



## Full Length Article

## Dimensionally reduced modeling of nitric oxide formation for premixed methane-air flames with ammonia content



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## ABSTRACT

The paper presented an experimental and numerical study on a combustion process of ammonia doped methane flames with the  $\text{NH}_3$  content in the fuel from 1% up to 5%. Tests were performed with an atmospheric pressure axisymmetric burner for stoichiometric mixtures and fuel-lean conditions ( $\phi = 0.63, 0.71, 0.83, 1.0$ ) with the substrates preheating up to 573 K. Numerical modelling involved three reduced dimensionally combustion models (0D IdealGasReactor, 1D FreeFlame, 1D BurnerFlame) with four detailed reaction mechanisms for the hydrocarbons and nitrogen chemistry (GRI 3.0, SanDiego, Konnov 0.6 and Tian). Comparison of the 0D/1D calculations results with a complete burner geometry modelling (3D Ansys EDC) was performed for GRI 3.0.

Experimental study shown that nitric oxide emission increased with the increase of  $\text{NH}_3$  content in the introduced fuel, however transition from  $\text{NH}_3$  to NO was incomplete and trend was not linear for the rising ammonia share. Doubling of the ammonia content in the fuel from 2.5% to 5% resulted in a rise of NO emission by only 55.5% (from 946 to 1471 ppmv) for the lean mixtures and by 48% (from 1451 to 2148 ppmv) for the stoichiometric conditions.

The numerical analysis results were in a good agreement with the experimental results for the lean mixtures and ammonia content up to 1% for all investigated combustion models and the kinetic reaction mechanisms. Trends of the nitric oxide emission obtained with the stoichiometric flames were valid for 1D models. However overall values were over predicted for an adiabatic 1D FreeFlame calculations and under predicted for 1D BurnerFlame, which showed a sensitivity of the nitrogen chemistry to the process temperature. Analysis with the 3D Ansys EDC model and GRI 3.0 provided the most accurate results up to 2.5% of the  $\text{NH}_3$  share in the fuel with a maximum relative differences values to the experimental results from 10% up to 24% for the lean and the stoichiometric conditions respectively.

## 1. Introduction

Nowadays changes in the environmental policy are redirecting research activity into the field of zero- $\text{CO}_2$  emission alternative fuels. Studies on  $\text{NH}_3/\text{H}_2$  and  $\text{NH}_3/\text{CH}_4$  flames in gas turbine conditions are representative of this novel approach [1], as well as the potential of using ammonia to charge the energy storage systems [2] as their primary energy source or co-firing fuel. Previously, ammonia doped flames were mainly investigated from the perspective of NO emission from combustion of fossil fuels doped with nitrogen species, in specified conditions even with the  $\text{NH}_3$  content equivalent close to 5000 ppm [3].

Unlike sulphur compounds,  $\text{NH}_3$  is not only a source of pollution, but could also be used as a reducing agent, depending on process temperature and flow conditions. Simultaneous ammonia reduction and

oxidation processes, which are taking place in the reaction zone, are considered to have a significant impact on large scale combustion applications. Industrial experience, followed by kinetics studies, show that maintaining a temperature below 1173 K at the point of ammonia injection into the combustion chamber is essential to minimize the formation of NO from the reducing agent in the SNCR type of FG DeNOx systems [4]. However, overall NO reduction process enabled by  $\text{NH}_3$  is very sensitive to changes of the system operating parameters, which leads directly to doubts in the numerical prediction of reaction products in cases where test conditions support the  $\text{N}_2$  formation path.

A variety of kinetic reaction mechanisms involve  $\text{NH}_3$  doping in substrates. Extended nitrogen chemistry is calculated in parallel or in combination with  $\text{C}_1$ - $\text{C}_3$  hydrocarbons oxidation models, such as Konnov 0.6 [5], GRI 3.0 [6], CRECK [7] or San Diego [8] with  $\text{NO}_x$

Abbreviations: RSM, Reynolds Stress Model; EDC, Eddy Dissipation Concept; SNCR, selective non-catalytic reduction; FG DeNOx, flue gas denitrification; MOQ, minimum orthogonal quality; AEQ, average element quality

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module. These mechanisms are commonly used in a number of applications, where chemical energy of the flammable mixture is provided mostly by the hydrocarbons. Mechanisms dedicated to  $\text{NH}_3$  flames with addition of methane are placed on the opposite side of the ammonia cofiring considerations, with Tian [9] mechanism being worthy of note in the specified context.

