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Carbon Neutrality

REVIEW

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Ammonia combustion and emissions in practical applications: a review

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Abstract

Ammonia is emerging as a viable alternative to fossil fuels in combustion systems, aiding in the reduction of carbon emissions. However, its use faces challenges, including NOx emissions and low flame speed. Innovative approaches and technologies have significantly advanced the development and implementation of ammonia as a zero-carbon fuel. This review explores current advancements in using ammonia as a fuel substitute, highlighting the complexities that various systems need to overcome before reaching full commercial maturity in support of practical decarbonising global strategies. Different from other reviews, this article incorporates insights of various industrial partners currently working towards green ammonia technologies. The work further addresses fundamental complexities of ammonia combustion, crucial for its practical and industrial implementation in various types of equipment.

Keywords Ammonia combustion, Ammonia emissions, Technology challenges, Alternative fuels, Applications

1 Introduction

Ammonia (NH_3) is a promising medium for energy storage of renewable energies and sustainable fuel for mobile and remote applications. It can be produced, similar to synthesised Hydrogen (H_2), from fossil fuels or renewable sources such as solar, biomass and wind [1]. Some advantages of ammonia over pure hydrogen include its lower cost of storage and transportation per unit of energy due to its higher volumetric density, easier handling and distribution, and better commercial viability [2, 3], which allows ammonia to be used as a zero-carbon hydrogen carrier [4]. Ammonia can directly be converted to electricity in a turbine, engine, or fuel cell. However, ammonia as energy storage faces four key challenges as illustrated in Fig. 1 [5]. Fig. 1 summarises the key challenges (or barriers) that require careful consideration, thus recognising urgent research and development needs for investigating the possibilities to improve the overall combustion properties of ammonia blends and address material science and component integrity before deploying any system commercially.

According to the International Energy Agency (IEA), ammonia production in 2020 accounts for 2% (8.6 EJ) of total final energy consumption and 1.3% (450 Mt) of CO_2 emissions as a consequence of ammonia production [6]. Ammonia production has significantly increased in response to the expanding ammonia market worldwide and diverse applications across various industries. Fig. 2 presents the historical data on ammonia production spanning from 1945 to 2022 and extends

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to include projections of its production up to the year 2050. The ammonia industry, considered one of the key industries for the global economy, faces strong challenges for the 2050 net-zero emissions scenario, hence it will need to reduce its emissions and increase sustainability. Since ammonia has been traded worldwide for more than 150 years, the maturity of the chemical's network is vast, making it the second most commercialised chemical on the planet. Therefore, knowing that ammonia can also be used for power generation and fuelling applications makes ammonia a great candidate to support the transition to a net zero-carbon economy. However, untacked emission-products of combustion processes are still a hot topic of research and development, topics that are approached in this work. The decarbonisation of ammonia production is discussed elsewhere, available in refs [6, 7].

Ammonia's adoption within the industrial scope as a source for generating power, heat, and propulsion through combustion systems is increasingly notable. Nonetheless, several relevant issues remain unaddressed, necessitating ongoing research and development efforts. These include the mitigation of emissions such as NOx, N_2O, residual ammonia, and carbon monoxide, particularly in scenarios involving co-firing. Simultaneously, enhancing the efficiency of these systems is imperative to improve their economic viability and further boost the advancement of ammonia-based technologies. This review, therefore, critically discusses the combustion fundamentals of ammonia, which are crucial for its application within industrial facilities, due to ammonia's


Key barriers for ammonia-based energy systems

Carbon-free synthesis of ammonia

This is critical because ammonia production methods are heavily reliant on fossil fuels and burning fossil fuels for this purpose severely releases carbon dioxide emissions into the Earth's atmosphere, which is extremely detrimental to the environment.

Power generation at utility-scale

This is important as most developments have focused on improving small-to-medium scale devices for transportation purposes. More importantly, pure ammonia combustion has several technical challenges include high auto-ignition temperature, low flame speed, narrow flammability limits, high heat of vaporization and high NOx emissions.

Public policy and safety regulations

They are essential to be implemented throughout health and safety impact analyses and the review of currently associated legalisation and end-user perceptions and acceptability.

Competitive economics

It is needed to undergo thorough economic studies in order to determine the potential of ammonia and its viability for use as energy systems.

Fig. 1 Key primary barriers that face viable energy system based on ammonia as the storage material

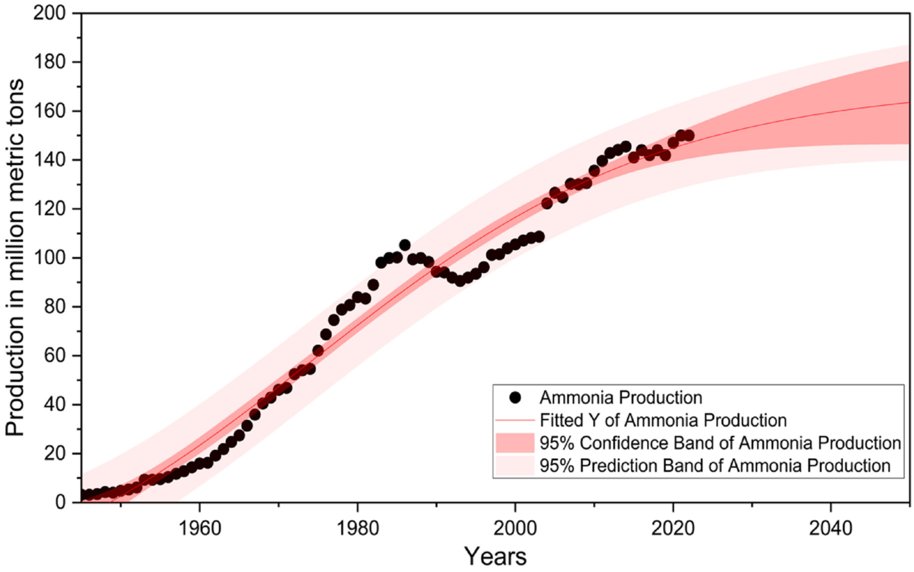

Fig. 2 Ammonia production from 1945 to 2022 and the future production estimates. Historical figures are available in refs [8, 9], while the predicted data has been modelled by the authors using the logistic growth function

potential as a sustainable energy vector that can significantly reduce carbon emissions in power generation and manufacturing process and help in transition into zero-carbon economy.

2 Combustion fundamentals

This section focuses on essential combustion principles acknowledged when utilising ammonia as a fuel source. While comprehensive discussions on this subject have been extensively documented in previous works [10–16], a thorough grasp of the technology demands an exploration of ammonia's unique combustion properties. This section includes aspects such as fuel sprays, ignition delay time (IDT), both laminar and turbulent burning velocity, extinction processes, and emissions (with emphasis on NOx). Furthermore, acknowledging some characteristics of ammonia is essential. The key properties of ammonia compared to other commonly used fuels are listed in Table 1.

