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Cheng, Qiang; Muhammad, Akram; Kaario, Ossi; Ahmad, Zeeshan; Martti, Larmi **Ammonia as a sustainable fuel**

Published in: Renewable and Sustainable Energy Reviews

DOI: 10.1016/j.rser.2024.114995

Published: 01/01/2025

Document Version Publisher's PDF, also known as Version of record

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Please cite the original version:

Cheng, Q., Muhammad, A., Kaario, O., Ahmad, Z., & Martti, L. (2025). Ammonia as a sustainable fuel: Review and novel strategies. *Renewable and Sustainable Energy Reviews*, *207*, Article 114995. https://doi.org/10.1016/j.rser.2024.114995

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Renewable and Sustainable Energy Reviews





Ammonia as a sustainable fuel: Review and novel strategies

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ARTICLE INFO

Keywords: Sustainable transportation Alternative fuels Internal combustion engine Ammonia Fuel-injection

Low-temperature combustion

ABSTRACT

Ammonia is increasingly recognized as a viable alternative fuel that could significantly reduce greenhouse gas emissions without requiring major modifications to existing engine technologies. However, its high auto-ignition temperature, slow flame speed, and narrow flammability range present significant barriers, particularly under high-speed combustion conditions. This review explores the potential of ammonia as a sustainable fuel for internal combustion engines, focusing on its advantages and challenge. The review draws on a wide range of studies, from NH₃ production, application, to the combustion mechanisms, that explore various strategies for enhancing NH₃ combustion in both spark ignition and compression ignition engines. Fundamentals and key approaches discussed include using hydrogen and hydrocarbon fuels as combustion promoters, which have been shown to improve ignition and flame propagation. Literature on fuel injection strategies, such as port fuel iniection, direct injection, and dual-fuel injection, are examined to highlight their influence on NH₃-air mixing and combustion efficiency. Furthermore, the review delves into advanced ignition technologies, such as lowtemperature plasma ignition, turbulent jet ignition, and laser ignition, which are explored for the potential to overcome the ignition difficulties associated with NH₃. After a comprehensive analysis based on the literature, the intelligent liquid-gas twin-fluid co-injection system (iTFI) emerges as a promising approach, offering improved combustion stability and efficiency through better fuel-air mixture preparation. By synthesizing the existing research, this review outlines the progress made in NH₃ combustion and identifies areas where further study is needed to fully realize its potential as a sustainable fuel.

Abbreviations

		RCCI
CH ₄	Natural gas	SI
CI	Compression ignition	SL
DDFI	direct dual-fuel injection	TDC
DI	Direct injection	TJI
DME	Dimethyl ether	
FC	Fuel cell	
GHG	Green house gas	
H ₂	Hydrogen	
HCCI	Homogeneous charge compression ignition	
ICCI	intelligent charge compression ignition	1. Introduction
ICEs	Internal combustion engines	
IDT	Ignition delay time	The everying concern
LBV	Laminar burning velocity	The growing concern
LTC	Low temperature combustion	carbon oxide (CO_2) has
NH ₃	Ammonia	as nitrogen oxides (NO
NOx	nitrogen oxides	drocarbons (uHC) origi
PCCI	premixed charge compression ignition	fied this concern Inte
PFI	Port fuel injection	incu uns concern. inte
	(continued on next column)	contribute to these emi

(continued)

RCCI	Reactivity controlled compression ignition
SI	Spark ignition
SL	Laminar flame velocity
TDC	Top dead center
TJI	Turbulent jet ignition

The growing concern over global climate change, particularly due to carbon oxide (CO_2) has been further intensified by such other pollutants as nitrogen oxides (NOx), particulate matters (PM), and unburned hydrocarbons (uHC) originating from fossil fuel combustion, has intensified this concern. Internal combustion engines (ICEs) significantly contribute to these emissions, accounting for around 20–25 % of CO_2 emissions from fossil fuel combustion worldwide [1]. These emissions

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https://doi.org/10.1016/j.rser.2024.114995

Received 22 December 2023; Received in revised form 8 October 2024; Accepted 8 October 2024 Available online 15 October 2024 1364-0321/© 2024 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY license (http://creativecommons.org/licenses/by/4.0/). stem from various sources such as vehicles (cars, trucks, motorcycles), aircraft, ships, and power generation reliant on fossil fuels, including gasoline, diesel, and natural gas. To achieve the ambitious goal of transforming into the first climate-neutral continent by 2050 in Europe, substantial reductions in CO_2 emissions from ICE-based transportation are imperative. The combustion system, pivotal to ICEs, offers considerable potential for optimization by utilizing carbon-free alternatives, for instance, hydrogen (H₂) and ammonia (NH₃) to immigate CO_2 emissions.

While H_2 emerges as a catalyst for a carbon-free economy, it faces challenges concerning storage, distribution, and its relatively low volumetric energy density (2.9 MJ/L at 70 MPa), hindering its widespread application [2]. Conversely, NH₃ possesses a high energy density (7.1 MJ/L) and established transportation network, presenting remarkable flexibility. These attributes position NH₃ as a practical contender for the next-generation energy transportation, storage, and power generation systems. Additionally, NH₃ offers innovative solutions to sustainability challenges within the energy industry.

By exploring the potential of alternative fuels, including NH_3 and H_2 , this transition holds the key to addressing the carbon emissions in the transportation sector and to fostering a sustainable energy future. Several countries have begun to incorporate NH_3 as a low-carbon fuel into their future energy policies. Notably, the U.S. House of Representatives published a draft legislation that explicitly classified NH_3 as a "low-carbon fuel" in January 2020 [3]. The UK's Royal Society also released a policy with a title of " NH_3 : Zero-Carbon Fertilizer, Fuel, and Energy Store", which provides valuable information and analysis to support the development of regulatory policies regarding NH_3 's utilization within the clean energy technology portfolio [4].

Upon evaluating the various applications of NH_3 for power generation, including ICEs, proton-exchange membrane fuel cells (PEMFCs), alkaline fuel cells (AFCs), and solid oxide fuel cells (SOFCs) for power pulsations, it becomes evident that the ICE boasts high efficiency and practicality. Although SOFCs exhibit superior efficiency compared to ICEs, but suffer from lower power density, load response capability, and cost-effectiveness. In contrast, ICEs offer the second-highest efficiency rating, featuring more efficient than both PEMFCs and AFCs when operated near their maximum power output. Additionally, ICEs present a cost advantage, greater robustness, and acceptable power density and load response characteristics [1].

When comparing NH₃-powered ICEs with conventional fossil-fueled ICEs, the technical performance is notably similar in power density, load response, part-load performance, and overall system efficiency. However, conventional ICEs produce significantly higher levels of harmful emissions, including uHC, NOx, and PM. Importantly, the CO₂ emissions stemming from conventional fossil fuel combustion in ICEs constitute the leading contributors to GHG emissions, thereby exacerbating climate change [2]. Hence, the industry and academia have shown substantial interest in H₂ and NH₃ as carbon-free fuel alternatives. Nevertheless, the liquid NH₃ exhibits a higher volumetric energy density compared to liquid H₂ [5]. Additionally, NH₃ shares storage similarities with propane, existing in a liquid state at room temperature (\sim 25 °C) when pressurized to \sim 10 bar or held at -33.4 °C under atmospheric pressure. Therefore, the primary advantages of NH₃ over H₂ as a fuel can be summarized as following five arguments:

- 1) Cost efficiency: NH_3 emerges as a cost-effective alternative with significantly lower fuel pricing and the added advantage of leveraging existing infrastructure (approximately 10.6–30.2 times less expensive than H_2) [6,7].
- 2) Volumetric energy content: Liquid NH_3 has notably higher volumetric energy content than liquid H_2 (approximately 1.7 times more than liquid H_2).
- 3) Established transportation and storage: NH₃ benefits from wellestablished transportation and storage technologies, evident from over 18 Mt of NH₃ traded internationally annually [7].

- 4) Safety measures: Despite NH₃ being toxic, its potent odor makes detection easy, and well-established safety protocols are widely practiced. In particular, for the NH₃ ships typically require a boil-off gas (BOG) system for vent recovery, similar to LPG (liquefied petroleum gas) tanker ships.
- 5) Renewable production: a wide scale of renewable energy sources such as solar, wind, hydro, etc can be adopted for the NH₃ production. However, it is essential to acknowledge that further research is necessary to unlock its full potential and feasibility.

The adoption of NH_3 as a fuel in ICEs can be categorized into three stages:

Stage I (1905–1942): During a shortage of regular fuels in Europe, NH₃-fueled ICE patents and commercial vehicles were developed. For instance, a Fiat patent in 1931 for NH₃ cars is cited by Cardoso et al. [8], and the NH₃ Cascale company in Italy developed limited-scale automotive applications between 1935 and 1936 [9]. In Belgium, approximately 100 buses ran on NH₃ during the Second World War (1941–1942).

Stage II (1960–1970): Detailed investigations into NH_3 -powered ICEs were conducted in conjunction with the U.S. Army's Energy Depot Concept initiative [10,11], primarily aimed at addressing fuel logistics for military operations, particularly in remote and critical areas [12,13].

Stage III (2003-present): a growing interest in the application of NH_3 as a carbon-free fuel for ICEs, encompassing both theoretical/numerical predictions and experimental tests [14,15]. This research has explored NH_3 combustion in spark-ignition (SI) engines, compression ignition (CI) engines, and gas turbines [16,17]. These endeavors have demonstrated the feasibility of using NH_3 as a alternative fuel in ICEs with minor modifications of the fuel systems and combustion chamber. However, additional research and development are essential to enhance operability and improve engine efficiency, while reducing NOx and $N_2Oemissions$ and NH_3 -slip before widespread applications.

