

CFD-CRN Modelling for Prediction of Pollutants in NH₃/H₂/Air Combustion

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Abstract

In the pursuit of a renewable, clean, highly-efficient fuel, hydrogen proves to be a promising alternative. But due to high diffusivity and low compressibility, the storage and transport of hydrogen are difficult. To address such problems, ammonia, which itself is a clean fuel, can be blended with hydrogen. This additionally solves the problem of low flame speed and flammability of ammonia as a fuel. In the present study, an experimental analysis of the emissions of a premixed swirling stoichiometric combustion of an 80/20 (by mole) mixture of ammonia and hydrogen is discussed. Such a discussion is important since a nitrogen-based fuel could be accompanied by large NO_x emissions. A methodology of coupled Computational Fluid Dynamics (CFD) and Chemical Reactor Network (CRN) has been discussed, which decouples the detailed chemistry from the hydrodynamics of the flow and hence, makes it computationally feasible to easily predict pollutant concentrations. A 2D axisymmetric CFD simulation followed by a CRN analysis, with a detailed reaction mechanism, has been performed. The CFD-CRN procedure described in the present study is able to replicate the NO concentration trend depicted in experimental results, at a minimal computational cost.

Keywords: CRN, Pollutants, Ammonia, Hydrogen, Combustion Simulation, Swirl

1. Introduction

While wind, solar and hydroelectric energies are established technologies, it is still challenging to replace combustion as the primary source of energy. The scarcity of fossil fuels and the widespread emissions causing global warming forces us to find alternatives to carbon-based fuels, which would have cleaner combustion products. Hydrogen is an excellent

alternative but the associated storage and transport problems make it unfeasible for its direct adoption.

To address this problem, hydrogen can be converted into ammonia, a hydrogen carrier, which can be easily compressed and transported. But the consequent reconversion of ammonia to hydrogen incurs high energy inefficiencies (up to 33% [1]). So, ammonia should be directly burned where possible. However, ammonia has drawbacks such as low flame speed, high ignition energy, narrow flammability limits and low flame stability.

Enrichment of ammonia with hydrogen increases the flame speed, flame stability, as well as ease of storage and transport. Although this blend would be zero-carbon emitting, it becomes necessary to evaluate the NO_x production and thereby, find ways to reduce these emissions, which not properly tackled can be highly detrimental to the atmosphere and climate change.

Valera-Medina et al. [2] studied a 50/50 blend of ammonia-hydrogen and reported that NO_x is higher for lean combustion. Valera-Medina et al. [3] reported that 70/30 and 80/20 blends by volume of ammonia-hydrogen are most stable. Nozari et al. [4] numerically examined lean ammonia-hydrogen-air mixtures at elevated pressures and reported total NO_x formation decreases under rich conditions. However, N₂O was not dissociated from NO. Pugh et al. [5] reported that NO_x emissions are sensitive to equivalence ratio and vary across several orders of magnitude. Findings denoted that the least NO_x concentration is achieved in rich conditions, although there is a high fraction of unburnt fuel and thus, reduced combustion frequency. Further findings showed that a secondary airflow could be highly effective in reducing emissions. Rocha et al.