Ammonia, as a potential renewable energy carrier, has recently gathered renewed interest in the energy sector, attributed to its high volumetric energy density and its practicality in terms of storage and distribution. The idea of using ammonia as a fuel date back to the early twentieth century, although pinpointing its first application is difficult due to its varied uses over time [21]. During World War II, Belgium powered buses with ammonia due to fuel shortages [21]. In the 1960s, the U.S.A. experimented with ammonia as fuel in the innovative X15 aircraft [22]. This era also saw investigations into the use of ammonia in gas turbines and reciprocating engines [23, 24]. The outcomes of these early studies remain unclear, but they laid the groundwork for the current interest in ammonia as a sustainable energy source.

Previous research has indicated that ammonia combustion has some drawbacks, such as low reactivity, slow burning, a limited flammability range, high auto-ignition temperature, and the high NOx emissions [17, 25, 26]. Ammonia's adiabatic flame temperature is lower than that of hydrogen and natural gas, recorded at 1800 °C (3272 °F), compared to 2110 °C (3812 °F) and 1950 °C (3542 °F), respectively. This lower temperature and the absence of CO_2 in the exhaust gases result in reduced radiative heat transfer, delaying combustion [27]. Ammonia's laminar burning velocity is also lower than that of hydrogen and natural gas, with recorded velocities of 0.07 m/s, 2.91 m/s, and 0.37 m/s, respectively [16, 28]. Its narrow flammability range poses additional ignition challenges. A major concern with ammonia combustion is the potential for NOx emissions. Although stoichiometric combustion of ammonia does not produce NOx, real-world conditions can lead to the formation of nitrogen-containing radicals and subsequent NOx emissions [28]. However, mature NOx removal technologies, such as selective catalytic reduction (SCR), can mitigate these emissions [29], and interestingly, ammonia from the fuel might be used in this process [26]. The risk of unburned ammonia is also a concern due to its toxicity [28], and ammonia can cause corrosion in materials, requiring careful material selection [24]. Several strategies have been developed to improve the combustion process, such as using gaseous ammonia, adding combustion additives, and employing swirlers and flame holders, all of which have shown to enhance combustion stability and efficiency while reducing NOx emissions [30–32]. Blending ammonia with other fuels is another area of exploration [11, 33], where the secondary fuel may act solely as an ignition aid or be part of a continuous blend. Efficiency improvements in ammonia combustion have been reported with the addition of auxiliary fuels like hydrogen and methane [34–36].

2.1 Fuel sprays

Ammonia is usually stored as a liquid and can be supplied to combustion systems in both gaseous and liquid forms. Extensive research has explored the use of gaseous ammonia for premixed or diffusion flames [37, 38]. However, there is potential for employing atomised liquid ammonia spray, offering benefits like shorter start-up times and reduced costs compared to its vapourised

Table 1 Key properties of ammonia and common fuels [17–20]

	Ammonia	Hydrogen	Methane	Propane	Methanol	Ethanol	Gasoline
Boiling temperature at 1 atm (℃)	-33.4	-253	-161	-42.1	65	79	25–215
Lower heating value (MJ/kg)	18.6	120.0	50.0	46.4	20.1	26.9	42.9
Flammability limit (Equivalence ratio)	0.63 ~ 1.4	0.10 ~ 7.1	0.50 ~ 1.7	0.51 ~ 2.5	0.55 – 2.9	0.66 – 2.4	0.7 – 0.4
Adiabatic flame temperature (℃)	1800	2110	1950	2000	1870	1920	2000
Auto-ignition temperature (℃)	650	520	630	450	465	425	192
Maximum laminar burning velocity (m/s)	0.07	2.91	0.38	0.43	0.50	0.47	0.58
Minimum ignition energy (MJ)	8.00	0.02	0.28	0.25	0.14	0.28	0.14

form [39]. Early attempts at direct liquid ammonia combustion in gas turbines [40, 41] were plagued by instabilities and frequent flame blow-outs. These challenges were occurred due to ammonia's high latent heat of vaporisation, low flame speed, and heating value. The low flame temperature of ammonia also poses a risk of inefficient N_2O to N_2 conversion, contributing to greenhouse emissions [42]. In order to mitigate these issues, it was suggested to co-fire liquid ammonia spray with faster-burning fuels such as hydrogen and methane, enhancing combustion stability and reducing N_2O emissions [13, 43].

Okafor et al. [39] applied the Mie scattering technique to observe ammonia spray characteristics in non-reacting conditions without and with airflow as shown in Fig. 3. They cooled the ammonia pipeline to prevent fuel vaporisation, eliminate cavitation in the nozzle, and facilitate stable combustion of liquid ammonia. The non-reactive liquid ammonia spray reached a height exceeding 150 mm in the absence of air flow as shown in Fig. 3a. However, this extent was not fully visible in the illustration due to the restricted height of the illuminating laser sheet used in the experiment. When combined with swirling preheated air, the height of liquid spray ammonia significantly reduced (Fig. 3b), allowing sufficient fuel vaporisation, mixing, and preheating. Airflow was also found to improve spray dispersion. In their study, they successfully stabilised liquid ammonia spray flame, as demonstrated in Fig. 4a. The liquid ammonia spray flame with a significant height (> 150 mm) was observed, while the flame volume within the combustor is small. In contrast, ammonia spray co-fired with methane resulted in a more stable and voluminous flame as shown in Fig. 4b. Another recent work [44] investigated a two-stage liquid ammonia spray combustion in a 230kW gas turbine combustor, and it was reported that sufficient compensation for heat extracted from droplet vaporisation was necessary to improve efficiency in the primary combustion zone, thereby reducing the quantity of unburned ammonia that was oxidised in the secondary zone.

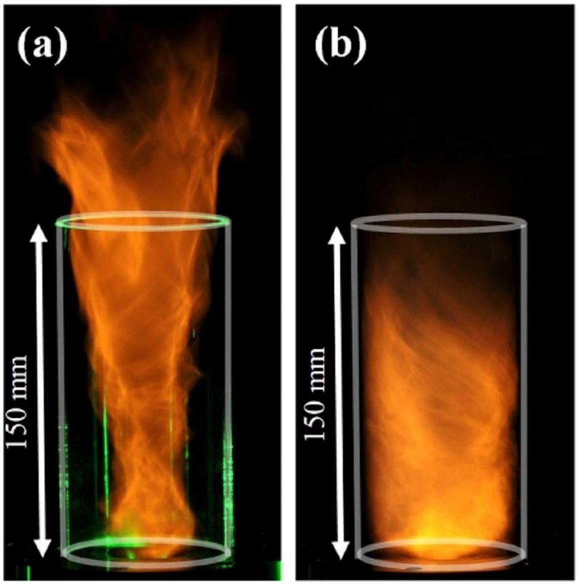

Fig. 4 Liquid ammonia spray flame at equivalence ratio = 0.9, inlet air temperature = 500 K, swirler mean inlet velocity = 21 m/s (**a**) pure ammonia (**b**) ammonia (70% in total energy) and methane [39]. (Reprinted from Fuel, 2021, Vol 287, Okafor, E.C., et al., Flame stability and emissions characteristics of liquid ammonia spray co-fired with methane in a single stage swirl combustor, Pages No.119433, Copyright (2023), with permission from Elsevier)