According to the literature review, this paper presents a novel approach to understanding and leveraging NH3 as a sustainable fuel in ICEs, especially in the transportation sector. Unlike previous studies that primarily focus on individual aspects of NH3 combustion, this paper offers a comprehensive review that integrates fundamental combustion studies with real-world engine applications. Additionally, this paper also introduces innovative strategies for overcoming the inherent challenges of NH3 as a fuel, particularly its high auto-ignition temperature and narrow flammability range.

The key contributions of this review paper are three-fold. First, a thorough analysis of current research on NH3 combustion in ICEs, including fuel properties, combustion characterisitcs, chemical kinetics, reaction mechanisms, considering both spark ignition (SI) and compression ignition (CI) engines, which helps identify the most critical challenges and potential solutions. Second, according to the literature review on the fundamental combustion studies and practical engine investigations, this paper explores and evaluates advanced injection and ignition technologies such as direct dual-fuel injection, air-assisted fuel injection, laser ignition, low-temperature plasma ignition and turbulent jet ignition (TJI), offering insights into the potential for enhancing NH₃ combustion efficiency. Last but not the least, the introduction of the intelligent liquid-gas twin-fluid co-injection system (iTFI) represents a significant innovation, which was designed to create homogeneous NH₃-H₂-air mixtures, aims to improve combustion stability and efficiency, thereby reducing the carbon footprint.

The innovation value in this paper lies in the interdisciplinary approach, combining insights from fundamental chemistry, engineering, and environmental science to propose practical, scalable solutions for NH_3 as a sustainable fuel. The iTFI system, in particular, represents a breakthrough in fuel injection technology, offering a novel method to optimize NH_3 combustion and reduce emissions. This innovative concept has the potential to transform the use of NH_3 in ICEs, paving the way for its broader adoption in the energy sector.

2. Methodology

This review paper takes a structured approach to analyzing, evaluating, and presenting the key aspects of ammonia (NH_3) as a sustainable fuel. To analyze existing research on NH_3 as a fuel, a comprehensive literature review is conducted of academic papers, technical reports, and industry publications that discuss the fundamental properties of NH_3 , its combustion processes, and its role as a hydrogen carrier. To compare NH_3 with other fuels, the data on various fuels (e.g., hydrogen, methane, gasoline) is collected and compared them with NH_3 using metrics such as lower heating value (LHV), emission profiles, flame speed, and energy conversion efficiency.

This study centres on technical evaluation of combustion strategies. To examine different combustion strategies for NH_3 , experimental data and simulation results from various studies that explore NH_3 combustion in ICEs are extensively reviewed and synchronized to examine the effectiveness of different ignition methods, injection strategies, and the impact of blending NH_3 with other fuels. The computational tools (e.g., computational fluid dynamics, chemical kinetics models) are also introduced to simulate NH_3 combustion under various conditions. This step may involve validating the model against experimental data to ensure accuracy.

To evaluate innovative approaches to enhancing $\rm NH_3$ combustion efficiency and reducing emissions, a novel strategy with combined intelligent liquid-gas twin-fluid co-ignition, advanced turbulent jet ignition and low temperature mode is expored based on theoretical models or experimental data to assess the feasibility and potential impact of these strategies.

The methodology chosen for this study provides a structured framework for investigating NH_3 as a fuel, ensuring that the analysis is thorough, evidence-based, and relevant for the energy sector. However, potential sources of error in this analysis stem from variability in experimental conditions, assumptions about future technology advancements, and reliance on current market data. These factors expose to inaccuracies in predicting the performance of NH_3 as a fuel and its

environmental impact. If these errors are significant, may skew the results, potentially overstating viability or underestimating the challenges of NH_3 application as a fuel. This "wrongness" could misinform policymakers and scientists, leading to misguided decisions that fail to meet climate change targets. To improve accuracy, future studies should standardize experimental conditions, update economic assumptions, and rigorously validate the proposed strategies in real-world applications.

3. NH₃ production and application

3.1. NH₃ production

The production of NH₃ can be traced back to the 1830s, as illustrated in Fig. 1 [47]. Prior to the industrial production of NH₃ via the Haber-Bosch process, natural fertilizers were primarily sourced from caliche in Chile and Guano in Peru [18]. In 1898, William Crookes played a pivotal role in igniting the growing interest in industrial N₂ activation [19]. The year 1903 witnessed a revival of an ancient method that applied electric arcs to convert atmospheric N₂ into nitric acid (HNO₃), a process originally explored by Henry Cavendish in 1784. This approach was reintroduced in the Birkeland-Evde process, marking a significant milestone in commercial NH₃ production [20]. In 1908, Fritz Haber succeeded in synthesizing NH₃ from N₂ and H₂, overcoming the challenge of a remarkably slow reaction rate under standard atmospheric temperature and pressure conditions [21]. Subsequently, Haber elevated both the reaction temperature and pressure by employing an osmium catalyst to accelerate NH₃ formation and shift the equilibrium. Later, BASF acquired the invention, and in 1913, Carl Bosch scaled up production, giving rise to the well-known Haber-Bosch process. This process operated at temperatures of 400-450 °C and pressures of 150-250 bar, employing a multi-promoted fused iron catalyst. This system is often referred to as Gen 1 (see Fig. 1) and remains the predominant commercially available method for NH3 synthesis. Fritz Haber and Carl Bosch were jointly awarded the Nobel Prizes in Chemistry in



Fig. 1. Historical development of N2 activation.

1918 (awarded in 1919) and 1931 for their groundbreaking contributions to N₂ fixation from the air and the invention of high-pressure chemical methods [22]. Presently, approximately 170 million tons per year, equivalent to 465 thousand metric tons per day (MTD), of NH₃ is produced globally.

Since the 1960s, numerous methods have been developed to study surface chemical reactions, with the aim of enhancing the efficiency of NH₃ production. In 2007, Gerhard Ertl received the Nobel Prize for his significant contributions in elucidating the details of a process of immense importance in artificial fertilizer production: the Haber-Bosch process, which involves the conversion of atmospheric nitrogen (N₂) to NH₃ using an iron catalyst [23]. The signing of the Paris Climate Treaty in 2015 provided substantial impetus for research aiming to decarnomize the N₂ activation. By renewing the H₂ production through steam-CH₄ reforming with renewable energy based water electrolysis, it becomes possible to eliminate the CO₂ emission by Haber-Bosch process. This shift could render NH3 production more sustainable and cost-effective, particularly through the smaller and local reactors, which is often referred to as Gen 2 [24]. Nonetheless, significant challenges must be addressed, including the synthesis of NH₃ at moderate conditions (pressures of 20-40 bar), intermittence of low-pressure H₂ influent from water electrolysis, sustainable separation of pure N₂ from the air, and the discovery of efficient low-temperature thermochemical catalysts at moderate pressures. Electrochemical reduction of N2 and H2O provides an attractive strategy in the pursuit of NH3 synthesis is electrochemical reduction of N2 and H2O, allowing for NH3 production directly at the point of consumption. This approach eliminates transportation costs and emissions and reduces problems related to excess fertilizer runoff, marking it as Gen 3 [25]. The energy requirements for this process, with the energy expenditure of $\Delta G^o = +678.5 k Jmol_{fixed N2}^{-1}$ or Δ $G^o = +19.9 G Jmol_{NH3}^{-1}$ for such a process can be powered by solar or wind in photo/electrochemical systems.

3.2. NH₃ application

NH₃ holds a pivotal role as an industrial chemical with extensive applications, both in its pure form and as a vital precursor for a diverse range of other chemicals [25]. Fig. 2 demonstrates an overview of existing NH₃ application, encompassing its role as a fertilizer, refrigerant gas, and in the production of explosives like ammonium nitrate, trinitrotoluene (TNT), and nitroglycerine. NH3 also finds application in textiles and pharmaceuticals. Remarkably, more than 80 % of globally manufactured NH₃ serves as an agricultural fertilizer.

In the context of decarbonization, the advancement of green NH₃ production has unlocked additional avenues of utility. First, NH3 can Renewable and Sustainable Energy Reviews 207 (2025) 114995

as serving as a carrier for H₂. Second, NH₃ can be used as a transport fuel, either through direct combustion in an engine or via chemical reactions with oxygen in a fuel cell to generate electricity for powering motors. Third, NH₃ facilitates the storage of thermal energy through phase changes from liquid to gas or solid to solid transformations. Several power technologies seamlessly integrate NH₃ (or NH₃-derived H₂) as an energy source, including NH₃-based fuel cells, ICEs, and gas turbines. Table 1 provides a concise summary outlining the advantages, challenges, and requirements associated with utilizing NH3 as a power source.

4. NH₃ as a renewable energy transportation media

4.1. NH₃ transportation and storage

With ongoing advancements in renewable energy technologies and the subsequent reduction in costs, it is logical to develop more efficient methods for transporting renewable energy from high-intensity regions to those with limited renewable energy resources [27,28]. This necessitates renewable energy in a transportable and commercially viable form.

The synthesis of fuels through the process of electrolysis using renewable energy, often referred to as power-to-X or e-fuels, has garnered significant attention. These fuels include liquid H₂, synthetic natural gas, methanol, and NH₃ [1]. Among these options, NH₃ stands out as a particularly advantageous choice as a transportation fuel. The key issue for the H₂ transporation related to pressurizing or liquefying H_2 is energy-intensive, which needs more than 30 % of the total H_2 energy plus the boil-off losses during transportation. Moreover, the infrastructure required for long-distance shipping of H₂ is still under development. In contrast, NH3 can be easily liquefied at around 10 bar pressure at room temperature. Moreover, the storage and shipping infrastructure for NH₃, including transportation regulations, have been established for many years. Furthermore, NH3 features a significant advantage in that it contains 40 % more H₂ (17.75 wt %) than methanol (12.6 wt %). Table 2 lists the advantages and disadvantages of various synthetic fuels in transportation and storage.