The two-direction character of reaction paths involving ammonia could raise questions about the range of mechanisms application in the case of the  $\text{CH}_4/\text{NH}_3$  mixtures combustion. Konnov et al. [10] challenged the uncertainties of NO formation prediction near flammability limits of investigated mixtures, containing 4370 ppm of  $\text{NH}_3$ . Their results were in agreement only with the measurements of Henshaw et al. [11], who investigated fuels with ammonia content of up to 4% and identified the main reactions as  $\text{N} + \text{NO} \rightarrow \text{N}_2 + \text{O}$ ,  $\text{NH} + \text{NO} \rightarrow \text{N}_2\text{O} + \text{H}$  and  $\text{NH} + \text{NO} \rightarrow \text{N}_2 + \text{OH}$ . Skreiberg et al. [12] considered lower temperature regimes and stated, that the presence of NO promotes the reduction path  $\text{NH}_3 \rightarrow \text{NH}_2 \rightarrow \text{N}_2$  directly or through  $\text{NNH}$ , rather than the sequence  $\text{NH}_3 \rightarrow \text{NH}_2 \rightarrow \text{NH} \rightarrow \text{N}$ , which is important in flames. Tian et al. [9] proposed  $\text{NH}_3$  reaction mechanism, based on Skreiberg et al. works, for a wide range of inlet ammonia content, up to 50% of  $\text{NH}_3$  in fuel by volume, where the fuel  $\text{NO}_x$  is extracted by using argon for oxygen dilution. The mechanism shows a widening of the reaction zone with the increase of the  $\text{NH}_3/\text{CH}_4$  ratio. Moreover,  $\text{CH}_3$ ,  $\text{CH}_2$ ,  $\text{CH}_2\text{O}$ ,  $\text{NH}_2$ ,  $\text{NH}$  and  $\text{HNO}$  were marked as the key species in mixture oxidation. Pathways crucial for the NO and  $\text{N}_2$  conversion start with  $\text{NH}_3$  decomposition to  $\text{NH}_2$ , which begins the entire  $\text{HNO} \rightarrow \text{NO} \rightarrow \text{N}_2$  and  $\text{NH} \rightarrow \text{NO} \rightarrow \text{N}_2$  sequences, occurring directly or through the  $\text{N}/\text{N}_2\text{O}$  middle step. The mechanism validation by Kobayashi et al. [13] shows good result accuracy for laminar burning velocity and equilibrium calculations when evaluated with an experiment at elevated pressures, but significant over prediction of NO mole fraction was observed for the simulations of lean mixtures.

Studies on the thermo-physical properties of ammonia doped methane flames show that  $\text{NH}_3$  reacts relatively late in the lean fuel burning conditions, which in turn leads to high  $\text{NO}_x$  emissions [14]. The addition of 4% ammonia concentration resulted in a 10% to 20% decrease in the flame velocities [11]. In the pre-ignition zone, the ammonia chemistry is blocked due to the absence of free radicals, which are consumed by methane–methyl radical ( $\text{CH}_3$ ) conversion [12]. Absence of O and H radicals reduces  $\text{NH}_2$  availability to form NO by other pathways, through HNO or NH and increase the probability of forming  $\text{N}_2$  instead of NO [15].

Modelling the complex reaction kinetics in a turbulent flow as the NO reduction and the  $\text{NH}_3$  oxidation involves large amounts of computing power. Two main approaches could be specified, a kinetic reaction mechanism reduction, with a probability of lowering the calculation results accuracy and reducing dimensions of the projected domain, excluding turbulence mixing interaction influence on the computed results. Recent investigations on increasing the Konnov mechanism computation performance by reducing species and reaction amount for the gas turbine conditions were performed using 0D, 1D, and 2D approach for a fuel with 61% content of  $\text{NH}_3$ . Valera-Medina et al. [16] used equilibrium and 1D models with a result that was an under-prediction of the  $\text{NO}_x$ ,  $\text{CO}_2$ ,  $\text{O}_2$  and unburned fuel amount, with over predicting of the CO emission at the same time. They also stated that numerical 0D and 1D codes can be used for reference and the trends can be representative for further research. Barbas et al. [17] involved 1D and 2D models in numerical calculations to obtain  $\text{NO}_x$  formation characteristics simulations with detailed chemical kinetics GRI 3.0 for 1.8% ammonia content and 2D simulation results provided reasonable predictions of trends in experimentally measured  $\text{NO}_x$  emissions with the addition of ammonia.

