Benchmarking of NH_3/CH_4 combustion mechanisms against an extensive collection of experimental data

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Abstract

Ammonia (NH_3) is a carbon-free fuel that does not directly release carbon dioxide (CO_2) when burned, making it an attractive alternative for clean energy production. This carbon-neutral property of ammonia is particularly appealing in the context of global efforts to mitigate climate change and reduce greenhouse gas emissions. However, the use of ammonia as a fuel source presents significant challenges due to its low flammability and the potential for high emissions of other pollutants, such as nitrogen oxides (NOx). Interestingly, the flame stability and combustion characteristics of NH_3 can be significantly improved by cofiring it with the highly reactive small molecule fuel, methane (CH_4). The addition of CH_4 to NH_3 combustion can improve the reactivity and overall performance of ammonia as a fuel. In this comprehensive study, a large amount of indirect experimental data on NH_3/CH_4 combustion has been collected from the literature. The data were collected in a variety of experiments, including ignition delay times from shock tube and rapid compression machine experiments, species and laminar burning velocities. The experimental data covers a wide range of conditions regarding methane/ammonia ratio, temperature, pressures, and equivalence ratio. This extensive data set provides a robust basis for evaluating the performance of different combustion mechanisms in predicting the behavior of ammonia-based fuels. The experimental data have been stored in ReSpecTh Kinetic Data (RKD) files which were used to automatically set up and run simulations of the corresponding experiments in the Optima++ framework code to assess the predictive power of the models. The performance of 16 combustion mechanisms in reproducing CH_4/NH_3 experimental data was evaluated and compared in detail. The results provide valuable insights into the current state of ammonia/methane combustion modelling and highlight the need for further development to improve the predictive capabilities of kinetic models.

Keywords: mechanism validation, Optima++, laminar burning velocity, ignition delay, jet stirred reactor

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1. Introduction

The global energy crisis and the urgent need to control greenhouse gas emissions have driven the advancement of carbon-neutral energy technologies to reduce fossil fuel consumption and CO_2 emissions. In recent years, significant research efforts [1,2] have focused on pure ammonia (NH_3) combustion. However, due to its inherently low flame speed and poor reactivity – characterized by low laminar flame velocity, short ignition delay time, and high minimum ignition energy [3] – pure NH_3 remains impractical as a standalone fuel for broad applications.

To address these challenges, extensive studies have been conducted to enhance NH_3 combustibility through blending with reactive fuels such as hydrogen (H_2)[4], diesel[5], dimethyl ether (DME)[6], and methane (CH_4). Alzueta et al. [7] recently investigated NH_3/CH_4 oxidation chemistry in a laminar flow reactor, demonstrating that rapid methane conversion via the reaction $CH_4 + NH_2 \rightarrow CH_3 + NH_3$ significantly enhances mixture reactivity. Han et al. [8]experimentally studied NH_3 premixed flames blended with H_2 and CH_4, revealing that both additives improve laminar burning velocities, albeit to varying degrees. Notably, fuel stability is a critical prerequisite for industrial applications, necessitating a comprehensive understanding of the fundamental combustion characteristics, stable combustion regimes, and underlying mechanisms of NH_3 and its blends.

Extensive research efforts have been dedicated to elucidating the combustion mechanisms of both ammonia and methane individually. However, for CH_4/NH_3 mixtures, previous studies have mainly explored limited aspects, such as simple combustion characteristics under specific conditions. There is a lack of comprehensive and systematic benchmarking effort that considers a wide range of experimental data covering various operating parameters and combustion environments. This gap in the literature highlights the need for the current research to provide a more holistic understanding of NH_3/CH_4 combustion mechanisms.

This study aims to benchmark the performance of NH_3/CH_4 combustion mechanisms against experiments under diverse combustion conditions. The study comprises the following steps: collecting experimental data and models from literature, and systematic quantitative performance evaluation of the models against the data collection.

2. Methodology

This investigation employs a quantitative comparison approach originally developed by Turányi et al. [9,10], which has been extensively validated through applications to diverse combustion systems in previous studies (see e.g. [10–14]). A key component of this methodology is the error metric, which quantifies discrepancies between computational predictions and experimental measurements. The deviation of a single simulation result (Y^{sim}) from the experimental data (Y^{exp}) should always be evaluated with the consideration of the corresponding experimental uncertainty (σ^{exp}: one standard deviation). Thus their deviation is normalized by σ^{exp} to define the uncertainty normalized signed simulation error (D):

$$D = \frac{Y^{\mathrm{sim}} - Y^{\mathrm{exp}}}{\sigma^{\mathrm{exp}}}. \qquad (1)$$

The mean square of these D error values over all data points and all data series defines the E error value, which characterizes the overall accuracy of the model on the whole data collection:

$$E = \frac{1}{N}\sum_{f=1}^{N_f}\sum_{s=1}^{N_{fs}}\frac{1}{N_{fsd}}\sum_{d=1}^{N_{fsd}} D_{fsd}^2. \qquad (2)$$

N is the number of experimental data series, and N_f is number of XML data files (containing a series of experiments), N_{fs} is the number of data series in the s-th data file, N_{fsd} is the number of data points in the s-th data series of the f-th XML data file. Each data series represents a collection of measurements for a specific physical quantity, acquired under near-identical experimental configurations with systematic variation of a single controlled condition parameter.