4.2. NH_3 as an H_2 carrier

To tackle the challenges associated with H₂ transportation and storage, the chemical carriers, for example, methanol [30], formic acid [30], and NH₃ borane [31] has gained significant attention. Among these carriers, NH₃ stands out as a promising contender due to its low production cost (Bosch-Haber process), impressive energy density (12.8



Fig. 2. Schematic of existing and expanded end uses of NH₃.

Fuel technologies applicable to NH₃ [26].

Application	Efficiency, %	Pre-treatment	Capital Cost (£/kW)	Advantages	Disadvantages	R&D Focus
Proton exchange membrane fuel cell	40–50	NH ₃ decomposition Trace NH ₃ removal	100 (mobile) 1300 (stationary)	Established technologies Suitable for mobile applications	Cost and use of platinum Sensitive to un-cracked NH ₃	Decreasing platinum use Efficient NH_3 decomposition
Alkaline fuel cell	50–60	None	1300 (stationary)	Non-use of platinum (or nobel metals) High tolerance of NH ₃	Low energy density Few commercial suppliers Requires carbon dioxide scrubbing	Increase in energy density Extend suitability Innovation for CO ₂ scrubbing Direct NH ₃ system
Solid oxide fuel cell	50–65	None	760 (stationary)	Established technologies Decompose NH ₃ in-situ Non-use of platinum (or nobel metals)	High-temperature operation Large-scale commercialization Corrosion of components	Extend suitability Combining heat and power Reduction of oxidation impacts
ICE	30–50	Partial decomposition is beneficial	30-45 (mobile) 1000 (stationary)	Established technologies Robust technologies High-power density	NOx gases and NH_3 slip Low efficiency	NH ₃ cracking systems Reduce NOx emiision Reduce unburned NH ₃
Boilers and Furnaces	85–90	None	150-350 (stationary)	Established technology at low NH ₃ content Robust technology High power output	Low efficiency of NH ₃ combustion NH ₃ slip and NOx emission Corrosion	Injection and combustion technologies Use new materials and innovative distribution concepts
Combined cycle gas turbine	55–60	Partial decomposition is beneficial	750 (stationary)	High power output Full-cycle development (heat, power cooling)	imcomplete NH ₃ combustion NH ₃ slip and NOx emission Technology under development	Efficient NH_3 combustion Reduction of NH_3 and NOx Larger power output

GJ/m³), and easy to be liquified at room temperature. Implementing an NH₃-based system for storing and transporting H₂ could be a pivotal step toward achieving a carbon-free society [32,33]. However, the wide-spread application of NH₃ as an H₂ carrier, particularly in transportation, faces limitations related primarily to the efficient decomposition of NH₃ into H₂ [33,34]. Overcoming this challenge relies on developing a rapid initiation and high-rate H₂ generation process that independent external energy input.

The realm of NH₃ decomposition catalysts has been extensively reviewed by Yin et al. [35] and Schüth et al. [36]. Among the available metal catalysts, such as Fe, Ni, Ru, and Ir, supported Ru catalysts exhibit exceptional activity. Additionally, a novel class of NH₃ decomposition catalysts, including substances such as sodium amide [33] and nonstoichiometric lithium imide [37], has shown promising efficiency. However, these highly active catalysts remains a challenge due to the endothermic nature of NH₃ decomposition, requiring continuous heating from an external heat source throughout the reaction.

Converting NH₃ to H₂ at the point of combustion involves an additional step for NH₃ decomposition, which reduces overall energy efficiency compared to burning NH₃ directly. Considering NH₃ has a lower flame speed and a narrow flammability range, which may need combustion promoters such as H₂ and other hydrocarbons to improve the combustion efficiency. In this context, on-board H₂ cracking within ICEs presents more versatility and reliability compared to fuel cells (FCs). The elevated temperatures generated during combustion or within hightemperature exhaust gases can supply sufficient energy for NH₃ cracking, offering a potential solution to the energy input challenge.

4.3. NH₃ on-board cracking

On-board H_2 fuel reforming emerges as a crucial technology for recovering waste energy, presenting a promising avenue to enhance the efficiency of NH₃ ICEs. This thermochemical energy is effectively tapped from the hot exhaust stream and comsumed through the endothermic catalytic reforming process. It involves combining gasoline with a fraction of engine exhaust gas within a catalytic reformer, leading to the production of a H₂-rich reformate. This reformate boasts higher enthalpy compared to the original gasoline supplied to the reformer and is reintroduced into the intake manifold: a concept referred to as reformed exhaust gas recirculation (rEGR) [38].

The on-board production of H_2 via the decomposition of NH_3 presents itself as a viable option, whether solely based on heat (as in Eq. (1)) or incorporating both heat and exhaust species like O_2 (as depicted in Eqs. (1) and (2) respectively). Leveraging stored NH_3 holds advantages over storing H_2 in highly pressurized vessels, especially in the context of passenger vehicles [39]. This strategic approach aligns with the guide-lines set forth by the Department of Energy, US. A commonly employed reformer hardware system is illustrated in Fig. 3, positions the reformer catalyst within the high-pressure EGR loop, strategically placed in close proximity to the exhaust manifold. This placement aims to maximize reformer catalyst temperatures, optimizing the reforming process.

$$NH_3 Decomp.: 2NH_3 \rightarrow 3H_2 + N_2 \Delta H^o = 46.19 \, kJ \,/ \, mol$$
 (1)

NH₃ Decomp. with Exhaust : $NH_3 + xO_2 \rightarrow 2xH_2O + (1.5 - 2x)H_2$ + 0.5N₂ if x < 0.75 (2)

5. Environmental and economic assessments

5.1. Environmental assessment

When NH₃ undergoes combustion in one-step mechanisms, the sole byproducts produced are pure N2 and water, both of them are friendly to the environment. This fundamental concept forms a closed and sustainable cycle, illustrated as the NH₃ economy in Fig. 4 [7]. Nevertheless, transitioning from an economy entrenched in hydrocarbon-based energy to one centered around NH₃ requires meticulous consideration for the environmental consequences linked to the NH₃ production and application. It is crucial to avoid merely replacing one crisis triggered by CO2 emissions with another concerning NH3 and NOx emissions. Fig. 4 delineates the principal atmospheric, terrestrial, and marine cycles involved in the exchange between N2 and various significant forms of "fixed" N2, where "fixed" refers to all inorganic, organic, or biochemical N₂ forms except N₂. An essential insight from this illustration is that anthropogenic N₂ fixation already contributes an annual quantity of fixed N₂ compounds roughly equivalent to that from natural processes. Recognizing that the recycling and re-release systems of the earth must ultimately manage this additional fixed N2 becomes vital, especially

The comparison of different synthetic fuels in transportation and storage [29].

Fuel	Storage Type	Advantages	Disadvantages
H2	Physical storage: Compressed gas, cold/cryo and liquid H ₂	Compressed gas: Mature technology Liquid H ₂ : High storage density (70.8kg/m3) Cryo-Compressed: High volumetric capacity	Compressed gas: Low volumetric density (<40kg/ m3), high compressing energy, etc. Liquid H ₂ : H ₂ loss, safety issue, high liquefaction energy Cryo-Compressed: high compression/ liquefaction energy
	Material based: Adsorben (Graphene),liquid organic H ₂ carries (Cycloalkanes), complex hydrides (LiBH4),etc.	Metal hydrides: Low operating energy Carbon materials: High H ₂ storage capacity Chemical hydrides (AlH3): Good volumetric capacity	Metal hydrides: Low gravimetric/ volumetric capacity, high operating temperature Carbon materials: Low volumetric density, loss of useable H ₂ , low operating temperature Chemical hydrides (AlH ₃): Thermal management required
Synthetic Natural Gas (SNG)	Physical storage: Compressed gas, cold/cryo, and liquid H ₂	Pipelines transportation 70 % less CO ₂ as compared to other fossil fuels No ashes left after combustion Inexpensive Automotive fuel Low pollutants (e. g., SOx, NOx) or soot emission	Low volumetric energy (4 times lower than petrol) The difficulty of leak detection Expensive gas infrastructure Stored and distributed under high-pressure
Methanol	Liquid	Higher volumetric energy density than H ₂ Easy to transport East ro be reformed by RE	Corrosive Need of catalysis Need of up-scaling and further development of methanol reactor unit
Green NH ₃	compressed gas, or liquid	Abundant feedstocks; same properties as liquified natural gas (LPG) Liquid NH ₃ density is 70 % more than liquid H ₂ and three times more than compressed H ₂ Green NH ₃ produced from water electrolysis has the potential to reduce \sim 1.6 % of current global emissions Easily liquified	Currently, the energy required from NH ₃ production comes from fossil fuels Lower reactivity compared to hydrocarbons Require corrosive- resistant components

considering that the collective strain on these natural mechanisms has two folded in the last century since the widespread implementation of Haber-Bosch technology.

5.2. Economic assessment

Recognizing the imperative for NH₃ to maintain competitive pricing

to facilitate its widespread adoption as a fuel, it becomes crucial to contextualize NH₃ costs concerning contemporary liquid fuels. Recently, the international price of NH₃ has varied within the range of USD 250-400 per tonne (cost and freight), mainly influenced by local natural gas or coal costs at production facilities [31]. For the purposes of our discussion within this paper, the approximate cost of NH₃ is USD 300 per tonne, encompassing transportation expenses. Table 3 lists a comparative examination of fuel properties and costs, comparing NH₃ with diesel, gasoline, liquefied petroleum gas (LPG), and marine bunker fuel (heavy fuel oil). The foundation for this comparison lies in an approximate cost per kWh, computed using their LHVs. It is worth noting that fuel prices can significantly diverge worldwide due to various variables, such as local taxation and delivery expenses. This analysis underscores the potential for NH₃ to compete with current fossil fuel prices for end-users, contingent upon the demonstration of at least equivalent energy efficiency in the technologies employed for end-use.

