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# Investigation of combustion behavior of ammonia with hydrogen and methane through numerical simulations

Muhammad Saqib Sahito[1,*], Suhail Ahmed Soomro[2], Masroor Abro[2]

[1] *Energy System Engineering Department*

[2] *Chemical Engineering Department*

*Mehran University of Engineering & Technology, Jamshoro*

[*] *Corresponding Author E-mail: saqib.sahito1@gmail.com*

**ABSTRACT**

Ammonia/hydrogen mixture fuel is viable option to be utilized in the vehicles. But the in-situ production of hydrogen from ammonia is a challenge along with its proper combustion and controlled emissions in NOx form. This project aims to study the basic combustion characteristics including the laminar burning velocity, ignition delay period, quenching distance and combustible limit of $NH_3/H_2$ and $NH_3/CH_4$ mixed gases fuel. The influence factors and changing rules of the laminar burning velocity, ignition delay period and quenching distance of the $NH_3$ and $H_2/CH_4$ mixed gas fuel under different conditions including pressure, temperature, mixture concentration and hydrogen blending ratio are researched. The 3D model of combustion chamber was developed and the concerned governing equations like Navier-stoke equation, energy equation and species transport equation were solved. It was observed that $NH_3$ combustion is poor in pure environment but with addition of hydrogen or methane it gets improved along with release of high amount of energy.

**Keywords:**

Ammonia
Combustion
CFD

## 1. Introduction

Fossil fuels possess very useful properties not shared by non-conventional energy sources that have made them popular during the last century. Unfortunately, fossil fuels are not renewable and are depleting rapidly. In addition, the pollutants emitted by fossil energy systems (e.g., CO, $CO_2$, CnHm, SOx, NOx, radioactivity, heavy metals, ashes, etc.) are greater and more damaging than those that might be produced by a renewable energy system. Since the oil crisis of 1973, considerable progress has been made in the search for alternative energy sources.

Hydrogen energy carries, stored at low or even ambient pressures, currently seem as an alternative solution for diesel-powered engines. hydrogen energy carrier can be directly combusted within the cylinder, i.e. methanol [1], or used as a carrier medium which is dehydrogenated before combustion [2]. The research assesses the benefits and limitations of ammonia ($NH_3$), an energy carrier with high gravimetric hydrogen density, as an alternative fuel for compression ignition engines.

Ammonia is an alkaline, colorless gas with a sharp pungent odour. The chemical's boiling and freezing points are 240 K and 195.5 K, respectively, with a density of 0.73 kg/m3 and autoignition temperature of 924 K under atmospheric conditions [3, 4]. About 80% of the global ammonia production is used in agriculture as fertilizer. The remaining 20% is used in refrigeration, purification of water supplies, and industrial manufacture of various products including plastics, explosives, textiles, pesticides, dyes, and other chemicals [3, 5, 6]. For instance, in the textile industry, ammonia is used in the manufacture of synthetic fibres, such as nylon and rayon. Ammonia also catalyzes the production of some synthetic resins. In the rubber industry, ammonia is applied to avoid the coagulation of raw latex [7, 8]. Ammonia is widely used in the industrial production of soda ash, and nitric acid (Ostwald process). Household cleaning solutions are mild, with 5-10% ammonia dissolved in water while ammonia solutions for industrial application are in concentrations of 25% or higher and are corrosive [7, 9, 10]. Concerning safety, ammonia is potentially hazardous to inhale. However, due to its strong, pungent smell, it can be detected easily in low concentrations (0.6e53 ppm) [11], well below its harmful limits. The "Immediately Dangerous to Life or Health (IDLH)" concentration level for ammonia is 300 ppm [6]. Moreover, being lighter than air, ammonia quickly dissipates into the upper atmosphere in case of accidental leakage.

