A computational study of ammonia combustion enhancement under non-thermal plasma

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Abstract: An inhouse 0D plasma assisted combustion (PAC) solver is developed to study ammonia/air combustion enhancement under non-thermal plasma. Uncertainties of the reaction kinetics due to a lack of experimental data for validation are examined by comparing different mechanisms used in the literature. A two-stage simulation is conducted by transferring the output from the 0D model as the input of a 1D flame using modified combustion mechanism.

Keywords: Plasma assisted combustion, carbon-free fuel, chemical kinetic mechanism.

1. Introduction

As a promising future green fuel, ammonia (NH_3) suffers from low flammability and high NOx emission [1]. Very recently, few numerical and experimental studies have shown great potential of using non-thermal plasma produced by nanosecond discharges (NSD) to reduce ignition delay time, extend lean blowoff (LBO) limits and to reduce NOx emission [2-4]. However, enhancement pathway for ammonia combustion remains largely unknown. The paucity of experimental data also impedes reliable computational studies which heavily depend on validated NH_3 plasma kinetics and N_xH_y related cross section data. There could be large uncertainties in published 0D PAC computational studies which usually assembled plasma mechanisms from dispersed sources.

This work is aimed at revealing uncertainties of ammonia PAC mechanism used in several 0D publications, calling for a collective effort in plasma kinetics development from experimental side. A two-stage simulation method is also proposed to conduct preliminary investigations of ammonia flame behavior under non-thermal plasma.

2. Methodology

A 0D PAC solver with a Python interface is developed by incorporating plasma kinetics solver ZDPlasKin [5] and the combustion chemical kinetics solver Cantera [6] using operator splitting method. The rate constants of electron impact reactions are solved by the Boltzmann equation solver BOLSIG+ [7] incorporated in ZDPlasKin. The 0D PAC solver uses an adaptative splitting time step range from $\sim 10^{-13}$ s to 10^{-7} s to balance accuracy and cost. The solver is validated against experimental data from Mao et al [8].

The plasma kinetic mechanism used in this work mainly incorporates updated NH_3/air plasma model from Faingold et al. [9] and NH_3/air combustion mechanism from Han et al. [10] which includes reactions of excited species such as $O_2(a^1\Delta_g)$ and $O(1D)$. Latest experimental work done by Bang et al. [11] on plasma assisted NH_3 cracking is referred to modify several reactions in NH_3 plasma mechanism.

To the authors' best knowledge, all 0D computational studies on plasma assisted ammonia combustion in the literature built their NH_3 plasma mechanisms based on Faingold and Lefkowitz [3]. The current work shows large uncertainties in the PAC mechanisms by comparing 0D results from mechanisms provided by available publications [4, 12] , as well as the mechanism used in this work. Temporal evolution of electron, key excited species and ignition delay time are compared.

A two-stage simulation is conducted in the following way. Firstly, fresh NH_3/air mixture is assumed to pass through a homogeneous plasma channel and experience certain number of NSD based on residence time, thus making 0D modelling possible. Secondly, the reformed mixture composition is examined, excluding all charged species and exited sate species with negligible concentration. Key excited species such as $O_2(a^1\Delta_g)$ are kept and their related plasma reactions are incorporated into a selected combustion mechanism, enabling their roles to be played in combustion process. Thirdly, the filtered output of 0D model is used as 1D or multi-D combustion simulations.

3. Partial Results

Fig.1 compares the temporal evolution of electron and $O_2(a^1\Delta_g)$ densities based on two different PAC mechanisms of NH_3/air. The NSD has a frequency of 33 kHz with fixed reduced electric field of 150 Td. Each pulse is terminated according to the designated discharge energy per pulse which is set to 0.5 mJ/cm^3. The initial gas temperature is 800 K and plasma gas heating is neglected since the total amount of deposited plasma energy is small. The black curves (referred as Taneja 2022) represent results from running our 0D solver with input mechanism files provided as supplementary materials in Taneja et al. [4].

Significant differences are observed. For electron number density evolution, Taneja 2022 has higher peak estimation than this work by an order of two. Taneja 2022 also presents a much slower electron recombination rate. On the other hand, $O_2(a^1\Delta_g)$ concentration in Taneja 2022 shows non-monotonic behaviour and quickly vanishes after 10 pulses when NSD is off. On the contrary, a steadily incremental accumulation of $O_2(a^1\Delta_g)$ is observed in current work and stays after the 10 pulses in the plotting range.

Such uncertainty may affect conclusions on ignition delay time prediction and plasma enhancement pathway analysis, significantly.

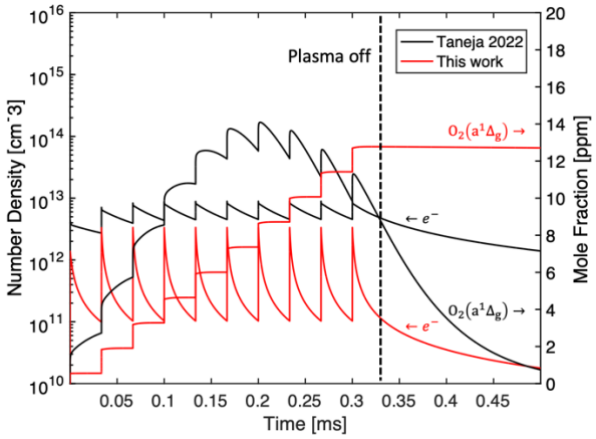

Fig. 1. Temporal evolution of electron and $O_2(a^1\Delta_g)$ for 10 pulses. Red curves: based on mechanism compiled in this work. Back curves: based on mechanism files provided by Taneja et al. [4] Dash line: plasma stops.

4. Comment

Large uncertainties of NH_3 plasma kinetic make it elusive to conduct valid modelling of plasma assisted ammonia combustion even in simple 0D reactors. Therefore, any conclusion based on unvalidated PAC mechanisms should be taken with caution. Computational studies are still important at this early stage of plasma assisted ammonia combustion to provide valuable insights into this promising topic and will evolve its accuracy and capability when more experimental studies come out.

A two-stage simulation technique can provide a feasible way to do computational study for 1D or multi-D PAC geometries provided that plasma channel can be simplified, considered as homogeneous and separated from the flame reaction zone. In this case, plasma assistance serves more like a reforming process of fresh gas. A fully coupled PAC solver higher than 0D with spatial variation is necessary for plasma/flame coupling process.

5. References

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