5.3. Key concerns on long-term ecosystem damage

Shipping NH₃ presents several hazards and health risks to marine life due to its toxic and corrosive nature. If NH₃ is accidently released into the marine environment, it can have severe consequences: Firstly, NH₃ is highly toxic to marine organisms, especially fish and invertebrates, which could cause damage to gills, disrupt normal respiratory functions, and lead to death. Even low concentrations can be harmful, affecting the reproductive systems and growth rates of marine species [45]. Secondly, NH₃ can accumulate in the tissues of marine organisms. Over time, this can lead to higher concentrations in the food chain, impacting not only individual species but also the broader ecosystem [46]. Thirdly, NH₃ can alter the pH of water and lead to oxygen depletion, which can disrupt the delicate balance of marine ecosystems, affecting the survival of various species that are sensitive to pH and oxygen concentration changes [47]. Last, NH3 is corrosive and can damage the structural integrity of shipping containers and vessels [48]. If a spill occurs, the corrosive nature of NH₃ can exacerbate the environmental damage, making containment and cleanup more difficult. Therefore, persistent NH₃ contamination can lead to long-term changes in marine ecosystems, including shifts in species composition, loss of biodiversity, and changes in the food web. Mitigating these risks requires stringent safety measures during NH₃ transport, including robust containment systems, monitoring for leaks, and rapid response strategies in case of spills.

6. Physical and chemical properties of NH₃ as a fuel

6.1. Physical and chemical properties

 NH_3 is a colorless gas with a sharp, pungent odor, lighter than air and disperses quickly when released into the atmosphere [1,9]. The liquid NH_3 has a density of 0.728 kg/m³ at 15 °C, providing a higher volumetric energy density compared to liquid H_2 . Remarkably, NH_3 proves highly suitable as a fuel in extremely cold conditions due to the low freezing point (-77.73 °C). Moreover, a high heat of vaporization (23.3 kJ/mol or 1371 kJ/kg at its boiling point of -33.35 °C) make the NH_3 can be managed as a liquid in thermally insulated containers. Table 4 lists the detailed physical and chemical properties of NH3.

The NH₃ molecule takes on a trigonal pyramidal shape, where three hydrogen atoms are bonded to the nitrogen atom along with an unshared pair of electrons. It exhibits polarity and substantial association due to robust intermolecular hydrogen bonding. Notably, NH₃ has a dielectric constant ($22 \text{ at} -34 \degree \text{C}$) lower than that of water ($81 \text{ at} 25 \degree \text{C}$), which means it is a good solvent for organic materials. Additionally, the challenges of non-flamable properties under ambient conditions could mitigate fire or explosion risks [49]. Furthermore, NH₃ showcases vapor pressure characteristics similar to LPG, enabling its storage and transportation using similar practices and precautions [49]. NH₃ demonstrates relatively low viscosity, potentially advantageous for fuel



Fig. 3. Reformer hardware layout (red arrows: hot (outlet), blue arrows: cold (inlet)) [38].



Fig. 4. Vision of the "Planetary nitrogen cycle" [1].

atomization and fine droplet formation within high-pressure fuel injection. The table below details the physical properties of NH₃.

6.2. NH_3 as a fuel compared with other fuels

The energy density of a fuel stands as one of the most critical factors in determining its viability as an energy vector [25]. Fig. 5 illustrates the energy densities of various fuels. While NH_3 demonstrates superior energy density compared to batteries, H_2 , CNG, and methanol. The pure NH_3 exhibits low laminar burning velocities (LBV), alongside high auto-ignition temperatures and ignition energies, which poses challenges when attempting to combust it in its pure form. Therefore, comprehensive insights into the NH_3 combustion in power pulsation systems is necessary. The comparison of the fuel proerties can be seen in Table 5.

6.3. Advantages and disadvantages of NH₃ as a fuel

Even though NH₃ becomes a potential alternative to immigate fossil fuels, considerable debates about its merits and drawbacks in industry and academia are nevere stoped. Based on the preceding discussion, Table 6 summaries the advantages and disadvantages of using NH₃ as a

Relative properties and costs of NH_3 compared with conventional liquid fossil fuels [40,41].

Fuel	P (Bar)	Density @15 °C (kg m ⁻³)	LHV @25 °C (kWh kg ⁻¹)	LHV @25 °C (MWh m ⁻³)	Cost (USD kg ⁻¹)	Cost (USD kWh ⁻¹)
NH ₃	10	603	5.18	3.12	0.30	0.058
Diesel	1	846	12.1	10.2	1.00 (USA)	0.083
LPG	14	388	12.6	4.89	1.00	0.079
					(Germany)	
Gasoline	1	736	12.1	8.87	1.81	0.15
					(Japan)	
Heavy	1	980	10.8	10.6	0.59	0.055
fuel					(Global	
oil					average)	

Note: Adapted and Updated from Zamfirescu and Dincer [40]. LPG, diesel, and gasoline costs from Global Petrol Prices, which include local taxes and delivery costs [42]. Heavy fuel oil prices from Ship & Bunker [43]. NH₃ price from Apodaca and Ewing [44].

Table 4

Physical and chemical properties of NH₃ [9,49].

Parameter @ Conditions	Unit	Value
Molecular weight	g/mol	17.031
Boiling point @1.013 bar	°C	-33.43
Density @1.013 bar, 15 °C	kg/m ³	0.728
Vapor pressure @ 0 °C	Bar	4.29
Vapor pressure @ 20 °C	Bar	8.55
Flammability range in air	% vol.	15.0-30.0
Viscosity@25 °C	µPa⋅s	10.07
Viscosity@-40 °C	mPa⋅s	0.276
standard enthalpy change of vaporization	kJ/mol	23.35
Flashpoint	°C	132
Autoignition temperature	°C	651
Explosive limits	%	15,0-33,6





fuel in ICEs.

7. Fundamental studies on $\rm NH_3$ and $\rm NH_3\text{-} based$ mixtures combustion

Developing efficient and clean NH_3 combustion necessitates a profound comprehension of the intricate chemical processes driving NH_3 combustion phenomena. Consequently, extensive research efforts have been dedicated to unraveling the fundamental characteristics of NH_3 combustion.

7.1. NH₃ reaction mechanism and reduction models

Over the past decades, extensive efforts have been directed towards building chemical kinetics models for NH₃ combustion. These models aim to precisely forecast ignitibility, LBV, flammability limits, NOx formation, and other critical parameters across varied NH3-based mixtures and conditions [50,51]. Miller and Bowman [52] achieved a significant milestone in the development of the NH₃ combustion mechanism, followed by Konnov and De Ruyck [53-55], who established several detailed mechanisms encompassing NH3 oxidation, decomposition, ignition, and flame structure. Later on, a comprehensive H/N/O mechanism involving 129 species and 1231 reactions with several corrections were made in subsequent NH₃ combustion mechanisms [56,57]. However, disparities persisted between different mechanisms and experimental data. Recent comparisons by Otomo et al. [58] highlighted that earlier models by Dagaut et al. [59], Mathieu et al. [60, 61], Klippenstein et al. [62], Tian et al. [63], and Song et al. [64] notably overestimated LBVs. Further detailed NH3 combustion mechanism is made by Shrestha et al. [65], featuring 1089 reactions and 125 species for H₂-CO-CH₄-NO-NH₃. Similarly, detailed models proposed by Miller-Bowman [52], GRI [66], and San Diego [67] for CH₄ combustion, accounting for nitrogen species like NH₃ and NO were studied for NH₃ oxidation [57,68]. While predicted and measured flame speeds aligned well [69], discrepancies surfaced under stoichiometric conditions with minimal NH₃ concentrations and fuel-lean conditions [70].

For combustion of NH₃ blends, additives such as H₂, CH₄, and other hydrocarbon fuels are considered to enhance ignition capability and flame velocity. This prompts the urgency to develop more precise, robust and compact reaction models for NH3-based multiple fuel combustion to reduce computational time. Efforts by Norström et al. [71] investigated six global mechanisms for NH3 oxidation, comparing them against a validated comprehensive mechanism comprising 56 chemical species and 340 reversible reactions from Streiberg et al. [56]. Similarly, studies by Xiao et al. [72] yielded five different-size reduced mechanisms derived from Konnov's mechanism [73] for NH₃/CH₄ combustion under gas turbine conditions. Rocha et al. [74] developed three reduced kinetic mechanisms for NH₃/H₂ mixtures using an optimization-based method [75] applied to the mechanisms of Okafor et al. [76], Mathieu et al. [61], and Otomo et al. [58]. Additionally, Okafor et al. [77] formulated a reduced kinetic mechanism for NH₃/CH₄ mixture combustion, validated for LBV and species concentrations in flames. Despite these optimizations improved predictions of LBV of NH₃ and NH₃-based blends, a post-optimization is still needed for ignition delays.

The major challenge remains in constructing a reduced mechanism capable of globally predicting LBV accurately. Future advancements might necessitate leveraging artificial intelligence-based algorithms to refine reduced mechanisms and bridge the gap between accuracy and computational efficiency.