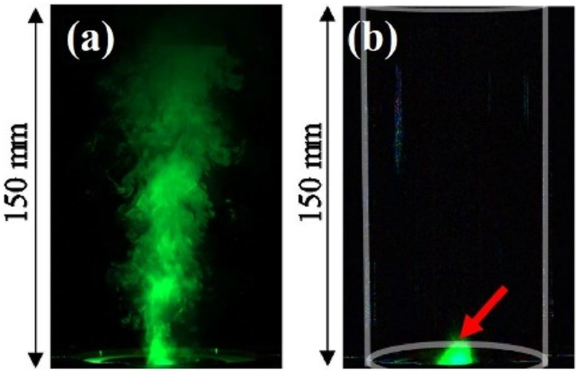

Fig. 3 Liquid ammonia spray (**a**) without airflow (NH3 flow rate = 10 g/min, instantaneous image); (**b**) with airflow (NH3 flow rate = 10 g/min, inlet air temperature = 500 K, swirler mean inlet velocity = 21 m/s, time-average of 50 images). Image is adjusted from [39] (Reprinted from Fuel, 2021, Vol 287, Okafor, E.C., et al., Flame stability and emissions characteristics of liquid ammonia spray co-fired with methane in a single stage swirl combustor, Pages No.119433, Copyright (2023), with permission from Elsevier)

Direct ammonia injection in engines typically results in a two-phase flow due to its low boiling point [45]. The atomised droplets' size is critical for effective fuel vaporisation, combustion, and emissions. Flash boiling of ammonia, occurs when the subcooled liquid is depressurised promptly below its saturation pressure, leads to a more uniform air/fuel mixture, comprising bubble nucleation, growth, and two-phase flow stages [46]. Comparative studies on ammonia sprays with other fuels like biofuel, ethanol, and gasoline showed distinct differences in spray shape and penetration. Pelé et al. [47] applied the Schlieren imaging to capture both liquid and liquid/vapuor ammonia sprays (Fig. 5a), and compared the spray penetration length with those of biofuel, ethanol, and gasoline (Fig. 5b).

Further studies on ammonia sprays have been conducted. Li et al. [48] investigated the characteristics of superheated ammonia spray at various fuel temperatures

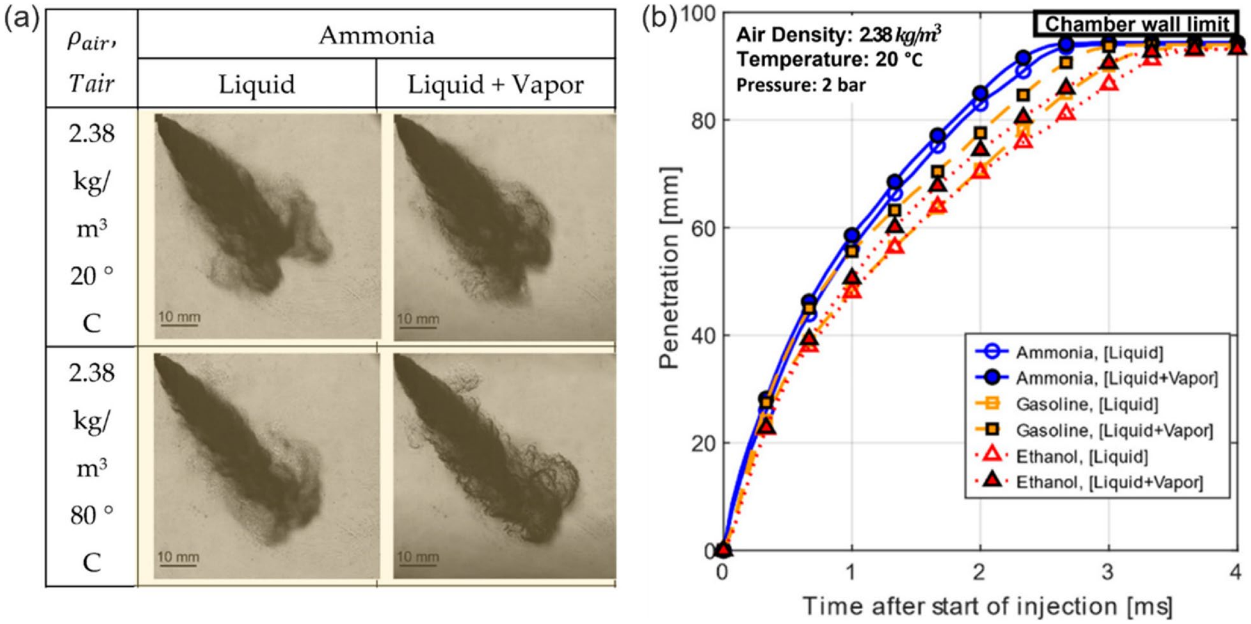

Fig. 5 Comparisons of liquid and liquid/ vapour NH3 sprays; (**a**) Spray shape and (**b**) Spray penetration. (Figure is adjusted from [47])

and ambient densities within a constant volume chamber (CVC). They found that the intensity of spray bubble explosions near the nozzle is dominated by the degree of superheating and the viscosity of the fuel across the flashing region. In terms of spray penetration, ammonia is found similar to diesel under both critical and initial flashing conditions. Similarly, Cheng et al. [49] employed Schlieren imaging to investigate the ammonia spray evolution processes in a CVC. The quantified spray characteristics (e.g., spray cone angle, penetration length, cross-sectional area, etc.) were compared with other fuels such as methanol and ethanol. Overall, ammonia showed longer penetration, a wider spray cone angle, and a larger cross-section area, alongside a faster evaporation rate compared to these fuels. Fig. 6a illustrates that an increase in injection pressure (40 to 100 bar) results in a longer spray penetration length, larger spray area, and a more turbulence within the spray. Conversely, as