Ammonia is also being considered as a hydrogen energy carrier. It contains 17.6 wt% of hydrogen compared with 2.5 wt% in methanol [12]. The volumetric hydrogen energy density in liquid anhydrous ammonia is significantly higher than that of liquid hydrogen and other conventional liquid fuels (i.e. methanol, ethanol, gasoline) [13]. Ammonia can be easily cracked to produce hydrogen for use in fuel cells or other hydrogen applications. As an energy carrier, it is considered a carbon-free green fuel, where its direct combustion could produce water and nitrogen only at low combustion temperatures. Ammonia is used as a fuel for transport vehicles and space heating [6]. It can also be used as a direct fuel in combustion engines or alkaline/high-temperature fuel cells [14] which have high resistance against residual ammonia. Whereas low-temperature fuel cells based on acidic membranes are easily degradable by ammonia concentration as low as 0.1 ppm [6].

Mikulčić et al. (2021) [15] studied numerical study of ammonia combustion chemistry under conditions resembling industrial ones. After literature review, three mechanisms of ammonia combustion that also include carbon chemistry were used for simulation of experimental premixed swirl burner with the aim of evaluating their performance. Takahashi, Fujitani [16] constructed a two-dimensional reactor model based on reaction kinetics and heat and mass transfer forcatalytic $NH_3$ decomposition. A reactor design that minimizes the effectof endothermic reaction was investigated. Frankl, Gleis, Karmann, Prager, Wachtmeister [17] studied numerically the use of ammonia and hydrogen in a high-pressure-dual-fuel (HPDF) combustion. The main fuels (hydrogen and ammonia) were direct injected and ignited by a small amount of direct injected pilot fuel. Shu, He, Ramos, Fernandes, Costa [18] reported the ignition delay times of $NH_3/CH_4/O_2$ mixtures diluted in Ar or $Ar/N_2$ over a temperature range of 900–1100 K, pressures of 20 and 40 bar, and equivalence ratios of 0.5, 1.0, and 2.0. The results demonstrate that a higher $CH_4$ mole fraction in the fuel mixture increases its reactivity, and that the reactivity decreases with increasing the fuel-oxygen equivalence ratio.

Research on alternative fuels thereby reducing dependence on non-renewable fossil fuels has become an important research direction in the field of transportation. Ammonia has been widely studied and researched as a substitute fuel of internal combustion engine [19-23]. Ammonia is easily liquefied, thus easy for storage. Meanwhile, it can also be used as a hydrogen carrier because ammonia can be pyrolyzed into $N_2$ and $H_2$ through catalytic reactor. Research shows that liquid ammonia can store as 1.77 times of hydrogen as that of liquid hydrogen, which makes ammonia can be used on fuel-cell vehicles as a hydrogen carrier, as well as on $NH_3/H_2$ mixed gas fuel engine as a new type of fuel system [24-29]. Based on fuel thermodynamic properties, combined with thermodynamics laws and the mathematical model of otto cycle engine, E. S. Strarkman theoretically calculated the thermal efficiency, cylinder pressure, temperature, specific fuel consumption and NOx emission of a single cylinder engine respectively fueled by liquid ammonia, gaseous ammonia and n-heptane. The results prove the necessity of ammonia blending with hydrogen [30, 31].

Frigo, Gentili [32] and Comotti, Frigo [25] studied the fuel supply system of $H_2$ / $NH_3$ mixed gas fuel engine respectively. Not directly using the hydrogen tank, only liquefied ammonia is used, and hydrogen gas input is controlled by the catalytic reactor, which is kept in the working temperature by taking advantage of the waste heat of exhaust gas. The feasibility of using liquid ammonia only as a fuel system is verified by experimental contrast, while hydrogen is obtained from on-board catalytic reactor which controls liquid ammonia decomposition.

Reiter, Kong [33] from the United States studied the combustion and emission characteristics of a dual fuel system fueled by ammonia gas and diesel, confirmed the feasibility of ammonia as an internal combustion engine fuel.