Experimental uncertainties are commonly presumed to adhere to a Gaussian distribution. Under this assumption, the error function E conforms to a reduced chi-square ($\sim\chi_\nu^2$) distribution. The square root of the error function (i.e. $\sqrt{E}$) corresponds to the uncertainty normalized root-mean square deviation (RMSD) of simulation from the experimental data, thus its value has an absolute meaning: it quantifies how many standard deviations of experimental uncertainty separate model predictions from experimental results on average. Ideally $\sqrt{E}\sim1$ for a perfect model, and $\sqrt{E}<2$ can be considered as an excellent model, where $\sqrt{E}<3$ corresponds to an acceptable or satisfactory model.

The validation of the mechanisms against the collected experimental data was carried out with the Optima++ simulation framework code [15] using OpenSMOKE++ [16] solver. Optima++ reads the RKD-format data files and runs the simulation tasks by invoking a selected solver. The latest version of the Optima++ code is available from the ReSpecTh site [17].

3. Experimental data collection

In this comprehensive study, a large amount of indirect experimental data on NH_3/CH_4 combustion has been collected from the literature. The data consists of ignition delay times (IDT) from shock tube (ST) [18–22] and rapid compression machine (RCM) experiments [23,24], and laminar burning velocities (LBV) [25–29]. The experimental datasets from 12

Table 1
Experimental data collection

Experiment	Ref.	N_{series}	N_{data}	x_{CH4}	p (atm)	$T_{(u)}$ (K)	φ	*dilu. ratio*	*dil./ox.*
ST – IDT	[18–22]	32 (31)	217(211)	0-100%	1.0–41.6	1181-2489	0.5-2.0	0.51-0.99	3.7-363
RCM - IDT	[23,24]	26 (25)	239(235)	0-100%	0.3–1.4	800-1300	0.5-2.0	0.7-0.8	2.92-7.94
LBV	[25–29]	36 (35)	376(354)	0-100%	1.0-5.0	298	0.213-1.50	0.382-0.765	0.996-4.88
Total		94 (91)	832 (800)	0-100%	0.3-41.6	298-2489	0.213-2.0	0.382-0.99	0.996-363

publications were encoded into 94 ReSpecTh Kinetics Data (RKD) Format Specification v2.5 XML files [30]. The summary on the size of the data collection and the covered condition ranges are shown in Table 1. Derived from the PrIMe Experimental Data Format [31], the RKD standard incorporates extended specifications to support storing of indirect measurements. These formatted files comprehensively document experimental parameters, raw observational data, and contextual metadata essential for computational validation and accurate reproduction of laboratory simulations.

4. The investigated mechanisms

Sixteen literature combustion mechanisms describing the combustion of NH_3/CH_4 mixtures were investigated in this study. Some mechanisms contained chemistry for larger hydrocarbons or oxygenated species (e.g. ethers), thus they were all reduced to contain nitrogen hydrocarbon chemistry up to C2 species. Table 2 lists all the mechanisms investigated and provides information about their size regarding the number of species and reactions before and after reduction. The mechanism identifiers were created by using the traditional short name or combining the names of the first author (with the exception of GRI-Mech) and the year of publication.

5. Results and discussion

Figure 1. show the performance of the mechanisms for laminar burning velocity measurements in stacked bar plot describing the distribution of pointwise errors. All mechanisms give predict more than 50% of the LBV data with more than $4\sigma^{exp}$ error. The best performing mechanisms are the X.Y.Zhang-2023, S.K Zhou-2023, and the R.Li-2019. Very bad performance is shown by the N. Lamoureux-2016, the GRI-Mech 3.0-2011, the P. Glarborg-2018 mechanisms, which could predict less 20% of the data within $3\sigma^{exp}$ error.

Figure 2. shows the performance of the mechanisms in predicting shock tube ignition delay time data. None of the mechanisms can predict 50% of the experimental data within $3\sigma^{exp}$ error. Best performance is shown by the J.C. Liu-2023 mechanism, followed by the Z.H. Wang-2021, P.Glarborg-2018, R.Li-2019, S.Burke-2014, S.Arunthanayothin-2021 models.