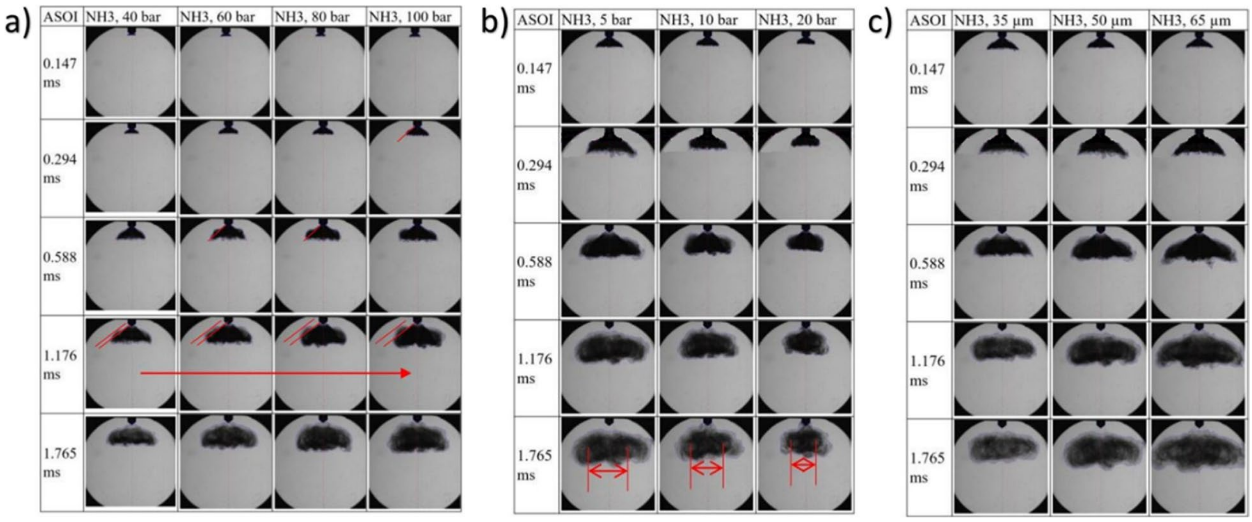

Fig. 6 Ammonia spray evolution process at (**a**) different injection pressure; (**b**) different chamber pressure; and (**c**) different injector needle lift. (Adjusted from [49, 50])

depicted in Fig. 6b, a rise in chamber pressure shortens the spray's penetration and reduces its area, likely due to the increased air drag force caused by higher ambient density. Furthermore, enhancing the injector needle lift leads to greater spray penetration and a more substantial spray area, as indicated in Fig. 6c.

2.2 Ignition delay time

The ignition delay time is a critical parameter for evaluating fuel combustion performance, which is equivalent to the prior-ignition duration of a homogeneous mixture under specific pressure and temperature conditions. Similar to other fundamental properties of combustion, the ignition delay is a key indicator of fuel reactivity and combustion response, frequently used in developing chemical kinetic mechanisms and predicting flame behaviours at specified conditions. Compared with conventional hydrocarbon fuels (e.g., gasoline, diesel, methane, etc.), ammonia presents higher ignition temperature and ignition energy, posing challenges for practical applications. This necessitates an exploration of its combustion characteristics and potential enhancement strategies.

Initial studies of ammonia's ignition delay began in the 1960s–80s by researchers such as Takeyama and Miyama [51, 52], Bradley et al. [53], and some subsequent studies [54, 55] employed shock waves to measure NH3 induction period in O_2/Ar mixtures. However, these early attempts lacked the precision and repeatability needed reliable model validation. Later, Mathieu and Petersen [56] expanded the experimental scope, providing a broader data range on NH3 (pressure from 0.14 to 3 MPa, temperature from 1560 to 2490 K, equivalence ratio from 0.5 to 2.0) and detailed information on ammonia's high-temperature oxidation of NH3 and NOx chemistry, leading to the development of a more accurate kinetic model for ammonia ignition delay and NOx emissions prediction. Shu et al. [57] further extended the pressure range for ammonia ignition delay measurements up to 40 bar, revealing significant pressure-related variation (i.e., the ignition delay was shortened with the increases in pressure, and results showed some differences with N2 and Ar mixtures).

Ammonia's low burning velocity (~0.42% of hydrogen) and high ignition energy (~4440 times of hydrogen) limit its applications in transportation. Its long ignition delay makes it inappropriate for singularly powering the engines running at high speeds. Hence, blending ammonia with ignition promotors (i.e., hydrogen, hydrocarbon fuels, DME, etc.) to enhance fuel reactivity and combustion performances. Many studies focused on the fundamental properties of NH3 fuel blends, particularly with zero-carbon NH3/H2 mixtures. Pochet et al. [58] investigated ammonia's ignition delay with hydrogen addition (0%, 10%, and 25%vol.) in an rapid compression machine at low temperature combustion conditions (low equivalence ratios: 0.2, 0.35, 0.5; high pressures: 43 and 65 bar; low temperatures: 1000–1100 K). The results showed that the hydrogen fraction must be higher than 10% to produce a significant promotion of the ignition delay. The equivalence ratio effect on the ignition delay of ammonia blends was obvious. Li et al. [59] implemented numerical investigations on the ammonia ignition delay at various hydrogen blending levels. They claimed that increasing the pressure and H2 addition ratios could promote NH3 ignition and decrease the ignition delay. Similarly, Chen et al. [60] reported that H2 addition changed the ammonia delay nonlinearly. Only 5% of the H2 blending ratio could reduce this value by about 60% relative to the pure ammonia at both pressures of 1.2 and 10 atm and temperature at 1600 K. Li and Chen's studies further confirmed the positive impact of hydrogen addition on reducing ammonia's ignition delay, highlighting the non-linear relationship and the significant reduction in delay with even modest hydrogen blending [60].

Further studies have also explored ammonia's ignition delay when mixed with hydrocarbon fuels. Dai et al. [61] examined NH3/CH4 mixtures with CH4 fractions up to 50%. CH4 exhibited a strong ignition-enhancing effect on NH3, which levels off at higher fractions, as the ignition delay time approaches that of pure CH4. Xiao et al. [62] studied the ignition characteristics of NH3/CH4 mixtures at different equivalence ratios (0.5, 1, and 2), different pressure (2 and 5 atm), and temperature (from 1369 to 1804 K) conditions. They found that the ignition delay to be more sensitive to CH4 at lower concentrations, with the effect of diminishing at higher ratios as shown in Fig. 7. In contrast, the ignition delay reduction was much slighter with a higher CH4 ratio at pressures of 2 atm and 10 atm. Fig. 8 shows that the mixture ignition delay decreased remarkably with increasing the initial temperature and mixture pressure. The numerical results presented that the mixture ignition delay rises slightly with increasing the equivalence ratio (see Fig. 9), which is consistent with the experimental results. Moreover, a sensitivity analysis performed in this study showed that the mixture ignition delay is most sensitive to the reaction of $H + O2 \leftrightarrow OH + O$, and the generated OH radicals promoted fuel consumption.