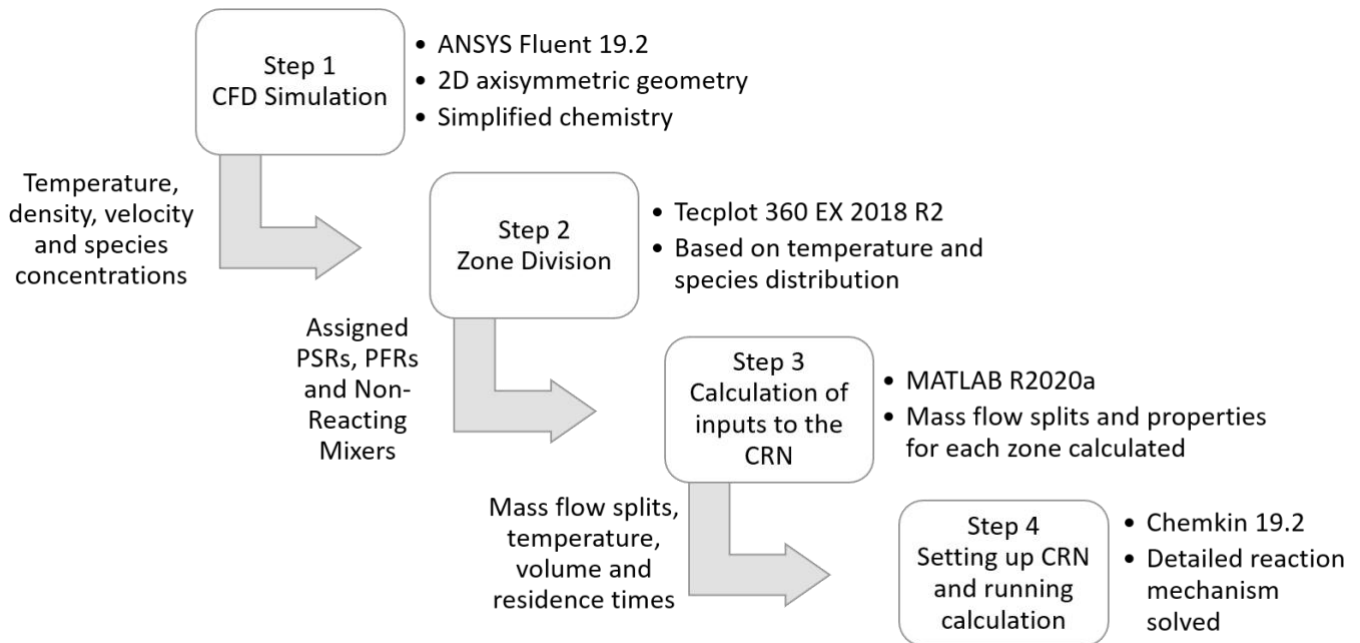


Fig. 1. CFD-CRN methodology

[6] reported that NH_3/H_2 flames have much higher NO_x than pure ammonia flames due to the generation of OH and O radicals. The NO in NH_3 flames is produced through the NH_3/O_2 chemical process. This explained the mitigation of NO at a higher equivalence ratio because the excess NH_3 in rich cases consumes NO. Mashruk et al. [7] studied various blends of ammonia/hydrogen at different equivalence ratios. Lean mixtures produce large N_2O and moderate to lean produce large NO. In the present work, the same experimental trends across different equivalence ratios discussed above are derived numerically for an 80/20 (%mol) blend.

CRNs have come to be widely used alongside CFD simulations. A brief overview of this CFD-CRN methodology is depicted in Fig. 1. A CRN involves zonal division of combustor flame field based on homogeneity (or unidirectional variation) in temperature and species distribution which is obtained from initial CFD simulations obtained using either (a) a reduced reaction mechanism and/or (b) combustion models (eg. Eddy-dissipation, FGM model etc.) that do not consider reaction rates in its calculations. This way, the initial CFD simulations are performed relatively quickly. The number of zones of CRN is usually way less than the number of cells in the CFD. For instance, a reduction of the CFD domain of thousands of cells to a few tens of properly selected zones is possible. These zones are then modelled as a combination of 0-D Perfectly Stirred Reactors (PSRs) or 1-D Plug Flow Reactors (PFRs). The CRN calculations are then run with a detailed reaction mechanism in each zone considering the reaction rates so as to predict the

pollutant concentrations in each of them. Subsequently, the pollutant concentrations can be determined at the exit of the combustor as these zones are connected in series. The fact that there are only few tens of cells (zones) in CRN as compared to thousands of cells in CFD makes it feasible to consider detailed chemistry (with reaction rates) in the CRN calculations as compared to the CFD simulations. Further, subsequent to CRN calculations, a sensitivity analysis could be performed to determine the possible pathway of pollutant formation (which is not possible with CFD simulations). In this manner, CFD-CRN is a tool for the prediction of pollutants' concentration, their pathways and rates of production.

The CFD-CRN methodology has been successfully implemented in previous works. For instance, Novosselov [8] predicted NO and CO emissions of lean-premixed industrial GT combustors and updated an eight-step global mechanism for methane with NO formation. DePape and Novosselov [9] furthermore expanded the applications to real-time CRN monitoring of combustion. Nguyen et al. [10-14] presented numerous reactor networks, for the prediction of NO_x during methane oxidation. Chen [15] applied the CRN technique to Correa-Gulati [16] and Dally et al. [17], flames, predicted pollutants and suggested a nine-step reduced mechanism for methane combustion using sensitivity analysis. There is abundant literature available about the application of CRN on methane combustion in various combustors, but not much about the same for ammonia-hydrogen combustion.

Driven by the research gap in the implementation of the CFD-CRN technique on ammonia-hydrogen blends for pollutant prediction, the present work is aimed at performing a 2D CFD simulation, build a CRN from it and run simulations with a detailed mechanism on CRN for pollutant prediction. The numerically predicted pollutant concentrations are then compared to experimentally determined figures.