7.2. Ignition behavior

Ignition delay time (IDT) is a vital factor in the combustion of fuels within ICEs, directly impacting the initiation of the chemical reaction. Compared to other fuels, NH₃ exhibits a notably higher ignition temperature (around 651 °C, in contrast to 230 °C for diesel) and demands a considerably higher minimum ignition energy (approximately 8.0 mJ, compared to 0.018 mJ for H₂). The challenging ignition properties of NH₃ is essential to blend existing fuels as 'promoters' to enhance NH₃ ignition and combustion, paving the way for NH₃ applications in ICEs [78,79]. Understanding the ignition behavior and knock resistance of NH₃ blends with combustion promoters through autoignition measurements at well-defined conditions is paramount. These measurements are vital for evaluating chemical mechanisms that describe NH₃ oxidation and facilitating models to simulate ignition behavior.

Early experimental studies on NH_3 ignition and oxidation in shock tubes date back to the 1960s–80s [80–83]. However, these initial shock

Comparison of the fuel properties for ICE application [25,49].

Properties	Gasoline	Diesel	Natural Gas	Gaseous H ₂	Liquid H ₂	DME	Ammonia
Molecular	C ₈ H ₈	C12H23	CH ₄	H ₂	H ₂	CH ₃ OCH ₃	NH ₃
Lower Heat Value, MJ/kg	44.5	43.4	38.1	120.1	120.1	28.43	18.8
Flammability limits, vol%	1.4–7.6	0.6-5.5	5–15	4–75	4–75	3–18	16-25
Laminar Flame Speed, m/s	0.58	N/A	N/A	3.51	3.51	N/A	0.15
Autoignition Temperature, °C	300	230	450	571	571	350	651
Absolute Minimum Ignition Energy, mJ	0.14	N/A	N/A	0.018	N/A	N/A	8.0
Octane Rating, RON	90–98	N/A	107	>130	>130	60.6	110
Fuel Density, kg/m ³	698.3	838.8	187.2	17.5	71.1	668	602.8
Energy Density, MJ/m ³	31,074	36,403	7132	2101	8539	18,991	11,333
Latent Heat of Vaporization, kJ/kg	305	270	N/A	N/A	445.6	467	1370

Table 6

Advantages and	disadvantages (of NH ₃ as a	fuel in	ICEs	[25,49].

Disadvantages
ligh latent heat of vaporization (1370 kJ/g)
ow flame speed (7 cm/s) potentically eads to incomplete combustion
ligh minimum ignition energy
ligh autoignition temperature
larrow flammability limit
otential high NOx and N_2O emissions

tube studies suffered from poor repeatability and limited experimental accuracy. A significant concern contributing to experimental uncertainty was the oversight of NH_3 adsorption on the stainless-steel equipment's contact surface [84]. Efforts to mitigate NH_3 adsorption involved measures like passivation, as suggested by Mathieu et al. [60], focusing on measuring IDTs of argon-diluted NH_3 in a shock tube. Pushing towards engine-like conditions, Shu et al. [85] extended shock tube IDT measurements of NH_3 to higher pressures, up to 40 bar. Besides to the IDT studies with pure NH_3 in shock tube, the IDT of NH_3/H_2 mixtures have been conducted in rapid compression machines (RCMs) by Pochet et al. [86], He et al. [87], Dai et al. [88], and Shu et al. [89].

Given rising concerns about NOx and NH₃-slip emissions resulting from NH₃ combustion, low-temperature combustion (LTC) strategies are gaining traction. However, most previous studies have not covered the temperature range (1000–1100 K). To enrich NH₃ IDT data, future research should prioritize exploring LTC conditions, encompassing low temperatures (900–1100 K), absence of dilution, lean conditions (equivalence ratios, ϕ , of 0.2–0.5), and elevated pressures (30–60 bar).

7.3. Flame structure and velocity

LBV serves as a fundamental indicator governing fuel combustion in practical energy devices, encapsulating diffusivity, exothermicity, and reactivity [90]. NH₃ features a low LBV (ranging from 5 to 13 cm/s) [91], impeding its application in certain energy devices. To enhance combustion, various experimental and numerical investigations have explored seeding NH₃ with more combustible fuels like CH₄, H₂, CH₃OH, $\rm C_2H_5OH,\,N_2O,\,CO,$ and syngas. These additives have shown potential for significantly boosting flame velocity and extending flammability ranges.

Figs. 6 and 7 present direct photographs of NH₃/air and NH₃/CH₄/ air premixed flames. In pure NH₃/air, the orange luminosity stems from NH₂, NH₃ α band spectra [91] and superheated H₂O vapor spectra. An increase in equivalence ratio intensifies the orange chemiluminescence due to NH₂ emission. The addition of CH₄ alters the flame luminosity due to the appearance of hydrocarbons, emitting blue luminosity.

Key findings from the study of NH3/air flame structures and burning velocities include: 1) Fuel-lean flames exhibit pronouncedly wrinkled structures at the flame front compared to stoichiometric and fuel-rich flames [92]. 2) The highest turbulent burning velocity values manifest in fuel-lean cases owing to a more wrinkled flame front with a lower effective Lewis number compared to fuel-rich cases. 3) The ratio of turbulent to laminar burning velocity (S_T/S_L) demonstrates nearly linear growth with turbulent intensity. Meanwhile, the ratio of wrinkled flame surface area to ensemble-averaged flame surface area sees only a slight increase with varying turbulent intensities [93].

Extensive studies on flame structures and burning velocities with various additives have shown that in NH3/CH4 flames, the ratio of turbulent to unstretched LBV decreases as the NH3/CH4 ratio increases. This is due to a reduction in flame surface density as the NH3 ratio increases, leading to a larger fractal inner cutoff [94–96]. The introduction of H2 has a nonlinear effect, increasing both unstretched LBV and Markstein length, with the latter showing a non-monotonic rise as the H2 ratio increases [97]. Studies on syngas, CO, and H2 further reveal the distinct chemical effects of H2 and CO additions on NH3 oxidation [98].

7.4. Flammability and stability

Even though the pure NH_3 combustion exhibits slow reaction rate, the presence of H_2 significantly increases the production of vital radicals such as OH, H, and O, which remarkably enhances the flammability and stability. This enhancement promotes conversion sequences, including NH to NO and, subsequently, NO to N_2 , especially near the lower flammability limits. Similarly, the conversion of NH_2 and NO to N_2 near the upper flammability limits is facilitated, thereby widening the flammability limits [95].

Studies exploring the lower flammability limits of $H_2/NH_3/air$ mixtures at various initial temperatures revealed a strong adherence to the



Fig. 6. Natural flame luminosity of NH₃/air premixed flames of $P_i = 0.1$ MPa at t = 100 ms. (a) $\phi = 0.8$, (b) $\phi = 1.0$, (c) $\phi = 1.2$ [91].



Fig. 7. Natural flame luminosity of premixed CH₄/air flame with increasing NH3 concentration [76].

Le Chatelier flammability limit mixing rule. Interestingly, in mixtures dominated by dissociated NH₃, autoignition dynamics often hinder the measurement of the fuel-rich flammability limit [96]. To improve NH₃ flame stability across diverse devices, several innovative mixing strategies have emerged. For instance, Khateeb et al. [27] presented findings on stability limits for technically-premixed swirl NH₃-air flames enriched with either CH₄ or H₂. Notably, the critical NH₃ fraction increases with pressure, being more substantial for NH₃-H₂ compared to NH₃-CH₄ blends.

These investigations demonstrate that when an adequate amount of H₂ is blended with NH₃ (χ_{NH3} < 0.9), flames with exceptionally lean equivalence ratios (φ < 0.7) can be stabilized, resulting in impressively low NO emissions (<200 ppm), irrespective of pressure. However, under such operating conditions, there is a risk of unacceptably high N₂O emissions if heat loss is extensive or residence time is insufficient. These findings highlight the delicate balance required to achieve both flame stability and emissions control in NH₃-enriched combustion scenarios.

7.5. NH₃ oxidization and emissions

Reducing NOx emissions such as NO₂, NO, and N₂O play a crucial role in ICEs development due to their significant impact on atmospheric chemistry. In particular, N₂O is an important greenhouse gas, contributes to positive radiative forcing and stratospheric ozone depletion, while NO influences tropospheric ozone production and atmospheric photochemistry.

 N_2O is notably formed through reactions involving the amine radical $NH + NO \rightleftharpoons N_2O + H$ and $NH_2 + N_2O \rightleftharpoons N_2O + H_2O$, especially under lean or high-pressure conditions. Additionally, atomic oxygen involvement in $H_2 + O \rightleftharpoons H + OH$ increases the radical pool. Reactions consuming N_2O in the thermal de-NOx process and reactions such as $N_2O + H \rightleftharpoons N_2 + OH$ and $N_2O + H \rightleftharpoons NH + NO$ are crucial for its consumption and production [99]. Regarding NO formation, HNO pathway dominantly generates NO in the rich-burn stage flame zone, while NH_i decomposition in the post-flame zone plays a significant role in NO reduction [27]. The concentration of N_2O peaks under lean conditions due to NO + HO2 $\rightleftharpoons N_2O + H$, while it remains minimal under stoichiometric conditions due to high temperatures [99].

The influence of additives in NH₃ combustion has been extensively studied with combiniation of C/N/H/O mechanisms [61,74]. Differences in NOx formation pathways between pure NH₃ and NH₃/CH₄ blends at $\phi = 0.7$ have been outlined. The NH₃ mole fraction (η_{NH3}) in CH₄/NH₃/air significantly affects combustion efficiency and NOx emissions, especially under fuel-lean conditions ($\phi < 1.0$), with maximum NOx emissions occurring at $\eta_{NH3} = 0.5$ [100]. Optimal conditions ($\phi \sim 1.1$) for minimized NOx and NH₃ emissions simultaneously have been identified [101]. Understanding the effects of OH concentration and temperature on CH₄/NH₃/air oxidation processes is enssential for emission control [102]. To address NH₃-slip, introducing H₂ as a promoter helps balance the trade-off between NO emissions and poor combustion efficiency [88]. For the diesel ignited NH₃ dual-fuel combustion, unburnt NH₃ releases from fuel-rich regions while NO produces from fuel-lean areas, with heat losses impacting NO and CO

emissions from NH₃ combustion.