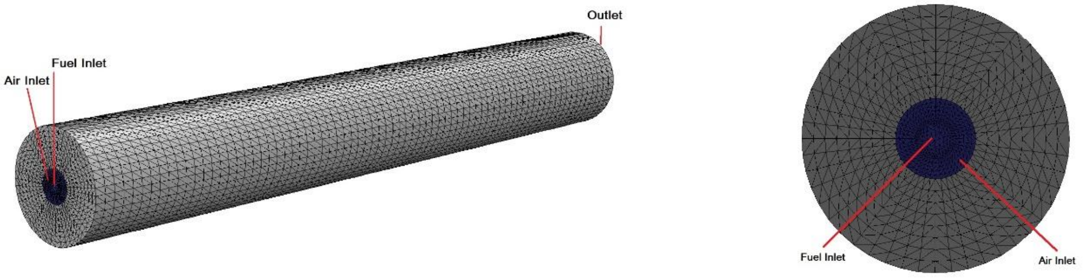
Although, ammonia has so many advantages, ammonia has poor combustion properties, slow combustion rate, low ignition limit, high ignition energy. The most effective solution is to mix ammonia with hydrogen. Hydrogen has high burning speed, low ignition energy and wide ignition limit, complementing with $NH_3$ combustion properties. On the other hand, hydrogen can be obtained by decomposition of ammonia through catalytic reactor (catalytic reactor), making full use of the advantages of ammonia liquid storage, avoid the difficult situation of hydrogen storage and transportation. $CH_4$ has also played important role when mixed with $NH_3$. This present research aims to study the basic combustion characteristics including the laminar burning velocity, ignition delay period, quenching distance and combustible limit of $NH_3/H_2$ and $NH_3/CH_4$ mixed gases fuel. The influence factors and changing rules of the laminar burning velocity, ignition delay period and quenching distance of the $NH_3/H_2$ and $NH_3/CH_4$ mixed gas fuel under different conditions including pressure, temperature, mixture concentration and hydrogen blending ratio are researched.

## 2. CFD model development

Computational Fluid Dynamic (CFD) modeling always starts with development of computational domain. In this, a grid geometry of the system used to develop with the help of advanced software. After the development of grid geometry (mesh), the concerned governing equations used to solve with numerical code and then solution is achieved. These steps are described in subsequent sub-sections.

### 2.1. Meshed Geometry of the Combustor

The geometry of the ammonia combustor was developed using Ansys DesignModeler software. The combustor is a cylindrical shaped pipe with one end has concentric inlets for fuel and air whereas other end is fully open for removal of exhaust gases. Fig. 01 shows a meshed geometry of combustor along with concentric fuel/air inlets. The important information regarding the dimensions of geometry and characteristics of mesh are tabulated in Table 01.



(a) (b)

Fig. 01: Meshed geometry of Ammonia combustor, (a) Schematic view of geometry (b) A view of concentric inlets

Table 01: Dimensions of Geometry and Characteristics of Mesh

| **Sr. No** | **Parameter** | **Value** |
|---|---|---|
| 1 | Combustor Length | 150 cm |
| 2 | Combustor Diameter | 20 cm |
| 3 | Fuel inlet Diameter | 3 cm |
| 4 | Air Inlet Diameter | 8 cm |
| 5 | Type of grid cells | Tetrahedron |
| 6 | No. of Cells | 48789 |
| 7 | Orthogonal Quality | 0.475 |

### 2.2. Governing Equations and Chemical Reactions

For solving the combustion of selected fuel mixtures, the steady state equations of mass, momentum, and energy were solved with help of commercial CFD code Ansys FLUENT. Along with these usual equations, species equations were also solved to calculate the generation and consumption of species due to chemical reactions. The flow was considered turbulent, so the turbulence was estimated using k-ε standard turbulent model. Due to combustion, high temperature is expected so the radiation transport of heat is considered and estimated with P-1 radiation model. All the selected equations are tabulated in Table 02.

### 2.3. Assumptions and Simulated Cases

In present research, steady state model computations were carried out with standard convergence criteria for the solution of mass, momentum, energy and species equations. Ammonia and hydrogen were considered pure species with standard rates of reactions. The kinetic expressions were taken from published literature.