2. Experimental Setup and Test Cases

The CFD-CRN technique is applied to the experimental setup of authors' previous work i.e., Mashruk et al. [7]. The swirl combustor to be simulated, as shown in Fig. 2, is a tangential

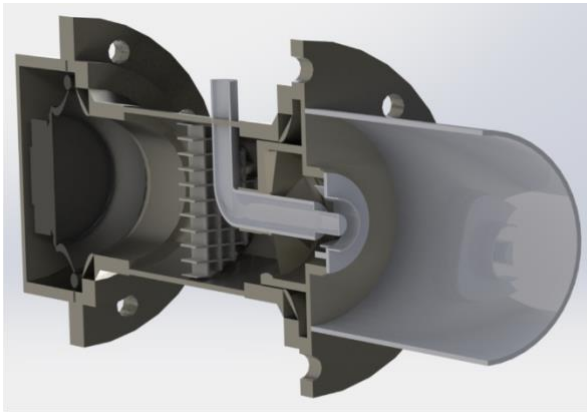


Fig. 2. Transverse cut section of 3D combustor

Air Volume Flow Rate (LPM)	Fuel Volume Flow Rate (LPM)	Equivalence Ratio (Φ)
229.3	37.69	0.55
210.2	37.69	0.6
168.2	37.69	0.75
132.8	37.69	0.95
120.1	37.69	1.05
114.7	37.69	1.1
105.1	37.69	1.2
90.1	37.69	1.4

Table 1: Air flow rates for different eq. ratio

atmospheric swirl burner [7]. Ammonia is premixed with air while hydrogen is added close to the combustion chamber. The flow rate for the fuel has been maintained constant while the air input has been controlled to vary the equivalence ratio. A swirling turbulent flow with premixed combustion has been considered, for an 80/20% (by mole) ammonia/hydrogen blend as fuel. Inlet is at the ambient condition of 288K, 1atm. The air flow rates are varied as shown in Table 1.

The ratio of tangential to axial velocity from the swirl number, to represent the swirling vanes in 2D, is calculated using the following formula:

$$S = \frac{2}{3} \frac{v_{\theta}}{v_x} \frac{1 - \left(\frac{R_i}{R_o}\right)^3}{1 - \left(\frac{R_i}{R_o}\right)^2} \quad (1)$$

Where S is the swirl number, v_{θ} is the tangential velocity, v_x is the axial velocity, R_i is the inner radius of the inlet and R_o is the outer radius of the inlet. In the present case, $S = 1.05$, $R_i = 6mm$, $R_o = 14mm$ and the axial velocity has been calculated using volume flow rate and cross-sectional area of the inlet.

2. Numerical Setup

A CFD simulation of the burner has been set up

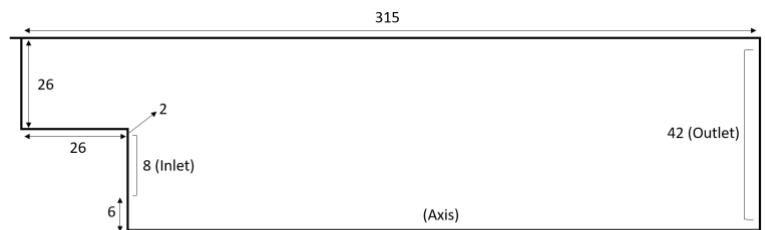


Fig. 3. 2D Axisymmetric Domain (all dimensions in mm)

in ANSYS Fluent 19.2, to simulate the experimental test cases described in the previous section using a 2D model (Fig. 3.) axisymmetric swirl case. A mesh of 80,000 cells had been found to be the optimum mesh for the study. A simplified 2D simulation efficiently cuts down on time and computational requirements. The turbulence closure has been achieved by using the RNG K-epsilon model, with standard wall treatment [18]. The partially premixed model has been used for species modelling. The Flamelet Generated Manifold (FGM) has been used with the 203 steps 31 species mechanism by Stagni et al. [19] used for flamelet generation. Since the flamelets are calculated in the reaction-progress space instead of physical space, the computations remain cheap despite the large mechanism. If finite-rate chemistry is involved, then a global mechanism or a reduced mechanism should be opted in CFD calculations in order to minimize the simulation time. The turbulence-chemistry interaction is set to Turbulent Flame Speed and the Zimont flame speed model has been used [20]. The ignition is achieved by patching the progress variable as "1" after the calculations had run with the Premixed Combustion, Pdf, Progress Variable Variance equations turned off.

