha_j \Psi_j(\xi) \]

where \( \alpha_j \) are deterministic coefficients, \( \Psi_j \) are suitably selected functionals of the random variable \( \xi \), while \( P \) is the truncation order. Legendre’s polynomials were selected in this work, and the coefficient estimation was carried out using the Dakota v6.16 framework using the methodology described here [20]. Sensitivity analysis was conducted by calculating the Sobol’s indices for each parameter, which represent their fractional contribution to the total variance of the model’s response (NO emissions in this specific case). They can be calculated as:

\[ S_i = \frac{V_{\xi_i} E_{\xi \sim i}(f(\xi | \xi_i))}{V(f(\xi))} = \frac{\sum_{i=1}^{L} \alpha_i^2}{\sum_{j=1}^{P+1} \alpha_j^2} \]

Results are reported in Figure 3. We can observe that only three parameters have a considerable contribution to the overall variance of the model, namely \( F_{a1}, U_q, \) and \( F_{a3} \). Therefore, the remaining four parameters can be excluded from the calibration procedure and set to their nominal values.

![Figure 3: Total Sobol’s indices for six selected cases normalized to sum 1](image)

**Bayesian Calibration of CRN model parameters on CFD data**

Data from CFD simulations, here considered to be high fidelity, can be used in a Bayesian setting to calibrate the CRN parameters, leveraging the Bayes’ theorem:

\[ P(\theta | D) = \frac{P(D | \theta) P(\theta)}{P(D)} \]

Where \( P(\theta | D) \) is the posterior probability of \( \theta \) given the observed data \( D \), \( P(D | \theta) = L(\theta) \) is the likelihood density, and \( P(\theta) \) is the posterior probability of the parameters and \( P(D) \) can be seen as a normalizing term. The model error is estimated via the approach developed by Sargsyan et al. [21] by embedding it directly into the model’s parameters:

\[ y_i \approx f(x_i, \theta + \delta) + \epsilon_i^d \]
where $x_i$ represent a given operating condition, and the term $\delta$ is the embedded model error, while $\epsilon_i^d$ represents the measurements’ noise. Calibration was carried out using the UQtk library developed by SANDIA National Laboratories, and results are reported in Figure 4, where a confident agreement can be observed between NO emissions predicted with the calibrated CRN and the emissions predicted by the RANS simulations.

**Figure 4:** Results of CRN calibration. NO emissions compared with CFD predictions and their confidence bounds.

**Conclusions**

In this work, model-to-model calibration was used to tune the parameters of a 5-reactor CRN of a multistage, mGT-like prototype combustor fed with pure NH$_3$. 15 CFD simulations were used as high-fidelity data to update first-guess, a priori distributions of CRN model parameters. The number of parameters to update was chosen by a preliminary sensitivity analysis, where Polynomial Chaos Expansion (PCE) was used to train a surrogate model of the CRN. The Bayesian calibration of the most influential parameters was then performed with model error embedding. Final NO predictions of the calibrated model are in great agreement with the CFD predictions. The advantage of this approach is that the CRN discrepancy is considered, and confidence bounds are also available. This approach is particularly useful when investigation of wide range of operating conditions is necessary.
References