Table 02: Selected Governing Equations

| **Equation Name** | **Mathematical Expression** | **Eq. No.** |
|---|---|---|

| | | |
|---|---|---|
| Continuity Equation | $\frac{\partial}{\partial x_i}(\rho u_{ij}) = S_m$ | (1) |
| Momentum Equation | $\frac{\partial}{\partial x_i}(\rho u_i u_j) = \rho \overline{g}_j - \frac{\partial P}{\partial x_i} + \frac{\partial}{\partial x_i}(\tau_{ij} - \rho \overline{u_i' u_j'}) + S_j$ | (2) |
| Energy Equation | $\frac{\partial}{\partial x_i}(\rho c_p u_i T) = \frac{\partial}{\partial x_i}\left(\lambda \frac{\partial T}{\partial x_i} - \rho c_p \overline{u_i' T'}\right) + \mu\Phi + S_h$ | (3) |
| Species Equation | $\frac{\partial}{\partial x_i}(\rho u_i C_j) = \frac{\partial}{\partial x_i}\left(\rho D_i \frac{\partial C_j}{\partial x_i} - \rho \overline{u_i' C_j'}\right) + S_r$ | (4) |
| Turbulence Model (*k-ε)* | $\frac{\partial}{\partial x_i}(\rho u_i k) = \frac{\partial}{\partial x_i}\left[\left(\mu + \frac{\mu_t}{\sigma_k}\right)\frac{\partial k}{\partial x_i}\right] + G_k - \rho\varepsilon$ | (5) |
| | $\frac{\partial}{\partial x_i}(\rho u_i \varepsilon) = \frac{\partial}{\partial x_i}\left[\left(\mu + \frac{\mu_t}{\sigma_\varepsilon}\right)\frac{\partial \varepsilon}{\partial x_i}\right] + C_{1\varepsilon} G_k \frac{\varepsilon}{k} - C_{2\varepsilon} G_k \frac{\varepsilon^2}{k}$ | |
| Radiation Model (P1) | $-\nabla q_r = aG - 4aG\sigma T^4$ | (6) |

Ammonia, methane and hydrogen combustion reactions are described via following equations.

$$4NH_3 + 3O_2 \rightarrow 2N_2 + 6H_2O \tag{7}$$

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O \tag{8}$$

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O \tag{9}$$

The combustion performance was evaluated by temperature profiles and fuel conversion rates. It was assumed that air contains 23 % oxygen and 77% nitrogen with respect to weight. Total 11 simulation cases were solved as per Table 03. One case is for pure ammonia, five cases for ammonia and methane mixture at various mass fractions and rest of five cases are for ammonia and hydrogen mixture at various mass fractions.

## 3. Results and discussion

The ammonia combustion was investigated numerically in pure and mixed environment with either methane or hydrogen. The results in terms of temperature of combustor and flue gases, fuel conversion and flue gas analysis were extracted and discussed in subsequent sub-sections.

### 3.1. Temperature of combustor and flue gases

The maximum inside temperature of combustor and exiting flue gas temperatures are shown in Fig. 02 for $NH_3$+$CH_4$ combustion and in Fig.03 for $NH_3$+$H_2$ combustion. The pure ammonia combustion shows a very less temperature i.e., in the range of 545 K (Fig.02) which is not very much feasible for any combustor. 30% mixing of $CH_4$ with $NH_3$ increased the temperature up to 752 K whereas 40 % $CH_4$ mixing little bit increase the temperature furthermore up to 812 K. However, the mixing of $CH_4$ with $NH_3$ 50% and more raised the combustor temperature appreciable up to 1723 to 1948 K (Fig.02). Hence the addition of 70% of $CH_4$ to $NH_3$ increased about 3.58 times the temperature as compared to pure $NH_3$ combustion.

Table 03: Feed compositions in various simulated cases.