Interestingly, NH_3 addition reduces soot formation during cocombustion with hydrocarbon fuels. Chemical kinetic analysis highlights C single bond N species from ethylene- NH_3 flames inhibit carbon participation in soot precursor formation, consequently reducing soot levels in flames [103]. This effect arises from NH_3 inhibiting soot inception and growth, rather than enhancing oxidation.

8. NH₃ application in ICEs

The historical journey of NH_3 as a vehicular fuel traces back to Sir Goldsworthy Gurney's proposal in 1822. The first small-scale motor was developed by NH_3 Casale Ltd. in 1905, with patents in Italy in 1935 and 1936 [49]. Norsk Hydro operated an ICE vehicle with H_2 derived from onboard NH_3 cracking as early as 1933 [5]. During World War II, fuels shartages in Belgium led to the exploration of compressed coal gas and later NH_3 a diesel replacement in the Belgian bus service. NH_3 powered ICEs gained significant attention during the 1960s through the U.S. Army, aimed at ensuring fuel supply security for remote critical operations. However, this initial project prioritized fuel logistics over cleaner energy and combustion alternatives.

In recent years, there has been a resurgence of interest in NH₃ as a clean alternative for ICEs, driven by theoretical/numerical predictions and experimental tests in SI and CI engines. These studies have demonstrated the potential of using NH₃ as a sole fuel in ICEs with requisite modifications to fuel systems and engines. Nevertheless, further research and development are imperative to improve operability, engine efficiency, while reduce NOx emissions for boarder application. The viability of pure NH₃ powered SI and CI engines presents significant challenges due to its poor ignition and combustion characteristics. As a result, blending NH₃ with highly reactivity fuels in a dual-fuel combustion mode has gained more attention. Fig. 8 outlines potential technical paths for NH₃-based ICEs.



Fig. 8. Different technical paths to NH₃-based ICEs.

8.1. NH₃ powered SI engines

There are several practical approaches to imporve engine performance when using NH_3 as a fuel in SI engines, for example, increasing spark energy, increasing compression ratios, supercharging/turbocharging, and blending more reactive fuels.

NH₃ was effectively used as the sole fuel in a single-cylinder SI engine, demonstrating operation within a compression ratio of 6-10 and an equivalence ratio of 0.8-1.4 by Starkman et al. [155]. However, subsequent findings by Sawyer et al. [104] in 1968 highlighted that NH₃ combustion led to higher NOx concentrations compared to hydrocarbon fuels. This disparity suggested a distinct NOx formation mechanism with NH₃ fuel, hinting at possible equilibrating processes between combustion and exhaust. Cooper et al. [16] later implemented supercharging techniques, plasma jet igniters and multiple spark plugs strategies to facilitate more efficient NH₃ combustion [105]. A comparative analysis of NH₃ and CH₄ combustion in an optical SI engine [106] revealed that NH₃ exhibited notably lower combustion stability and power capability than CH₄. The findings revealed that the initial flame development process accounted for the majority of the burning time loss in NH₃ combustion. Additionally, NH₃ flame propagation showed higher sensitivity to flame stretch and primarily relied on its response to turbulence for propagation. One proposed method to enhance NH₃ combustion involves inducing turbulence within the combustion chamber. However, the degree of swirl is critical, as insufficient swirl has negligible effects while excessive swirl might extinguish the flame due to the slower propagation of NH₃ flames [104].

To address the challenges of NH_3 combustion in SI engines, fuel additives have emerged as promoters to enhance ignition behavior and flammability. Various additives have been tested alongside NH_3 in dual-fuel modes in SI engines, including gasoline, CH_4 , H_2 .

Blending NH3 with gasoline enabled knock-free operation with higher compression ratios and can achieve similar loads with gasolineonly [107]. However, the overall efficiency, peak pressures, and exergy of NH₃/gasoline blends were slightly lower due to the lower energy density, heating values, and flame speed of NH₃. Additionally, NH3-Gasoline blends exhibited high NOx emissions due to the involvement of more nitrogen atoms in NOx formation and NH₃ slip from incomplete combustion. To mitigate this, post-catalyzers such as three-way catalytic converters and SCR systems were recommended to reduce NOx, unburned NH₃, uHC, and CO emissions. Sechul et al. [106, 108] observed a reduction of approximately 28 % in CO₂ emissions with replacing 50 % of CH₄ with NH₃. However, this approach faced limitations due to lambda value restrictions affecting combustion efficiency and emissions characteristics. The inadequate time for complete combustion led to instability and unburned fuel, while increased NH3 fractions delayed combustion, elevating fuel-bound NOx emissions.

 H_2 emerged as a noteworthy promoter for NH₃-based SI engines due to its superior combustion properties and carbon-free nature. Studies from Mørch et al. [17] and Li et al. [109] highlighted that optimal energy ratios for H_2 /NH₃ around 7.5%–10 % offered improved efficiency and power. To compensate for intake air dilution, raising compression ratios was suggested due to the higher octane rating of NH₃/H₂ mixtures [106]. To reduce NOx emissions, varying H₂ fractions based on load and speed were proposed [110]. It has been observed that H₂ exhibited an acceleration effect during ignition and initial combustion stages, higher H₂ fractions compromised efficiency due to increased wall heat losses [109]. Studies by Westlye et al. [111] emphasized Nitrogen-based emissions from H₂-enriched NH₃-fueled SI engines, showcasing comparable NOx emissions to gasoline due to lower flame temperatures. These findings underscored the potential of NH₃ as a combustion inhibitor and provide valuable insights for H₂-fueled engine development.

8.2. NH₃ powered CI engine

Using NH₃ as fuel in CI engines presents significant challenges due to

its notably high autoignition temperature ($630 \sim 651$ °C) [2] and substantial latent heat of vaporization (1370 kJ/kg) [112]. In comparison, diesel exhibits autoignition temperatures of 226–233 °C and a latent heat of vaporization of 270 kJ/kg [113]. Nevertheless, several potential methods could address the challenges, such as employing an exceptionally high compression ratio (>35), applying a combination of high compression ratios and elevated intake air temperatures [114], and implementing a pilot or dual-fuel system using H₂ [115,116] or hydrocarbon fuels like biodiesel [117,118], DME [119,120].

Wagner T.O. of American Oil Co. emphasized the need to extend the investigations on NH₃ combustion to CI engines in 1965 [12]. The finding suggested that the engine could initiate with kerosene, after a warm-up period then replaced with NH3 with a compression ratio of 35:1. Gray et al. [105] assessed performance and emissions with the same components and lubricants as original engine, and no substantial issues were found with NH3 combustion in CI engines. Interestingly, high-temperature glow coils proved to be more effective ignition sources than standard glow plugs or spark plugs [78]. However, the extensive challenges associated with burning NH₃ in CI engines were highlighted by Garabedian and Johnson [121], who proposed converting CI engines to SI mode as a short-term solution at US Army Tank-Automotive Center in 1966. Starkman et al. [122] confirmed that successful combustion of liquid NH3 in a diesel engine at nominal compression ratios required adopting a spark plug for fuel ignition. Meanwhile, Pearsall [123] encountered issues while operating an AVDS-1790 Vee Twin CI engine with a compression ratio of 30:1 with liquid NH₃ direct injection to overcome the high auto-ignition temperature and limited flammability range (15.15-27.35 % in dry air). However, the available literature on CI engines solely using NH₃ is limited and relatively dated, mainly because of implementing NH3 in a single-fuel concept poses significant challenges, primarily due to the excessively high compression ratios required, which would be impractical in real-world applications.

To address the challenges posed by high ignition temperature and low reactivity of NH_3 , blending NH_3 with highly reactive fuels such as diesel provides a promising solution, as demonstrated through both experimental and numerical studies [124–126].

The concept of using diesel or biodiesel as a pilot fuel for diesel-NH₃ dual-fuel combustion has been extensive explored since 1966. Gray et al. [105] concluded that employing NH₃ as a vapor fuel alongside diesel in a CI engine raised no significant issues. Bro and Pedersen [127] found that achieving high specific fuel consumption required a substantial diesel fraction (approximately 50 %) in the NH₃-diesel blend, resulting in notable NO emissions and unburnt NH₃ fractions. More recently, Reiter and Kong [128] operated a turbocharged diesel engine in a dual-fuel mode using NH₃ up to 95 % of the energy share ratio, noting that NOx emissions decreased minimally with low NH₃ fumigation but increased significantly with high NH₃ fumigation levels.

Niki et al. [129,130] investigated the effect of NH_3 fumigation on a CI engine at various loads, discovering that an increase of NH_3 energy fraction led to higher NOx and unburnt NH_3 emissions. Interestingly, the nitrous oxide (N_2O) and NH_3 slip were reduced with increasing engine load. In a subsequent study in 2019, Niki et al. [131] proposed diesel pre-injection as a practical method to reduce NH_3 slip in NH_3 -diesel dual-fuel engines, maintaining comparable thermal efficiency to diesel-only combustion, while with higher N_2O and NOx emissions. Bicer and Dincer [132] compared the life cycle GHG emissions of green NH_3 -powered vehicles to diesel-powered ones reported significantly lower emissions for NH_3 -based vehicles. Furthermore, engine tests with biodiesel and NH_3 demonstrated similar performance characteristics to diesel- NH_3 blends [117,118].