Recently, another study conducted by Dai et al. [63] revealed that mixing ammonia with Dimethyl Ether (DME) noticeably affects the autoignition delay time of ammonia. Their findings show a notable shift in the ignition delay versus temperature curves for ammonia/DME blends, with differences of approximately 250 K compared to pure ammonia. Their study contributed to the development of an innovative reaction

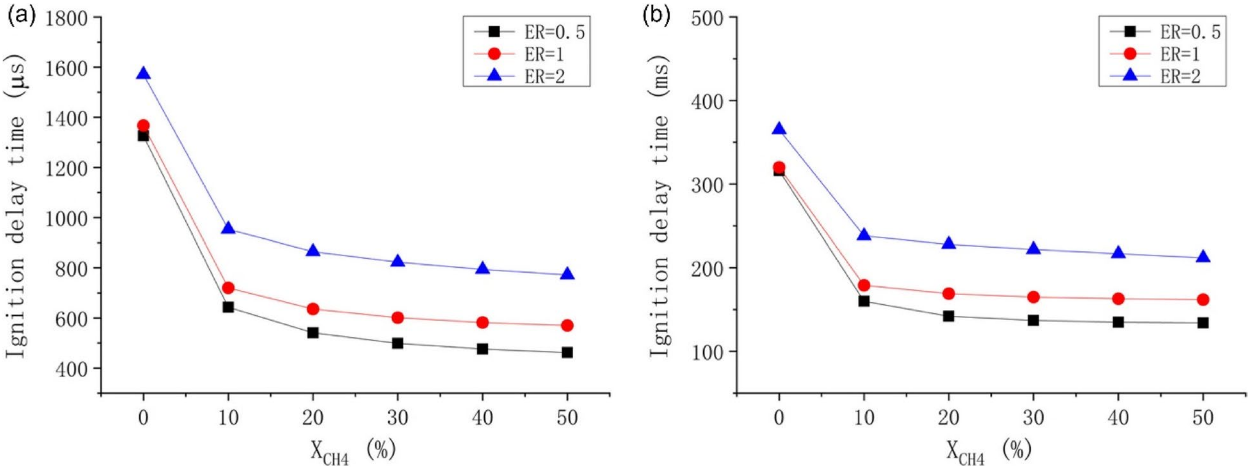

Fig. 7 The CH4 concentration effect on ignition delay of the NH3/CH4/air mixtures at T = 1550 K. (**a**) P = 2 atm. (**b**) P = 10 atm. (Figure is adjusted from [62]) (Reprinted from Int J Energy Res., 2020, Vol 44, Xiao, Hu., et al., Experimental and modelling study on ignition delay of ammonia/methane fuels, Pages No.6939–6949, Copyright (2023), with permission from Wiley)

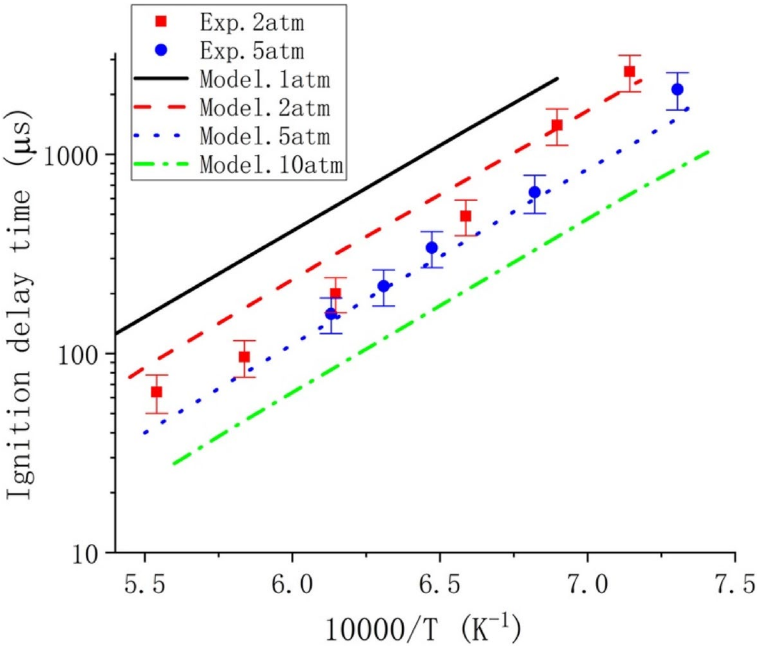

Fig. 8 The pressure effect on ignition delay of the NH3/CH4/air mixtures (φ = 1, XCH4 = 40%). (Figure is adjusted from [62]) (Reprinted from Int J Energy Res., 2020, Vol 44, Xiao, Hu., et al., Experimental and modelling study on ignition delay of ammonia/methane fuels, Pages No.6939–6949, Copyright (2023), with permission from Wiley)

mechanism for simulating these specific blends. Fig. 10 clearly compares the effects of CH4 (Fig. 10a) and DME (Fig. 10b) additions on ammonia at identical conditions (equivalence ratio: 0.5, 6.0 MPa). Another investigation of NH3/DME mixtures by Issayev et al. [64] concluded that adding DME as low as 5% could significantly enhance the ignition properties and reduce the ignition delay of ammonia.

Recent research has investigated the effects of adding heavier hydrocarbons to ammonia. Yu et al. [65] investigated the autoignition of NH3 blended with up to 60% n-heptane (C7H16) in an O2/Ar environment. Utilising a Rapid Compression Machine (RCM) at pressures of 1.0 and 1.5 MPa and temperatures ranging from 635–945 K, they focused on equivalence ratios of 1 and 2. Although this study contributed to the development of

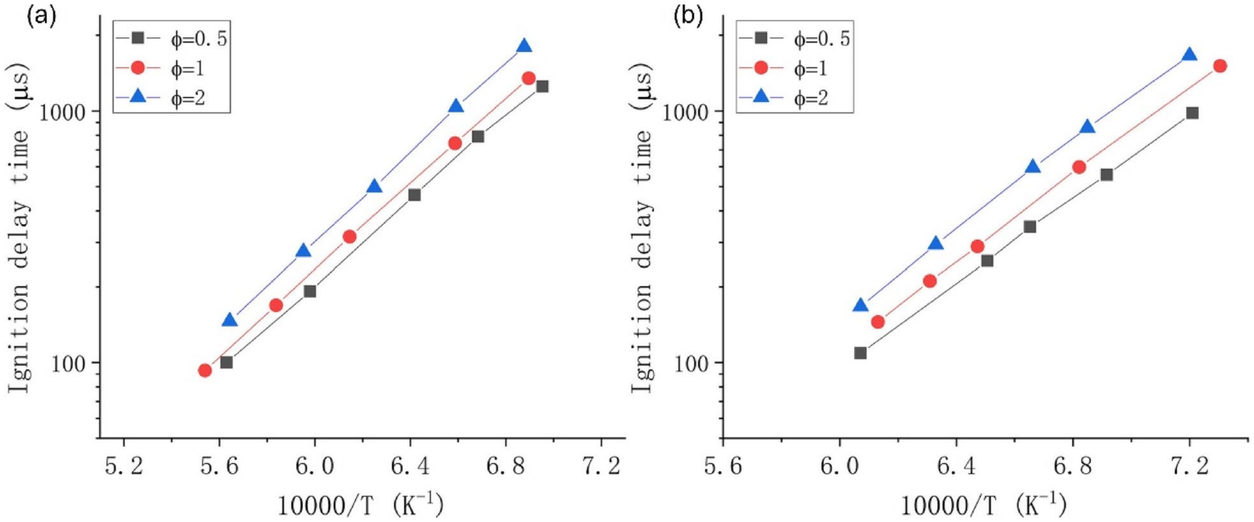

Fig. 9 The equivalence ratio effect on the NH3/CH4/air mixtures ignition delay. (**a**) P = 2 atm. (**b**) P = 5 atm. (Figure is adjusted from [62]) (Reprinted from Int J Energy Res., 2020, Vol 44, Xiao, Hu., et al., Experimental and modelling study on ignition delay of ammonia/methane fuels, Pages No.6939–6949, Copyright (2023), with permission from Wiley)