| **Sr. No** | **Case Name** | **Total Fuel** | **$NH_3$** | **$CH_4$** | **$H_2$** | **$NH_3$** | **$CH_4$** | **$H_2$** | **Air** | **Air (20% Excess)** |
|---|---|---|---|---|---|---|---|---|---|---|
| | | Kg/sec | % | % | % | Kg/sec | Kg/sec | Kg/sec | Kg/sec | Kg/sec |
| 1 | 100A | 0.01 | 100 | 0 | 0 | 0.01 | 0 | 0 | 0.061 | 0.073 |
| 2 | 70A_30M | 0.01 | 70 | 30 | 0 | 0.007 | 0.003 | 0 | 0.096 | 0.115 |
| 3 | 60A_40M | 0.01 | 60 | 40 | 0 | 0.006 | 0.004 | 0 | 0.104 | 0.125 |
| 4 | 50A_50M | 0.01 | 50 | 50 | 0 | 0.005 | 0.005 | 0 | 0.117 | 0.14 |
| 5 | 40A_60M | 0.01 | 40 | 60 | 0 | 0.004 | 0.006 | 0 | 0.13 | 0.156 |
| 6 | 30A_70M | 0.01 | 30 | 70 | 0 | 0.003 | 0.007 | 0 | 0.139 | 0.167 |
| 7 | 70A_30H | 0.01 | 70 | 0 | 30 | 0.007 | 0 | 0.003 | 0.148 | 0.178 |
| 8 | 60A_40H | 0.01 | 60 | 0 | 40 | 0.006 | 0 | 0.004 | 0.174 | 0.209 |
| 9 | 50A_50H | 0.01 | 50 | 0 | 50 | 0.005 | 0 | 0.005 | 0.204 | 0.245 |
| 10 | 40A_60H | 0.01 | 40 | 0 | 60 | 0.004 | 0 | 0.006 | 0.235 | 0.282 |
| 11 | 30A_70H | 0.01 | 30 | 0 | 70 | 0.003 | 0 | 0.007 | 0.261 | 0.313 |

The mixing of $H_2$ with $NH_3$ has more advantageous as only 30% mixing of $H_2$ with $NH_3$ raised the temperature of combustor up to 1923 K. The maximum temperature 2424 K is achieved by mixing of 70% $H_2$ with $NH_3$. The flue gas temperature was found little less than the inside combustor temperature which is due to some heat transfer thorough walls during its travel along the length of combustor. Overall, it is seen that mixing of methane or hydrogen with ammonia put positive effect on combustion, but hydrogen has better impact over methane.

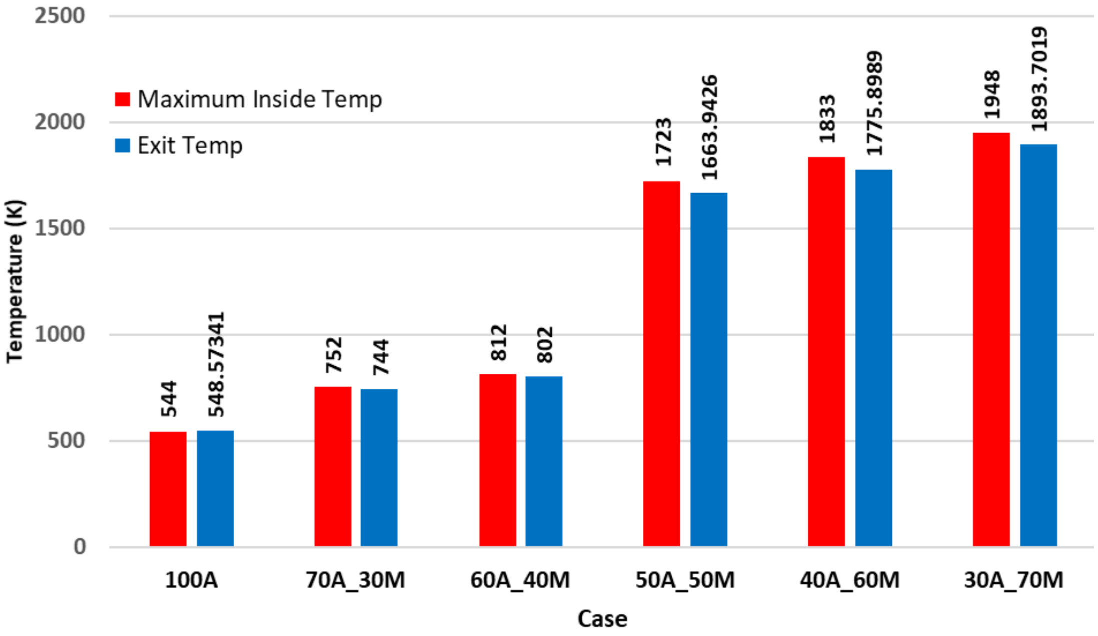


Fig. 02: Temperature of combustor and exit flue gases for various cases of $NH_3$+$CH_4$ mixing at different mixing ratios

