

Abstract

The critical greenhouse gas CO₂ accounts for 30% of global warming, and demands decarbonized energy sectors to minimize the consumption of carbon-based fuels. Renewable fuels, such as ammonia, hydrogen, dimethyl ether, and biodiesel are proving to be promising candidates for replacing fossil-based hydrocarbon fuels. The potential of ammonia as a green fuel has recently caught the attention of several organizations. Hydrogen is also a candidate for green power production. However, difficulties associated with hydrogen storage, distribution, and transportation are significant drawbacks of its wide-scale adoption. Whereas, benefits of ammonia in terms of storage and transportation over hydrogen make it a suitable alternative for gas turbine and internal combustion engine applications. The energy density of ammonia is close to diesel and gasoline, making it a promising renewable fuel for applications requiring these hydrocarbons. However, its wide adoption is hindered by slower reaction kinetics and higher NO_x emissions from ammonia combustion. Fuels such as hydrogen or methane are added as ignition promoters to enhance its ammonia combustion characteristics.

In computational simulations, accuracy significantly depends on the detailed reaction mechanism. Even though there have been extensive studies on ammonia oxidation kinetics, the available reaction model yields inconsistent results for ignition delay time and laminar burning velocity for NH₃/air and hydrogen added ammonia flames, particularly for auto-ignition characteristics at low-intermediate temperatures and high-pressure conditions.

In the first objective of this thesis, we have developed a new reaction model which can effectively predict the autoignition characteristics of NH₃/air mixture at a low-intermediate temperature and high-pressure conditions. The newly proposed reaction model consists of 32 species and 259 reactions. The developed model shows a good prediction of ignition delay time at low-intermediate temperature and high-pressure conditions compared to available models. To further check the accuracy of the developed reaction model, CFD simulations are also performed using the present developed model, and the computational results using the developed model closely follows the experimental results. Furthermore, the model also shows a good prediction of laminar burning velocity at atmospheric temperature and higher pressure up to 5 bar. The chemical kinetic modeling shows that the reaction subsets of NH₂ and H₂NO subspecies profoundly affect the autoignition behavior of ammonia/air mixture at a low-intermediate temperature and high-pressure conditions.

In the second objective, a new reaction model is developed based on the available literature for NH₃/H₂/air mixture by keeping the number of species and the reactions the same.

Extensive model validation is performed for laminar burning velocity and ignition delay time at different conditions using the newly developed model. The model shows good prediction with experimental results and closely follows the experimental trends compared to available reaction models. The chemical kinetics results show that the increase in hydrogen enrichment increases the intensity of the chain branching reaction $O_2 + H \rightarrow OH + O$ and chain propagation reaction $OH + H_2 \rightarrow H + H_2O$ and produces more H, OH, and O radicals. The increase in hydrogen enrichment increases NO_x emissions. At higher initial pressure, the NO emission decreases due to the NO consumption through the reactions $N_2O + NO \rightarrow NO_2 + N_2$ and $NO + HO_2 \rightarrow NO_2 + H$.

In the third objective, we studied the possibility of applying dual oxidizers for NH₃/CH₄ blends. A significant increase in laminar burning velocity, CO, and NO emissions is observed with increased air replacement percentages with H₂O₂. Furthermore, a significant decrease in ignition delay time (by a factor of 100) is also observed. The concentration of OH and HO₂ radicals together is mainly responsible for the laminar burning velocity variation at air replacement percentages higher than 20. However, OH radicals mainly govern the burning velocity at an air replacement percentage of less than 20. A linear correlation is observed between the laminar burning velocity variation and peak mole fraction of OH +HO₂ radicals. Intermediate radicals HNO and HCO closely govern the NO and CO production.

In the fourth objective, experimental and computational study is performed to understand the combustion and emissions characteristics of a NH₃/CH₄-air fueled self-recuperative high aspect ratio annular tubular porous burner for gas turbine power applications. In this burner, an annular zone surrounds the combustion chamber to preheat the incoming air, while two perforated discs are mounted between the combustor and annular zone for uniform distribution of air. Also, eight Zirconia foams (4 of 10 PPI and 4 of 20 PPI) are stacked at the upstream side of the combustor. Results show that the porous foam inclusion significantly reduces the mass fraction of OH radicals exiting the combustor compared to without porous cases. The insertion of porous foams shows better recirculation of heat, and stable combustion is observed for higher percentages of ammonia and higher thermal inputs. The NO and CO emissions are lower for the stack combination of 10+20 PPI compared to the stack of 10 PPI.

Keywords: *Ammonia based fuels, chemical kinetics, CFD, NO_x, CO, CO₂, porous combustion, gas turbines.*