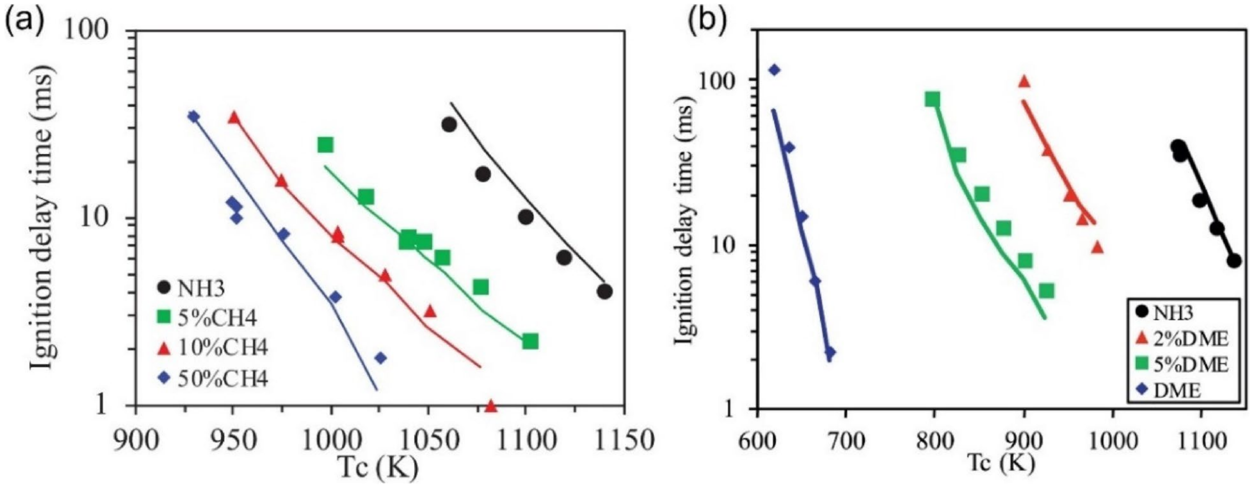

Fig. 10 A comparison between the change in ignition delay time for NH3 (equivalence ratio: 0.5, 60 bar) when blended with **a**) CH4 [61] and **b**) DME [63]. Markers represent experimental data and lines from numerical simulation. (Fig 10.A Reprinted from Combustion and Flame, 2020, Vol 218, Dai, L., et al., Autoignition studies of NH3/CH4 mixtures at high pressure, Pages No.19-26, Copyright (2023), with permission from Elsevier). (Fig 10.B Reprinted from Combustion and Flame, 2021, Vol 227, Dai, L., et al., Ignition delay times of NH3 /DME blends at high pressure and low DME fraction: RCM experiments and simulations, Pages No.120-134, Copyright (2023), with permission from Elsevier)

a new reaction mechanism, it noted considerable differences between the experimental results and simulations. In another study, Li et al. [66] explored blending ammonia with methanol (CH3OH), varying the proportions up to 100%. Their experiments, conducted at pressures of 2.0 and 4.0 MPa and temperatures between 845–1100 K, covered equivalence ratios of 0.5, 1, and 2. Similarly, Feng et al. [67] examined the effects of mixing diesel with ammonia, with diesel concentrations reaching up to 50%. This research, carried out at pressures from 1.0 to 2.0 MPa and temperatures of 670–910 K, spanned equivalence ratios from 0.5 to 1.5, using an RCM. Each of these studies offered insights into how different hydrocarbons alter NH3's autoignition behaviour. Notably, even a small addition of 1% CH3OH to ammonia could lead to a significant reduction in ignition temperature, approximately 100K. This highlights the substantial influence that hydrocarbon blends can have on ammonia's ignition characteristics, a topic that continues to be a rich area for ongoing research.

2.3 Laminar burning velocity

Flames are produced by a sustainable chemical reaction where the unburnt air/fuel mixtures are heated and converted into products. The Laminar Burning Velocity (LBV) is an essential concept in combustion theory, representing the rate at which flame front expands relative to the quiescent unburned gas. LBV has been extensively studied over decades due to its critical importance in understanding combustion reactions. The speed of flame propagation is typically faster for fuels with higher LBV values [68]. LBV is defined by the following equation [69]:

$$V = \sqrt{\alpha w/\rho} \tag{1}$$

where α (cm^2/sec) and ρ (g/cm^3) are the thermal diffusivity and density of the combustion mixture, respectively. w (g/cm3 sec) is the empirical rate of the reactant mixture conversion.

The LBV contains information relevant to thermodynamics, chemical kinetics, and species transport, which is of great importance for studying the reaction mechanisms of ammonia-premixed flames [70–72]. Early investigations include Partington et al.'s 1924 study [73] on ammonia combustion and explosion characteristics. The first measurement of NH3/O2 mixtures' LBV was by Murray and Halle [74] in 1951. At almost the same time, Ausloo and Tiggelen [75] used the Bunsen flame area method to estimate the LBV of NH3/O2 and NH3/N2O/N2 mixtures with different nitrogen fractions. Later in 1955, Cohen [76] examined ammonia's LBV at high temperatures, finding values of 0.23 m/s and 0.15 m/s for ammonia fractions of 22% and 22.5%, respectively. Further studies [77–79] compared ammonia's LBV with other fuels, such as methane (CH4) and propane (C3H8). Fig. 11 illustrates how ammonia flame velocity varies with its concentration in the air. Notably, ammonia's maximum flame velocity, lower than 0.08 m/s, is about one-fifth that of hydrocarbon fuels due to its lower reactivity. Despite advancements in measurement techniques, there remains a deviation of ~ 25% between different determination methods, especially in fuel-rich mixtures. However, current mechanisms effectively reflect these trends, as shown in Fig. 11. The following sections summarise findings concerning the temperature and pressure dependencies of ammonia LBV, as well as the effects of increased oxygen concentrations and variations in mixtures.