Dimethyl Ether (DME), an alternative to diesel, has been explored as a pilot fuel for NH₃-based dual-fuel combustion, allowing for NH₃ content up to 80 % [119,120]. However, understanding the operational variations when testing engines with NH₃ and (bio)diesel/DME fuel blends is still a subject of ongoing research and requires substantial development before practical implementation in vehicles. The literature review highlighted a renewed focus on the implementation of NH_3 in CI engines as a strategy to mitigate GHG emissions. However, significant research gaps remain in the understanding the NOx and N_2O formation mechanisms in NH_3 /diesel dual-fuel engines.

8.3. NH₃ in low-temperature combustion modes

In the pursuit of reducing NOx formation in CI engines, alternative combustion strategies, including low temperature combustion (LTC) or lean operation have emerged. However, the exploration of NH₃ as a fuel in LTC modes is rarely reported.

An effort to develop a homogeneous charge compression ignition (HCCI) engine with a 40:1 compression ratio at Sandia National Laboratory has been proposed, but results remain unavailable [209]. Zhang et al. [133] pioneered a physical-based model featuring a unique trajectory synthesizing mechanism, which determined optimal compression ratios (CR = 25) and piston trajectories (symmetric trajectory with $\Omega = 0.5$) for NH₃ HCCI combustion. Pochet et al. [86,134,135] experimentally explored NH₃/H₂ blends in HCCI combustion at compression ratios of 16:1 and 22:1, revealing that NH3 fractions up to 70%vol could significantly reduce NOx emissions with exhaust gas recirculation. Djermouni et al. [136] identified NH₃ as the best performer in HCCI engine performance, followed by H_2 and methanol. Shafiq et al. [137] demonstrated the feasibility of reducing NOx emissions in HCCI mode by adding H₂ peroxide to enhance NH₃ combustion. Rudolph et al. [138] delved into converting typical NH₃ and amine-containing off-gases from chemical plants into fuel-rich operated HCCI engines, focusing on carbon recovery and simultaneous work and heat output. Wang et al. [139] adopted argon as a diluent to improve the ignition and combustion of NH₃ in an HCCI engine, which significantly enhancing ignition behavior and thermal conversion efficiency. Lastly, Bakir et al. [140] and Tian et al. [63] emphasized the role of H₂ in promoting NH₃ combustion for future engine applications under HCCI mode.

Within more advanced combustion concepts, reactive controlled compression ignition (RCCI) has attracted more attention due to its expanded operating range. Ramachandran et al. [118] implemented RCCI operation with NH₃ and biodiesel using a split injection strategy, resulting in higher brake thermal efficiency and lower brake specific energy consumption, although NOx emissions increased compared to single diesel pilot injection. Dupuy et al. [141] systematically investigated efficiency and emissions in a single-cylinder CI engine under RCCI mode, revealing higher CO₂eq levels under lean and rich conditions compared to the NH₃/air mixture around stoichiometry. Zhang et al. [142] examined n-heptane/NH₃ combustion under RCCI conditions in an optical engine, demonstrating that NH₃ combustion is significantly sensitive to mixture reactivity and turbulence. Further studies by Xu

et al. [143,144], Elumalaiet al. [145], and Niki [146] focused on optimizing NH_3 energy share ratios and diesel injection timing in RCCI combustion, highlighting the challenges of high NH_3 -slip and emissions of NOx and N_2O in NH_3 -diesel RCCI combustion.

Li et al. [147] compared NH₃ fuel injection systems under premixed charge compression ignition (PCCI) and gas phase port injection combustion modes, revealing that phase port injection combustion exhibited lower overall NO emissions and higher efficiency than PCCI. Fig. 9 illustrates the concept from Lee and Song [148], which simulated a PCCI-like strategy for NH₃ combustion without secondary fuel. The results shown lower NOx emissions but suffering from reduced combustion efficiencies, unburned fuel, and limited operating ranges. Wu et al. [149] estimated the impact of NH₃ energy fraction, the timing of diesel injection, and diesel injection pressure on PCCI mode, achieving a gross indicated thermal efficiency of 49.7 % and significantly reduced GHG emissions compared to pure diesel mode.

In summary, these varied approaches demonstrate potential benefits such as reduced NOx emissions and enhanced efficiency, while often facing challenges related to unburned fuel, poor combustion phasing control, and limited operational ranges.

9. Possibilities of novel technologies for clean and effective $\ensuremath{\text{NH}}_3$ combustion

9.1. Possible injection strategies on NH3-based combustion engines

 NH_3 integration into SI engines primarily occurs through two injection strategies, including port fuel injection (PFI) and direct injection (DI). Port NH_3 injection may displace the supplied air, impacting the volumetric efficiency of the engine. Conversely, direct NH_3 injection offers better volumetric efficiency, which could also significantly reduces in-cylinder temperature due to its high latent heat. Notably, NH_3 is more suitable for lower engine speeds due to more time for the complete combustion, making it a more prevalent choice for marine engines [6].

A critical factor influencing engine performance is quality of fuel atomization, which subsequently affects vaporization and spray distribution. Therefore, improving NH₃ atomization and vaporization is pivotal for efficient combustion. Fig. 10 illustrates a general categorization of NH₃-based ICE technologies based on the fuel injection and ignition strategies. Four main technologies with 12 strategies are summarized as 1) pure NH₃ combustion with SI and CI, 2) dual-fuel combustion with NH₃ pre-mixed combustion, 3) single- or dual-fuel combustion with advanced pre-chamber ignition, 4) dual-fuel with diffusion combustion (dual-fuel direct injection). The complexity and cost of these applications increase exponentially from 1) to 4).



Fig. 9. Schematic of the NH3 PCCI combustion strategy proposed by Lee [148].

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Fig. 10. Feasibilities of the NH₃ as a fuel in ICEs.

For SI engine applications, several studies have focused on pre-mixed combustion, employing injectors positioned on the manifold to mix the NH_3 with air outside of the cylinder. PFI injections typically maintain pressures lower than 10 bar to mitigate complexity and costs [150]. While existing knowledge from gasoline engines indicated that the PFI engines are less efficient than the DI engines. Considering the properties of NH_3 , DI strategies can deliver higher power due to improved combustion efficiency and the ability to run at higher compression ratios [151,152]. NH_3 DISI engines often adopt moderate high-pressure strategies ranging from 50 to 200 bar [153]. This higher injection pressure enhances fuel atomization and vaporization rates, enabling more precise control over the fuel volume and combustion process across various operating conditions.

In CI applications, extensive investigation into DF combustion strategies involve a premixed NH₃/air mixture ignited with pilot diesel. Implementing high-pressure direct injection of NH₃ in CI engines has emerged as a viable solution for achieving cleaner and more efficient combustion in heavy-duty or marine engines [154]. Over the past few decades, diesel engine injection pressures have escalated from 50 MPa to over 250 MPa, with continued forecasted growth. Experimental studies on high-pressure NH₃ injection are currently limited, but higher injection pressures notably impact spray performance, influencing subsequent combustion and emission processes in CI engines. Forecasts suggest super-high (250–300 Mpa) [155,156] or even ultra-high (>300 Mpa) [157] injection pressures may be realized soon. Existing studies indicate the significance of injection pressure in determining spray characteristics, particularly in enhancing spray penetration and atomization performance.

To further enhance engine performance, investigations into direct dual-fuel injection (DDFI) or intelligent charge compression ignition (ICCI) strategies have explored injecting both low and high reactivity fuels into the combustion chamber using two independent high-pressure injectors [158,159]. These concepts, optimized with high-pressure direct injection (HPDI) for diesel-CH₄ and diesel-H₂ by Westport [160] and L'orange [161], which offer more flexibility in fuel injection and

mixture formation. However, accurately controlling the reactivity gradient remains a challenge in DDFI, particularly in controlling low reactivity fuel injection post low-temperature heat release (LTHR) near the top dead center (TDC).

9.2. Possible ignition strategies on NH₃ combustion

For optimizing combustion in SI engines using NH_3 and charge dilution, effective spark ignition becomes pivotal, especially when incorporating strong cylinder flow. Enhancing the ignition source is crucial to extend tolerance for poor mixture ignitability and allowing for higher levels of mixture dilution. The benefits of a robust ignition source include reduced IDT, late spark timing (close to TDC), and accommodate various in charge motion and mixture homogeneity. Fig. 11 demonstrates the most feasible ignition strategies for NH_3 applications in ICEs.

Conventional single-spot ignition spark ignition systems suffer from a limited ignition source, which requires higher discharge power [6, 162] or prolonged spark glow durations [163]. Zheng et al. [164] developed a strategy that introduces multiple ignition kernels using a multi-pole spark igniter with three electrically independent spark gaps, exhibiting superior combustion phasing and lower variability than single spark discharges. Advanced ignition methods like laser ignition [165], corona ignition [166], microwave ignition [167], non-equilibrium plasma discharge [168], and pre-chamber ignition [169] generate volumetric ignition sources, offering robustness in controlling ignition of diluted mixtures through distributed and multiple ignition sites. Notably, using NH₃ as fuel requires higher compression ratios, faster compression rates, and much higher ignition energy compared to other fuels. Future engines operating under extreme pressure and temperature demand high-voltage (>40 kV) for reliable ignition, making laser ignition, nanosecond plasma ignition, and prechamber ignition active research areas for NH₃ ignition.