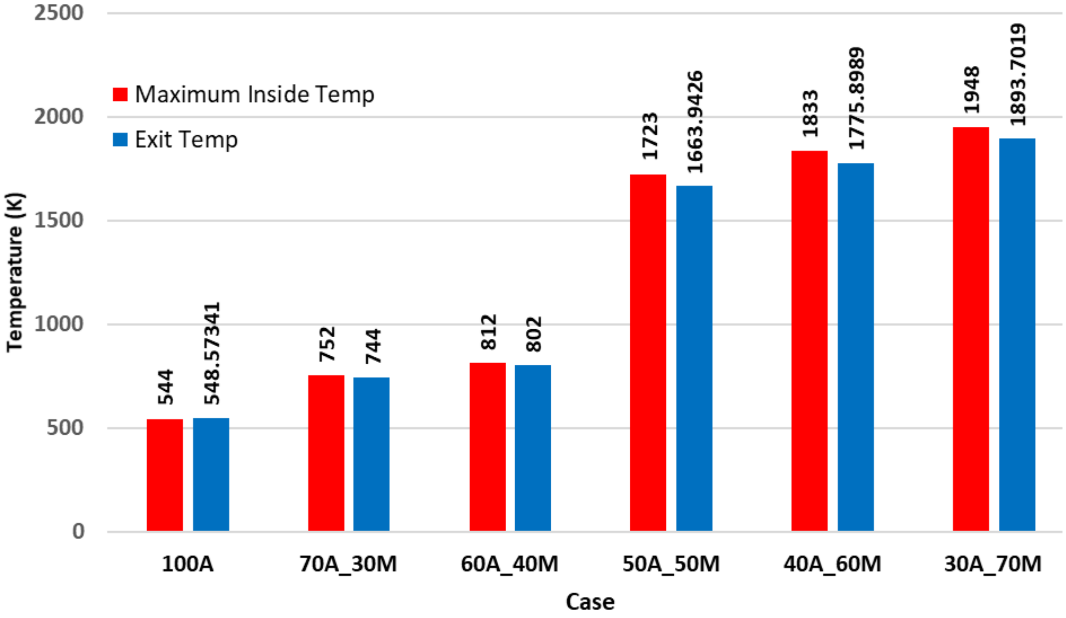


Fig. 03: Temperature of combustor and exit flue gases for various cases of $NH_3$+$H_2$ mixing at different mixing ratios

Fig.04 shows the temperature contour diagrams of all the cases of $NH_3$+$CH_4$ and $NH_3$+$H_2$ combustion, From the contours one can observe that the most of combustion completely occurs up to 20% of combustor length. The $NH_3$+$H_2$ shows a higher temperature zone as compared to $NH_3$+$CH_4$ combustion.