The results accuracy of the geometric model dimension reduction vary, especially for border cases, near stoichiometric conditions and for very lean mixtures. Limited flammability and extended interaction between hydrocarbons and nitrogen compounds under these parameters

implies introducing detailed kinetic reaction mechanisms, which reduces the possibility of using complete combustion geometry for calculations. This paper provides a study on the dimensionally reduced modelling accuracy with commonly used detailed chemistry mechanisms to predict NO emission from industrial axisymmetric burner as an introduction to the investigation of low emission ammonia applications in heat and power generation for energy storage systems

## 2. Research methodology

### 2.1. Experimental setup

Experimental investigations were performed in order to evaluate the accuracy of the dimensionally reduced flames modelling for  $\text{CH}_4/\text{NH}_3$  mixtures with air as an oxidizer. Overall NO emission in the flue gases from a premixed combustion of methane and ammonia was measured. Results were presented as a function of the equivalence ratio and ammonia fuel content by volume for a constant inlet substrates temperature.

Previous investigations showed, that a percentage change of the overall NO emission as a function of substrates inlet temperature could be neglected for the ammonia doped methane axisymmetric flames for the air preheated from 373 K up to 573 K. Moreover, inlet mixture preheating resulted in a lowered ammonia slip with the flue gases and a better flame stabilization, which in effect widened the range of tested equivalence ratios.

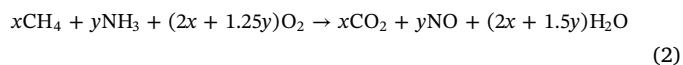
An axisymmetric, atmospheric pressure burner, with inner diameter of 25 mm was placed in a quartz glass tube combustion chamber with a diameter of 75 mm. The premixed flame was stabilized by a hot wall. The outer ceramic combustion chamber was insulated to prevent the influence of a major heat flux to the environment on the NO formation and approximate measurements to the adiabatic state.

The flue gas composition was measured in the flue gas duct located before the cooling system using a set of gas analyzers (Emerson Rosemount) NGA 2000 with the following sensors: infrared ( $\text{CO}$ ,  $\text{CO}_2$ ), chemiluminescence (NO) and paramagnetic ( $\text{O}_2$ ). The linearity for the NO, CO and  $\text{O}_2$  analysers was  $\pm 1\%$  of full scale, while the uncertainty in the calibration gas concentration used for the calibration of gas analyzers was  $\pm 2\%$ . However, this can be misleading since the same set of calibration gases is used for all tests. The measured emission values were recalculated for 0%  $\text{O}_2$  concentration in the exhaust gas using the formula (Formula 1):

$$\text{NO}_{0\% \text{O}_2} = X_{\text{NO}} \cdot [20.9 / (20.9 - X_{\text{O}_2})] \quad (1)$$

where  $X_{\text{NO}}$  is the measured fraction of nitrogen oxide;  $\text{NO}_{0\% \text{O}_2}$  is the recalculated nitrogen oxide mole fraction at 0% of oxygen, and  $X_{\text{O}_2}$  is the measured  $\text{O}_2$  mole fraction as a percentage of the flue gases.

Flow conditions during the test were kept at a stable level, due to a constant mean jet velocity. The oxygen amount for the each test was calculated using the formula (Formula 2):



The mass flow rate of methane, ammonia as well as air were controlled by Brooks Mass Flow controllers SLA series, with an accuracy of 0.2% of full scale. Preliminary test was performed for pure  $\text{CH}_4$  as a reference fuel, while other tests were carried out for a mixture of  $\text{CH}_4$  and  $\text{NH}_3$  (Table 1). The methane and ammonia were mixed with air about 150 mm from the nozzle exit. The substrates temperature was measured one diameter upstream using 1 mm thermocouple type J with a measuring range of up to 1020 K and an accuracy of 1%. Experimental variables matrix is given below (Table 1), as well as the scheme of test rig (Fig. 1).

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