AMMONIA/HYDROGEN COMBUSTION IN A GAS TURBINE: UNCERTAINTY QUANTIFICATION TO CALIBRATE THE CRN MODELING

R. Lamioni*, A. Mariotti*, M.V. Salvetti*, C. Galletti*
rachele.lamioni@unipi.it
* Dipartimento di Ingegneria Civile e Industriale, University of Pisa, Pisa 56126, Italy

Abstract
To tackle climate change, we need to incorporate renewable energy sources on a large scale. One potential solution is the use of hydrogen, which can be produced from excess wind and solar power, as an alternative energy vector that can be burnt in existing devices with little modification. In this context, ammonia, as a hydrogen carrier, is very appealing as it can be easily liquified and distributed through the existing infrastructure.
This study aims to explore the feasibility of using ammonia-hydrogen mixtures in industrial settings. This exploration is made using a network of chemical reactors (CRN) to reproduce a gas turbine system that can improve our understanding of the energy conversion process involving hydrogen, ammonia, and their mixtures. The study aims also to propose a novel methodology to identify the most significant parameters of the CRN model and calibrate the network, leveraging uncertain quantification techniques. This will enable the development of a CRN model that can accurately predict gas turbine system emissions under varying operating conditions, such as pressure, temperature, equivalence ratio, and mixture.

Introduction

The quest for cleaner gas turbine combustion technologies has gained traction in recent years. One of the potential solutions is the adoption of premixed combustion of ammonia-hydrogen blends; this promises to reduce carbon emissions while maintaining high combustion efficiency. Indeed, the use of NH_3-H_2 blends seems to be effective in overcoming the drawbacks related to these green energy vectors. Indeed, ammonia combustion presents some challenges, including low reactivity and high NOx emissions compared to conventional hydrocarbon fuels. On the other hand, hydrogen has a higher reactivity with high flame speeds which can favor the flashback phenomenon. The challenge, therefore, is to develop combustion technologies suitable for NH3, H2, and their blends. Previous experimental and numerical works have demonstrated the potential of NH_3-H_2 mixtures in reducing NOx emissions in rich conditions (ϕ>1) [1]. For instance, Pug et al. [2] showed that multistage NH3 combustion, where rich and lean combustion is performed sequentially, results in lower NO emissions. However, studying new fuel mixtures

through experiments or full-scale numerical simulations is prohibitively expensive due to the need to analyze a wide range of operating conditions, such as mixture, equivalence ratio, injection pressure, and temperature as well as the possibility to devise novel geometrical configurations. To address this problem, simplified-order models such as chemical reactor networks (CRNs) can represent a valuable tool for quickly analyzing a broad range of operational parameters. CRN models have already been used to characterize emissions in combustion processes and are useful as detailed kinetic simulations of practical systems [3-4]
In this study, we use Uncertainty Quantification (UQ) techniques to calibrate the parameters of a CRN model emulating a premixed gas turbine system operating with NH_3-H_2 blends. Such techniques are necessary since the behavior of these new mixtures in the operating conditions of gas turbines is not known. The NOx emissions predicted through the kinetic simulations of the CRN model will be compared with the experimental data of the system. The calibrated model, with quantified uncertainty, can then be employed to study a broad spectrum of operating conditions and identify a robust optimum operating point that minimizes pollutant emissions.

Case study

The case study is the high-pressure generic swirl burner (HPGSB), housed within the high-pressure optical chamber (HPOC) [5]. The HPGSB uses quartz windows and a cylindrical quartz burner confinement to provide optical access to the flame. The HPGSB is modular and can be operated with a wide range of swirl numbers. Previous works [5-6] include further information on the design and operation of this high-pressure combustion rig test facility.

Chemical Reactor Network model

The CRN model comprises a Perfectly Stirred Reactor (PSR) [7] and a Plug Flow Reactor (PFR) [8]. The PSR represents the primary combustion zone, while the PFR mimics the secondary combustion zone.
Figure 1 shows a schematic diagram of the CRN model constructed using five reactors: one mixer into which our blend enters, three PSRs that represent the flame zone, and two recirculation zones due to the swirl burner configuration. Finally, the last reactor characterizes the uniform zone of the system (PFR).