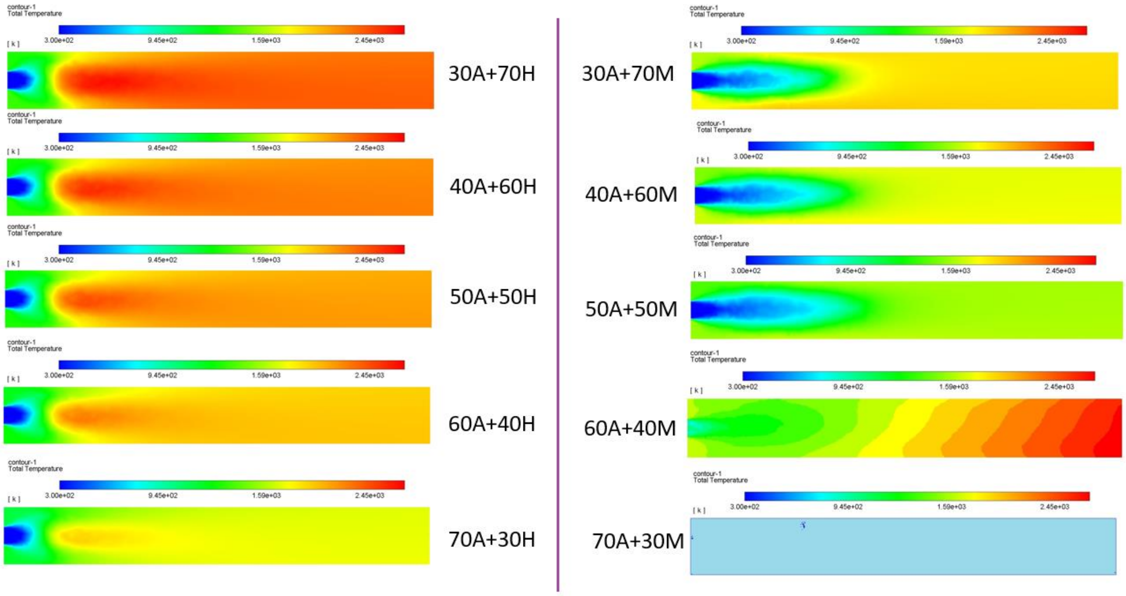


Fig. 04: Temperature contours for all the cases

### 3.2. Conversion of Fuel

The conversion of fuel was calculated for all the cases and presented in Fig. 05. From the figure it has observed that ammonia alone has lowest conversion i.e., 8% whereas mixing of $CH_4$ or $H_2$, increases the overall conversion rates of mixed fuel. Mixing of 50% to 70% either $CH_4$ or $H_2$, has almost same effect on the conversion of mixed fuel however the lesser mixing ratios for CH4 i.e., 30 or 40% has less conversion (in the range of 13 to 23%) as compared to mixing of $H_2$ with same fractions. The highest conversion was achieved 70% with mixing of either $CH_4$ or $H_2$. Overall it can be noted that H2 has better effects on conversion as compared to $CH_4$ in all mixing ratios.

### 3.3. Flue gas analysis

Flue gas analysis is also an important aspect to estimate the performance of any combustion process. The flue gas primarily contains the mass fractions of fuel species like in our case $NH_3$, $CH_4$ and $H_2$ along with $CO_2$ and $H_2O$ are important in general. The mass fractions of various important flue gas components for all solved cases are shown in Fig. 06. From the figure it is observed that unburnt ammonia is maximum when it is combusted in pure form due to its low combustibility with specified conditions. But this unburnt combustion of $NH_3$ is significantly reduced by addition of $CH_4$ and $H_2$. The $CO_2$ form the combustion of $CH_4$ (due to reaction 2) can be seen for the cases of $NH_3$ combustion mixed with $CH_4$. It has also noticeable that mixing of $H_2$ with $NH_3$ is environment friendly option as there is zero emission of $CO_2$ due to unavailability of any carbon-based fuel material.