2.3.1 Pressure dependence

Recent research by Kanoshima et al. [81] focused on measuring the LBVs of NH3/air flames at elevated pressures. Their findings, alongside those of Hayakawa et al. [82], closely align with the numerical simulations using the detailed reaction mechanisms by Otomo [83] and Han [84], and the reduced mechanism by Okafor [85]. As shown in Fig. 12, the LBV peaks around an equibalance ratio of 1.1 across various cases, with the peak value decreases with higher initial pressure. Furthermore,

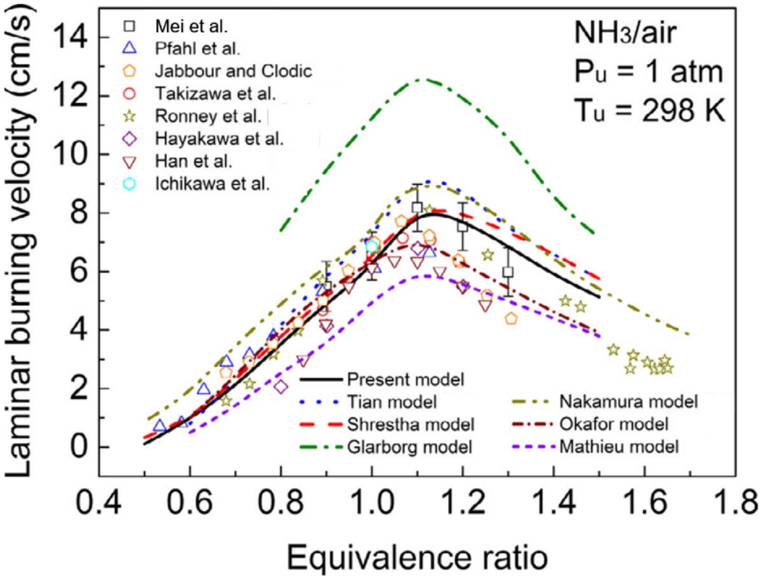

Fig. 11 Flame velocity variations with ammonia content in the air [80]. (Reprinted from Combustion and Flame, 2019, Vol 210, Mei, B., et al., Experimental and kinetic modelling investigation on the laminar flame propagation of ammonia under oxygen enrichment and elevated pressure conditions, Pages No.236-246, Copyright (2023), with permission from Elsevier)

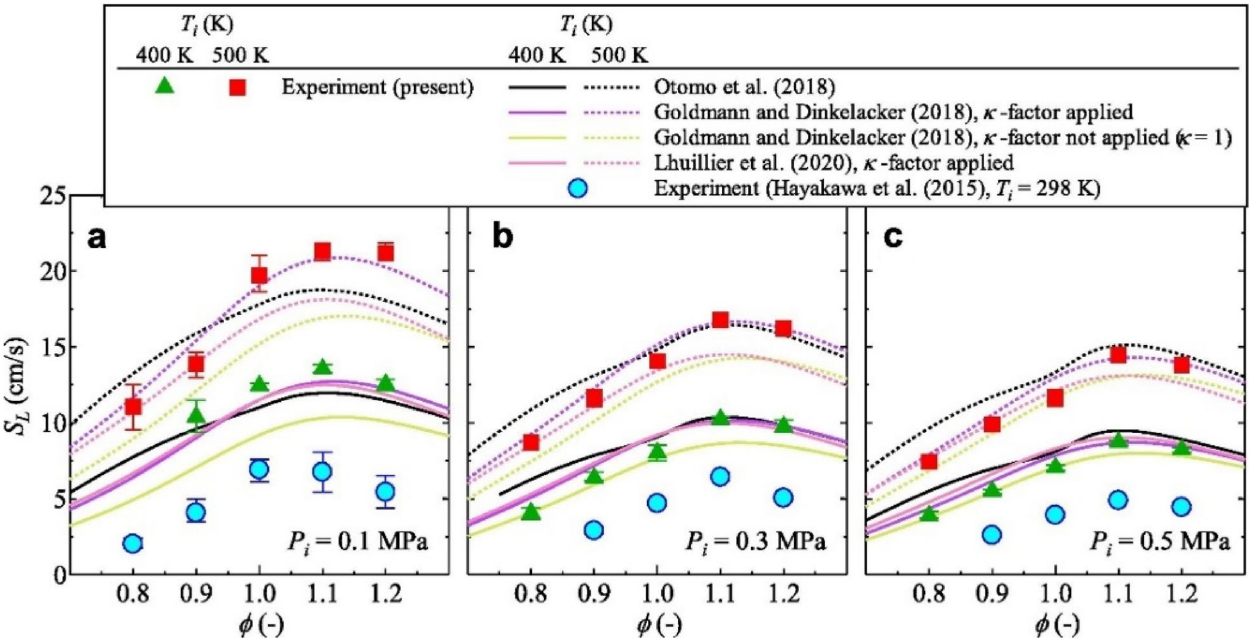

Fig 12 Experimental and numerical values of unstretched laminar burning velocities of ammonia/air flames: (**a**) Pi = 0.1 MPa; (**b**) Pi = 0.3 MPa; (**c**) Pi = 0.5 MPa [81]. (Reprinted from Fuel, 2022, Vol 310, Part B, Kanoshima, R., et al., Effects of initial mixture temperature and pressure on laminar burning velocity and Markstein length of ammonia/air premixed laminar flames, Pages No.122149, Copyright (2023), with permission from Elsevier)

Hayakawa et al. [82] studied the LBVs of NH3/air flames at elevated pressures using five detailed mechanisms [86–90]. Their experimental results confirmed that the burning velocity decreases as initial pressure increases for equivalence ratios of 0.9, 1.0, and 1.2. Notably, the trends in both experimental and simulation results mirror those observed in hydrocarbon flames, contrasting with Ronney et al. [71] finding, which indicated that little pressure influence on burning velocity. Particularly at an equivalence ratio of 1, the initial pressure was found to be significantly affected the flame propagation speed. Jin et al. [91] found a similar phenomenon in CH4-NH3 mixtures, attributing the LBV decrease to changes in the combustible fuel volume, which alter the mixture's density with varying initial pressure, and impacts on chemical reactions. Their investigation into the Rate of Production (ROP) revealed that the peak concentrations of radicals such as H, NH2, NO, NO2, and HCN generally decrease with increasing initial pressure. While the peaks of H, NH2, and NO initially rise then fall with increasing equivalence ratio, the peak of NO2 continuously drops and that of HCN consistently rises.