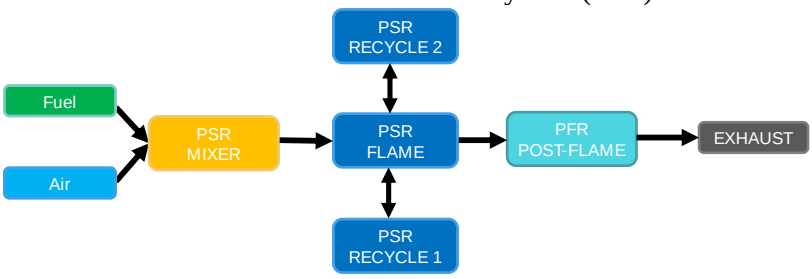

Figure 1: Chemical Reactor Network.

The CRN code is written in Python using Cantera [9], an open-source software for thermodynamics and reaction kinetics.
The CRN is tested against the experimental data from the Gas Turbine Research Center of Cardiff University that refer to a 50%H_2-50%NH_3 [5] blend with an equivalence ratio of $\phi = 0.52$ and atmospheric pressure. In the CRN we use the kinetic mechanism from Stagni et al. [10] with 31 species and 203 reversible reactions. The reactor volumes represent a fundamental parameter for a correct estimate of the emissions in terms of NOx as the volumes impact the residence time in each zone of the system. Hence, uncertainty quantification is employed here to help identify these volumes by calibrating the CRN. More specifically, three ratios between the volumes of the reactors involved are considered:

- V_{PSR}/ V_{PFR}, with the PSR volume represented by the contributions of the flame (V_f) and two recirculation (V_{r1} and V_{r2}) reactors. $V_{PSR} = V_f + V_{r1} + V_{r1}$.
- V_{r1}/V_f, the ratio between the first recirculation and the flame volumes reactors.
- V_{r2}/V_f the ratio between the second recirculation and the flame volumes reactors.

To quantify NO_x emissions from the combustion process, the molar fractions of NO and NO_2 are measured in the PFR reactor.

Uncertainty Quantification procedure

A stochastic sensitivity analysis is carried out, using the generalized Polynomial Chaos (gPC) in its non-intrusive form, to investigate how the reactor volumes of the CRN affect the NOx emissions. This analysis allows us to obtain continuous and accurate response surfaces in the parameter space, with an affordable number of deterministic simulations.
In the gPC approach, the dependence between a quantity of interest, $Z(\varepsilon)$, and the vector of independent uncertain parameters, $\boldsymbol{\zeta}(\varepsilon)$, is expressed using of a polynomial expansion [11]. The uncertain parameters are considered as random variables with a given probability density function (PDF). So, employing term-base indexing:

$$Z(\varepsilon) = \sum_{j=0}^{\infty} b_j \Psi_j \left(\boldsymbol{\zeta}(\varepsilon)\right)$$

where ε is an aleatory event, $\Psi_j\left(\boldsymbol{\zeta}(\varepsilon)\right)$is the j-th gPC polynomial and b_j is the corresponding projection coefficient.

The response surface is truncated to the limit O that can be calculated as follows:

$$O = \prod_{k=1}^{N} (T_k + 1) - 1$$

where N is the dimension of $\boldsymbol{\zeta}(\varepsilon)$, the index k identifies the k-th element of that vector, and T_k is the maximum order of the corresponding polynomial. The coefficients b_j of the expansion can be computed as follows:

$$b_j = \frac{\langle Z, \Psi_j \rangle}{\langle \Psi_j, \Psi_j \rangle} = \frac{1}{\langle \Psi_j, \Psi_j \rangle} + \int_\zeta Z\, \Psi_j \omega(\zeta) d\zeta$$

where $\omega(\zeta)$ is the weight function connected to $\Psi_j(\zeta)$. In this work, the integral above is approximated using a Gaussian quadrature formula and the optimal polynomial family Ψ_j is related to the shape of the PDFs of the uncertain parameters. Since we assumed uniform PDFs, Legendre polynomials are used. The same range of variation is selected for the three parameters, i.e. [1/5,5]. The total volume of the gas turbine system is $V_{TOT} = V_f + V_{r1} + V_{r2} + V_{PFR} = V_{PSR} + V_{PFR}$. The polynomial expansion is truncated to the 3rd order in each dimension. Thus, 4 quadrature points are needed for each variable (Gauss-Legendre points), whose values are summarized in **Table 1** and, using a tensor grid 64 simulations are carried out to evaluate the stochastic response surfaces.
The variability of the output quantities is described in terms of total variance as $\sigma^2 = \sum_{j=1}(b_j^*)^2$,with $b_j^* = b_j|\Psi_j|$, $|\Psi_j|$ being the norm of the j-th polynomial. The partial sensitivity of the quantities of interest to a single input parameter or a combination of them is computed using the variance decomposition method proposed by [12]. The related Sobol indexes I_i is defined as the ratio between the partial variances σ_i^2 i.e., the variance only due to the i-th uncertain input parameter, and the total variance σ^2, as $I_i = \sigma_i^2 / \sigma^2$.