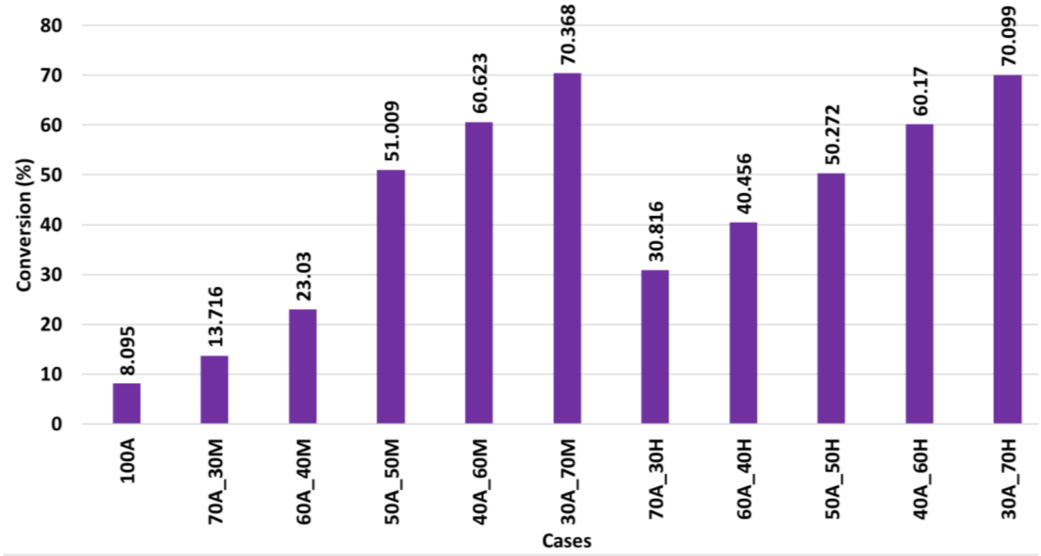


Fig. 05: Fuel conversion at different mixing rations of $NH_3$ with $CH_4/H_2$

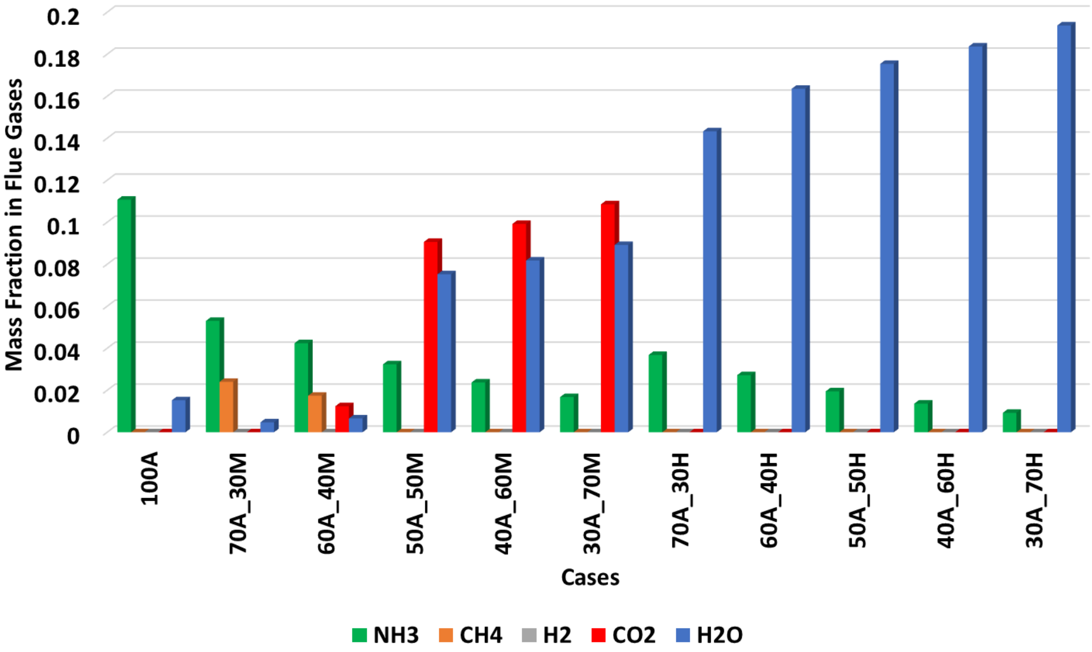


Fig. 06: Flue gas analysis for different cases

## 4. Conclusion

The combustion of ammonia ($NH_3$) was studied in its pure form and after mixing of $CH_4$ and $H_2$ individually at different mass ratios. The mixing of CH4 or H2 was carried at 30, 40, 50, 60 and 70% with $NH_3$. Numeric computations were carried out using standard CFD code Ansys FLUENT. From the research, it is concluded that 100% $NH_3$ showed very low conversion (8%) which is not feasible for any combustion system. The addition of $H_2$ or $CH_4$ with ammonia overall showed a better combustion. The maximum temperature was achieved 2424 K with 30% $NH_3$ and 70% $H_2$.

100% $NH_3$ showed minimum temperature in the combustor i.e., 544 K. 70% $H_2$ or 70% $CH_4$ addition with $NH_3$ showed maximum conversion (70%) of fuel. The mixing of both $H_2$ or $CH_4$ with $NH_3$ showed an overall better combustion in terms of exhaust temperature. $NH_3$ with $H_2$ mixture showed no $CO_2$ in flue gases and hence considered a better option for no $CO_2$ emissions. The work could be extended for comparison of experimental work with simulation results. The variation in equivalence ratio could be studied in future studies. The transient simulations could be performed to investigate the dynamic effects of the combustion.

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