Regarding laser ignition [170,171], photolysis has been employed to measure minimum ignition-energy densities for stoichiometric homogeneous mixtures of $H_2/O2/NH_3$, $H_2/air/NH_3$, and $CH_4/O2/NH_3$. These



Fig. 11. Possibilities of the ignition strategies for NH₃ application in ICEs.

measurements exhibit substantially lower than kinetic modeling predictions, indicating a potential role of hot H atoms from NH_3 photolysis in facilitating ignition.

Nanosecond plasma ignition has been extensive investigated into NH_3/air mixtures, demonstrating ignition enhancement through nonequilibrium excitation in nanosecond pulsed plasma discharges [172, 173]. The optimum ignition enhancement occurs at specific electric fields where the production of electronically excited N_2 and the dissociation of NH_3 and O2 are most effective, especially at lean conditions.

Prechamber ignition, also known as turbulent jet ignition (TJI), can effectively enhance the combustion of NH_3 in the main chamber. Zhou et al. [174] provided a comprehensive review on prechamber ignition, highlighting its advantages, such as offering multipoint ignitions and fast jet flame velocities, which extend lean-combustion limits or EGR tolerance under ultralean conditions. Active H_2 -fueled prechambers (reactivity-controlled turbulent jet ignition, RCTJI) have been developed to control chemical reactivity in NH_3 engines, overcoming challenges related to ignition and low flame velocity [175,176].

However, several potential knowledge gaps remain for future research in various NH_3 ignition strategies, including 1) Detailed exploration of fluid-mechanical and chemical-kinetic processes governing ignition sources and fuel-air mixtures. 2) Fundamental studies on spatial ignition, self-generated turbulence, and identifying flame regimes under prechamber combustion. 3) In-depth analysis of balancing the energy released by combustion reactions with the heat transfer to electrodes and the surrounding cooler gas is crucial for refining ignition system designs.

9.3. Low-temperature combustion strategies

LTC is a cutting-edge engine technology offers low NOx emissions for NH_3 combustion while maintaining high fuel efficiency. Among the LTC strategies, such as HCCI, PCCI, RCCI, ICCI, and PPC, early fuel injections play a crucial role by allowing adequate time for air-fuel mixing prior to combustion. However, certain LTC approaches might not be viable for future automotive and power generation applications due to challenges in regulating the heat release rate and the absence of robust combustion phasing control mechanisms.

In NH_3 combustion within LTC modes, precisely control the ignition timing and combustion phasing becomes essential. The optimization of thermal efficiency and expanded engine loads relies on managing equivalence ratio, temperature, and component stratification within

specific temporal and spatial scales. This control ensures the regulation of burning rates and combustion phasing across various operating conditions, allowing stable performance from low to high loads, while maintaining high thermal efficiency and low emissions.

9.4. Intelligent twin-fuel Co-injection strategy

Addressing the challenges related to NH_3 ignition and its slow flame speed, one promising approach involves leveraging H_2 as a combustion enhancer. Achieving optimal mixture homogeneity between H_2 and NH_3 is vital for ensuring ignition stability, promoting combustion, reducing NOx formation, and minimizing NH_3 slip. To facilitate efficient and clean NH_3 combustion in ICEs, a intelligent liquid-gas twin-fuel injection (iTFI) system has been proposed at Aalto University. The advantages of this novel strategy include.

- 1) Flexibility in the control of the liquid-to-gas ratio (NH_3/H_2) based on the injection pressure and injection duration.
- 2) The premixed NH₃ and H₂ injection can further enhance the atomization process of liquid NH₃ and improve the fuel-air mixture quality.
- 3) A more homogenous NH_3 - H_2 reduces the instability of the flame during the combustion.

Fig. 12 depicts the 1D GT-power and 3D CFD simulation of this innovative system, which involves blending high-pressure liquid NH_3 (>300 bar) with gaseous H_2 (>100 bar) before injection. By injecting the liquid NH_3 into H_2 prior to combustion, an accurate control over the liquid/gas ratio and homogeneous high- and low-reactivity fuel mixtures allows for controlled dual-fuel mixtures and varying degrees of stratification within the cylinder.

Additionally, the iTFI system, when combined with a prechamber, offers a volumetric ignition source for NH_3 and H_2 blends, further enhancing the ignition and combustion stability. The comprehensive concept of the application of iTFI system is demonstrated in Fig. 13. There is a clear path forward for realizing the concept. Firstly, high-pressure liquid fuels (>300 bar) injected into the gaseous-filled (>100 bar) mixing chamber. Secondly, the liquid-gas mixture injected into the pre-chamber (additional gaseous fuels can be used for ignition promoter). Lastly, the liquid-gas mixture enters the main chamber and mixed with air.

Further studies on liquid-gas twin-fluids injection are proposed to



Fig. 12. A intelligent liquid-gas twin-fuel co-injection strategy for NH₃ application.



Fig. 13. The concept of the liquid and gas twin-fuel injection for the NH₃-H₂ dual-fuel combustion in ICEs.

provide high-fidelity engine-based data for a deeper understanding of the liquid-gas co-injection and fuel-air mixing processes in pre-chamber and main-chamber.

10. Conclusion and future research

 $\rm NH_3$ is expected to become an important decarbonized fuel for power propulsion and generation. Whether used directly, partially decomposed, or blended with other fuels, several significant advantages

emerge. However, applying NH_3 in combustion presents notable challenges, including low flame speeds, limited flammability range, and a propensity to form NOx emissions. This review paper comprehensively analyzed the potential of NH_3 as a sustainable transportation fuel and proposed several novel strategies for efficient and clean combustion. The following conclusions summarized the current status of NH_3 as a fuel from a combustion perspective:

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- 1. NH_3 can provide an efficient and dependable long term energy solution for the power pulsation systems. However, owing to the low flame propagation speed, long ignition delay time, limited flammability boundaries, and modest flame radiation and temperature, further optimization of the multiple fuel blends, fuel injection and ignition strategies, and combustion modes is necessary for efficient and clean combustion.
- 2. Dual-fuel combustion stands as a prevalent strategy for enhancing the combustion efficiency of NH_3 . Literature reviews highlight diverse combustion promoters blended with NH_3 , including H_2 (in its pure form or partially cracked NH_3), methane, gasoline, diesel, biodiesel, DME, and syngas. Among these fuels, H_2 is the most effective combustion promoter for NH_3 , while CH_4 proves to be the least effective, based on the available literature.
- 3. The combustion chemistry of NH_3 significantly deviates from that of hydrocarbon fuels. Existing NH_3 -relevant chemical kinetics mechanisms exhibit discrepancies in predicting LBV and ignition delay time, largely attributable to inaccuracies in the rate parameters of critical nitrogen family reactions. Therefore, improving the predictive capabilities of chemical kinetics models through expanded target flame conditions and in-depth exploration of C-N crossing reactions becomes paramount.
- 4. Recent investigations into NH₃-H₂ dual-fuel combustion highlight that introducing H₂ enhances the O/H radical pool without altering oxidation pathway, resulting in elevated burning velocity and increased NOx generation compared to NH₃-air flames. Reactions involving nitrogen-containing radicals, OH, and NO, alongside H₂NO oxidation, become more prominent with H₂ substitution. In contrast, CH₄ addition resembles NH₃ oxidation by enriching the O/H radical pool and facilitating crucial C-N interaction reactions. Other alternative dual-fuel combinations like diesel-NH₃, gasoline-NH₃, and DME-NH₃ remain relatively unexplored, delving into these could significantly broaden the scope for the clean and efficient utilization of NH₃ in ICEs.
- 5. Low-temperature combustion, including techniques such as HCCI, RCCI, and PCCI, has gained substantial attention due to its potential to reduce NOx emissions while enhancing efficiency across a broad range of operating conditions. Additional NOx reduction strategies continue to be investigated, including very lean burning, flameless combustion, and humidification.
- 6. To broaden applicability of NH₃ in ICEs, extensive discussions have centered on injection strategies, ignition methods, and combustion modes. Injection strategies encompass port fuel injection (PFI), direct injection (DI), air-assisted direct injection (AADI), dual-fuel direct injection (DFDI), and high-pressure direct injection (HPDI). Notably, dual-fuel direct injection shows promise for optimizing engine performance and emissions. Ignition strategies encompass spark ignition (both conventional and multi-pole), laser ignition, plasma ignition (nanosecond plasma, corona, micro-wave), and prechamber ignition. Low-temperature plasma ignition and prechamber ignition appear as favorable solutions.
- 7. To achieve efficient and clean combustion, a novel strategy introduces the concept of liquid-gas twin-fuel (e.g., H₂, NH₃) highpressure co-injection, coupled with an active pre-chamber to furnish a dependable volumetric ignition source. This concept embodies intelligent mixing-controlled combustion (iMCC) aimed at enhancing efficiency and reducing emissions.

In summary, the application of NH_3 as a fuel in combustion-based technologies holds significant promise, but it requires addressing its unique challenges through innovative strategies and a deeper understanding of its combustion characteristics. However, the limitations identified in this review underline the need for further research to address gaps in understanding NH_3 combustion across different systems, standardize experimental conditions, and refine economic and environmental impact assessments. Future studies should focus on exploring the behavior of NH_3 in diverse combustion environments, validating proposed strategies in real-world applications, and continuously updating the analysis in line with technological advancements and policy changes.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Ossi Kaario reports financial support was provided by Nordic Energy Research. Qiang Cheng reports financial support was provided by Merenkulun Säätiö. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The financial support from Nordic Maritime Transport and Energy Research Programme of Nordic Energy Research (NER) with the project name of Concepts of ammonia/hydrogen engines for marine application (CAHEMA) and Merenkulun Säätiö (Grant No. 20230052) with the project name of FlexiFuel Combustion: A Twin-Fluid Jet Strategy for Carbon-Neutral Combustion in Marine Engine. are acknowledged.

Data availability

Data will be made available on request.

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