Table 1: Quadrature points for the three input parameters.

Quadrature Points	1st	2nd	3rd	4th
V_{PSR}/V_{PFR}	0.534	1.786	3.414	4.666
V_{r1}/V_f	0.534	1.786	3.414	4.666
V_{r2}/V_f	0.534	1.786	3.414	4.666

Results: effect of the PSRs and PFR volumes
Figure 2 shows the emissions in terms of NOx predicted by the 64 CRN simulations with different flame (PSR) volume values. The black dashed line is the experimental data for the test case considered with 50%H_2 and 50%NH_3 ($\phi = 0.52$, $p = 1.0$ atm and $T = 288$ K). With $V_f = 0.13\ V_{tot}$ the estimated NOx value agrees with the experimental data, which corresponds to 7 ms of the flame residence time.

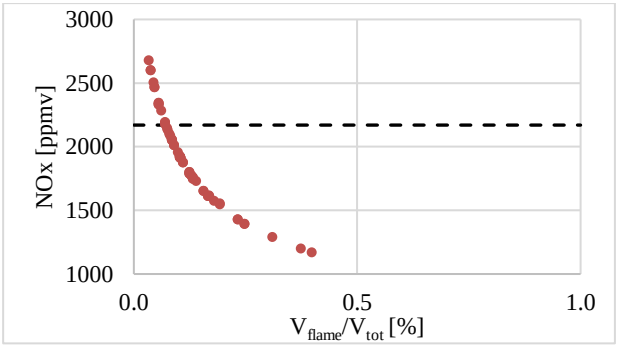

Figure 2: NO_x emissions with respect to the PSR (flame) residence time compared with experimental data from [3] (dashed line).

Errore. L'origine riferimento non è stata trovata. shows the Sobol indices representing the sensitivity to the choice of PSR and PFR volumes on the prediction of NOx emissions. The contributions of the interaction between the three selected parameters (the ratios V_{r1}/V_f, V_{PSR}/V_{PFR}, and V_{r2}/V_f) are negligible (<1%). The greatest contribution (43%) is given by the A/B ratio which represents the ratio between the volume of PSR and PFR. This result confirms that to have a good estimate of NOx emissions it is essential to correctly estimate the residence times, in particular, the one of the PSR of the flame as can be seen from the equal importance of the ratios V_{r1}/V_f and V_{r2}/V_f (28%).

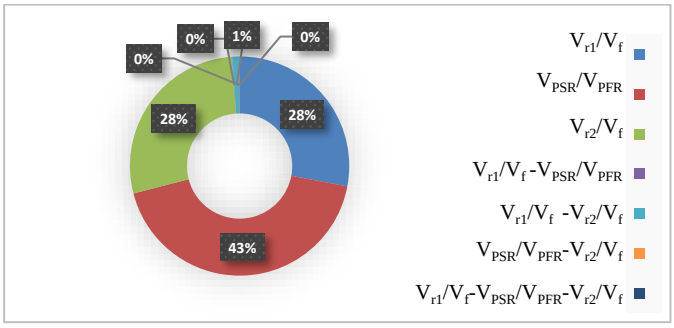

Figure 3: Sobol indexes for NOx emissions.

Conclusion

In this work, a chemical reactor network is developed and implemented to emulate a gas turbine system fueled with an ammonia-hydrogen mixture using the Cantera software. The uncertainty quantification techniques are used for the network calibration, by assessing in detail the effect of the volume of each individual reactor. Indeed, the volumes are a fundamental parameter affecting the correct estimation of NOx emissions. It is found that the parameter which has the main impact is the volume of the flame zone.

This work provides a first step for a CRN model that can be further employed to

devise combustor geometry modification, as well as to evaluate the effect of operating parameters such as temperature, pressure, and mixture composition on combustion efficiency and emissions.

Acknowledgment

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