Table 2
Investigated literature mechanisms for NH_3/CH_4 combustion

#	Mechanism ID	N_{spec}	N_{spec}[1]	N_{reac}	N_{reac}[1]	Ref.
1	P.Glarborg-2018	151	151	1397	1397	[32]
2	R. Li-2019	128	128	957	957	[33]
3	X. Y. Zhang-2023	152	151	1388	1385	[34]
4	N. Lamoureux-2016	123	66	934	519	[35]
5	S.K. Zhou-2023	169	169	1268	1268	[36]
6	S.Arunthanayothin-2021	155	100	2426	1054	[37]
7	X.L.Han-2020	69	53	312	243	[38]
8	GRI-Mech 3.0-2011	53	51	325	311	[39]
9	Z.H. Wang-2021	91	75	444	375	[40]
10	J.C.Liu-2023	170	127	1207	889	[41]
11	X.Y. He-2023	180	177	1406	1406	[42]
12	S. Burke2014	156	101	2437	1065	[43]
13	E.C. Okafor-2017	59	57	356	342	[44]
14	K.P. Shrestha-2025	291	166	2405	1622	[45]
15	J.X. Ren-2025	49	49	278	278	[46]
16	M.V. Manna-2024	175	114	1394	1122	[47]

[1] Including chemistry only up to C2 species.

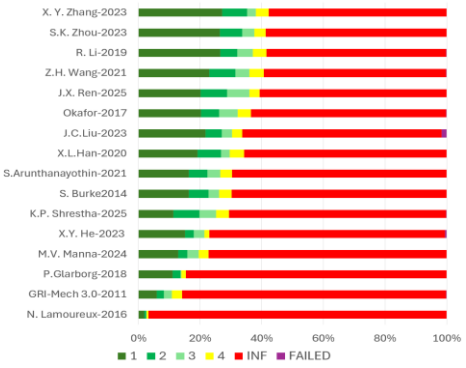

Fig. 1. Distribution of the $1\sigma^{exp}$ - uncertainty normalized absolute simulation errors ($|D_{fsd}|$) of laminar burning velocity experimental data points shown as a stacked bar plot.

Figure 3. shows the performance of the mechanism in predicting rapid compression ignition delay time data. Half of the mechanisms give bad predictions for all the data point, very few mechanisms perform actually very well: the J.X. Ren-2025, the X.Y. He-2023 around 75% of the data within 3 σ^{exp} error. Acceptable performance is shown by the Li-2019, the J.C. Liu-2023 and the X.Y. Zhang-2023 mechanisms.

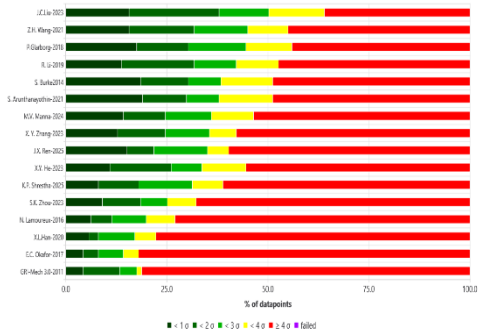

Fig. 2. Distribution of the $1\sigma^{exp}$ - uncertainty normalized absolute simulation errors ($|D_{fsd}|$) of experimental shock tube ignition delay experimental data in a stacked bar plot.

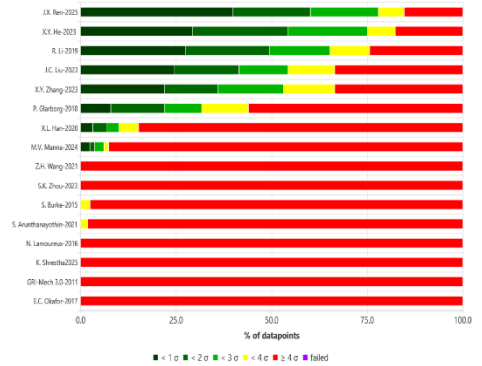

Fig. 3. Distribution of the $1\sigma^{exp}$ - uncertainty normalized absolute simulation errors ($|D_{fsd}|$) of experimental rapid compression machine ignition delay experimental data in a stacked bar plot.

Conclusion

Our study has shown that ammonia-methane combustion mechanisms fail to predict a significant portion of the experimental data with acceptable accuracy. However, it is important to note that the data has not been filtered for consistency and some bad points were not identified and discarded yet.

While the latest methane and ammonia mechanisms have evolved into highly predictive models, the interactions between NH_3 and CH_4 are substantial and not adequately described by current models. Therefore, further developments are needed in this regard.

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