From the results obtained from CFD, the velocity, temperature and density fields along with the species concentrations fields are employed for the CRN modelling which is described in the next section.

3. Results

3.1. CFD Simulations

Fig.4a depicts the temperature contour for the axisymmetric domain and Fig.4b is the magnified view of the flame zone divisions. There exist two recirculation zones: a central toroidal recirculation zone (CTRZ) and a recirculation zone in the corner of

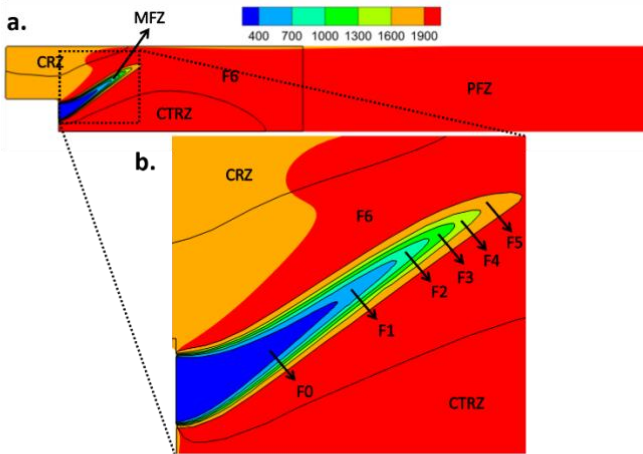


Fig.4: a.Division of zones for axisymmetric domain based on temperature contour (in K) for $\phi=1.0$ (CTRZ: Central Toroidal Recirculation Zone, CRZ: Corner Recirculation Zone, MFZ: Main Flame Zone, PFZ: Post Flame Zone) b.Close-up division of the flame zones

the combustor (CRZ), which is a common feature in a confined swirling flow, like in a swirl combustor. The CTRZ stabilizes the flame by effectively mixing the incoming reactants with hot products, thereby, matching the flow and flame velocities. Consequently,

From the CFD solution, the computational domain is divided into zones, based on homogeneity in temperature and species concentrations (Fig. 4). This division roots from the definition of Perfectly Stirred Reactors (PSRs) which have spatially uniform pressure, temperature and species concentration. So, to get a zone as close as possible to the homogeneous assumption of PSRs, the division of all the zones, except for the recirculation zone and post flame zone, is based on the temperature range. The species distributions have been found to closely follow similar distribution as the temperature. The recirculation zone is defined as the zone of axial backflow or mathematically, the region with negative axial velocity. Recirculation zones formed in the case of swirl burners are crucial for the stabilization of flame. In a CRN, these are often characterised by high residence times, large volumes and low mass flow rates. These are often responsible for the ignition as they recirculate the hot products close to the inlets.

Residence time is calculated using the following:

$$Residence\ Time = \frac{Volume}{\left(\frac{Mass\ Flow\ Rate}{Density}\right)} \quad (2)$$

The post flame zone is where the streamlines become parallel to the axis and parameters like temperature and species concentrations vary but only in the flow direction i.e., there is perfect mixing in the direction transverse to the flow. These zones are modelled as Plug Flow Reactors (PFRs).

Based on the above guidelines, the present

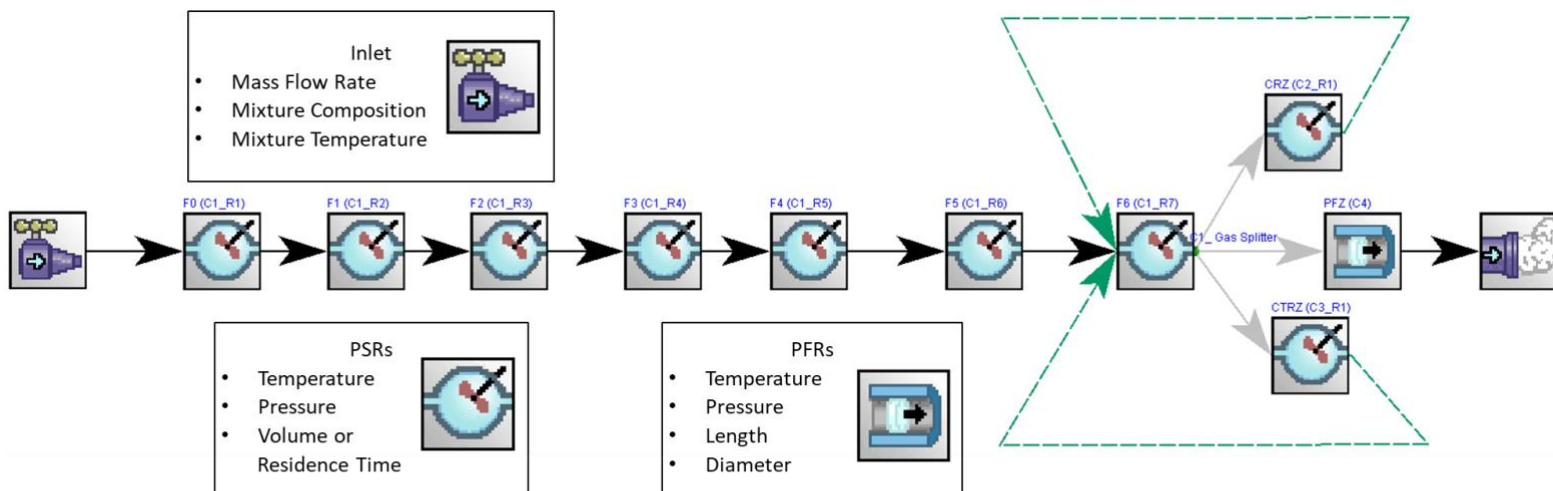


Fig. 5. CRN and required inputs in Chemkin-Pro

the flame stabilises in the shear layer formed due to the CTRZ.

flame field has been divided into 9 PSRs and 1 PFR along with an inlet and an outlet, as shown in Fig.5,

which is constructed using Chemkin-Pro 19.2. The inputs for respective reactors as shown in Fig. 5 and flow splits in-between zones are calculated. The detailed Stagni reaction mechanism [19] has been used for pre-processing. The CRN calculation is run and the NO ppmvd 15% O_2 is obtained at the end of the PFR length.

Fig. 6 compares the NO emission results from the experiment [7] to that predicted by the CRN, for different equivalence ratios for the 80/20 NH_3/H_2 blend. The optimum operating equivalence ratio is accurately predicted by the CRN to be slightly richer than the stoichiometric, in agreement with the experimental results. The sudden increase in NO in the lean regime is also predicted by the CRN. The predicted results show an increase in the NO as the equivalence ratio is decreased from stoichiometric. At $\phi = 0.75$, the NO concentrations reach a maximum, which accurately matches the experimental findings. The value of the lean equivalence ratio below which NO begins to decrease, i.e., 0.75, is accurately predicted by the CRN. The maximum deviation in predicted concentration is 45% which occurs for the lean regime. This can be reduced by taking a larger number of zones with narrower variations in temperature and by including heat loss from the walls into the CRN. Alternately, a 3D CFD simulation instead of 2D could also be employed to better predict flow and flame field accurately which eventually will lead to building of a better CRN. The NO concentrations are accurately predicted by the CRN close to the stoichiometric mixture. It is also predicted that the contribution of NO to total NO_x decreases as the mixture is made leaner, as seen from the

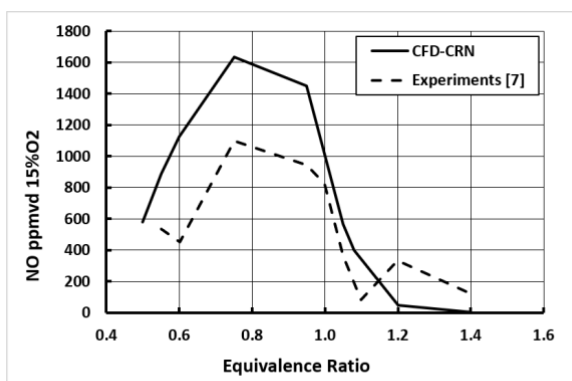


Fig. 6. Comparison of CFD-CRN results with experiments

experimental literature. The key take away point from the present analysis is that by employing a low-cost 2D CFD-CRN approach the correct trend of NO (and other pollutants) can be predicted as in the experiments (which are very expensive to perform and requires a lot of infrastructure to perform in the first place). Thus, CFD-CRN approach is shown to be a cost-effective way

to predict the correct trends in ammonia/hydrogen-air flames.

4. Conclusion

A CFD-CRN simulation has been presented as a tool for the prediction of pollutants in the combustion of ammonia hydrogen blends. The CFD is used to establish the flow field while the detailed chemistry is solved using the CRN. A tangential atmospheric swirl burner from the authors' previous work has been simulated. It predicts a slightly richer than stoichiometric mixture to be the optimum for pollutant evasion, in agreement with the experimental results for the same combustor. The prediction of the emission trends using the CFD-CRN technique saves immensely on the required time and the costs.

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