2.3.2 Temperature dependence

The literature on how temperature influences the LBV of ammonia flames is relatively limited. Han et al. [92] used a heat flux burner to investigate ammonia flames across a temperature range from 298 k to 448 K, incrementing in 7K steps. They observed an increase in LBV from ~ 7 cm/s to ~ 15 cm/s, with the peak consistently occurring at an equivalence ratio (ϕ) of 1.1. From this data, they derived a temperature coefficient, which varied from 2.5 in the lean region to 1.85 at an equivalence ratio of 1.1. Lhuillier et al. [93] conducted a study on ammonia flames under spark-ignition (SI) engine conditions, introducing hydrogen and methane additions up to 15% and examining equivalence ratios between 0.9 to 1.1. In their further experiments, they increased both pressure (to 5.4 bar) and temperature (to 445 K) simultaneously. Shrestha et al. [94] further extended the scope of research by exploring temperature ranges up to 473 K, pressures up 10 to bar and hydrogen ratios up to 30%, and varying oxygen ratios up to 30% in a spherical stainless steel combustion chamber. Their findings indicated that a 30% addition of both oxygen and hydrogen could approximately triple the LBV of ammonia (by a factor of ~ 2.8). Despite these advancements, discrepancies persist in the data, particularly in fuel-rich mixtures. The lack of high-temperature data points highlights the need for further experimental work with varied setups to improve the validation of data and understanding in this area.

2.3.3 Oxygen content dependence

Considering ammonia's inherently low burning velocity and the limitations of certain experimental setups, researchers have developed methodologies to augment its burning velocity. One approach is the blending with faster-burning fuels, but another significant method is the enrichment of the oxidiser with oxygen. Li et al. [95] investigated the effects of increased oxygen concentrations, up to 30%, in ammonia flames. They found that LBV could be enhanced by as much as 2.6 times compared to combustion with air. Liu et al. [96] conducted experimental studies on ammonia-oxygen flames at ambient conditions, noting the highest LBV at around an equivalence ratio of 0.8, which is more characteristic of lean mixtures, reaching speeds exceeding 100 cm/s. Sun et al. [97] performed numerical investigations using the ND mechanism to validate their results against Liu et al.'s [96] experimental data. They found good agreement in the lean and stoichiometric regions, though the model tended to overpredict in the rich mixtures. Mei et al. [80] further examined the impact of oxygen content, increasing it to 45% and confirmed the nearly linear trends observed by Li et al. Their studies extended to pressures of up to 5 atm, confirming the decreasing trend of LBV in air and revealing that the Markstein length tends to increase for $\phi < 0.8$ and decrease for $\phi > 0.8$ with higher pressures. Expanding on this research, Zhang et al. [98] used the same experimental apparatus to compare the effects of He and N2 in O2-enriched mixtures (as illustrated in Fig. 13). Their experiments, conducted at pressures up to 7 atm, demonstrated higher LBVs in mixtures containing helium.

2.3.4 Mixture dependence

Ammonia's inherently low reactivity and challenging ignition characteristics have led to its combination with ignition promoters like hydrogen and various hydrocarbon fuels, including methane, gasoline, and DME. The type of fuel blended with ammonia plays a critical role in influencing its burning velocity. Research has revealed that the equivalence ratio (ϕ) has a particularly an extraordinary impact on the unstretched LBV of ammonia. Typically, the LBV reaches its peak around $\phi = 1.1$, consistent across different initial pressures [80, 99], as illustrated in Fig. 14. The addition of other fuels, akin to hydrogen, has been found to enhance the LBV of ammonia flames. This includes mixtures with methane (CH4) [86, 100, 101], carbon monoxide (CO) [84], syngas [102], and dimethyl ether (DME) [103, 104]. In this context, Wang et al. [102] conducted a study to dissect the thermal, diffusion, and chemical effects influencing the LBV when ammonia is mixed with these fuels. Their findings indicated that chemical and diffusion effects primarily govern the variations in LBV. Furthermore, the impact of ammonia chemistry becomes more significant under fuel-rich conditions. The subsequent sections will delve into these mixing effects on the LBV in greater detail.

Ammonia mixture with hydrogen Lee et al. [105, 106] conducted studies on the LBVs of ammonia-hydrogen mixtures at 0.1 MPa, reporting that an increase in hydrogen concentration resulted in higher LBVs. In another work, Han et al. [98] used a heat flux burner to investigate the impact of adding hydrogen, carbon oxide and methane, on NH_3/air mixtures. They found that with up to 40% hydrogen, the maximum LBV shifted toward slightly richer conditions compared to pure ammonia. Lhuillier et al. [107] observed a significant increase in LBVs of ammonia flames, increased from 7 cm/s (0% H_2) to approximately 80 cm/s, when the hydrogen ratio was raised from 0 to 60% at 298 K, as can be seen in Fig. 15.

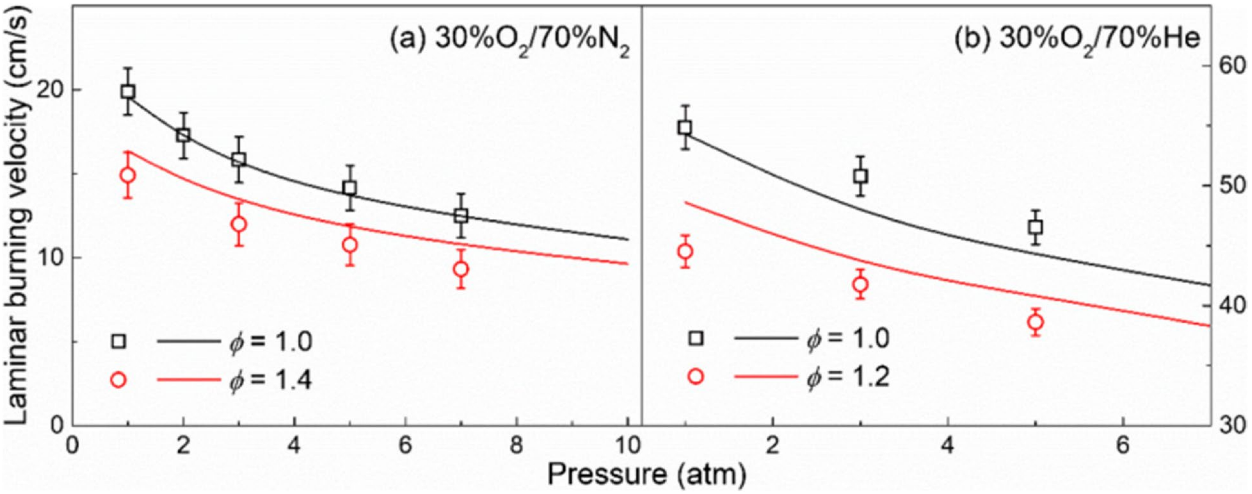

Fig. 13 Measured and simulated LBVs of NH3/O2 at T=298 K, P= 1−7 atm, with a dilution of **a**) N2 and **b**) He [98]. (Reprinted from Energy & Fuels, 2022, Vol 36(15), Zhang, J., et al., Unravelling Pressure Effects in Laminar Flame Propagation of Ammonia: A Comparative Study with Hydrogen, Methane, and Ammonia/Hydrogen, Pages No.8528-8537, Copyright (2023), with permission from American Chemical Society)

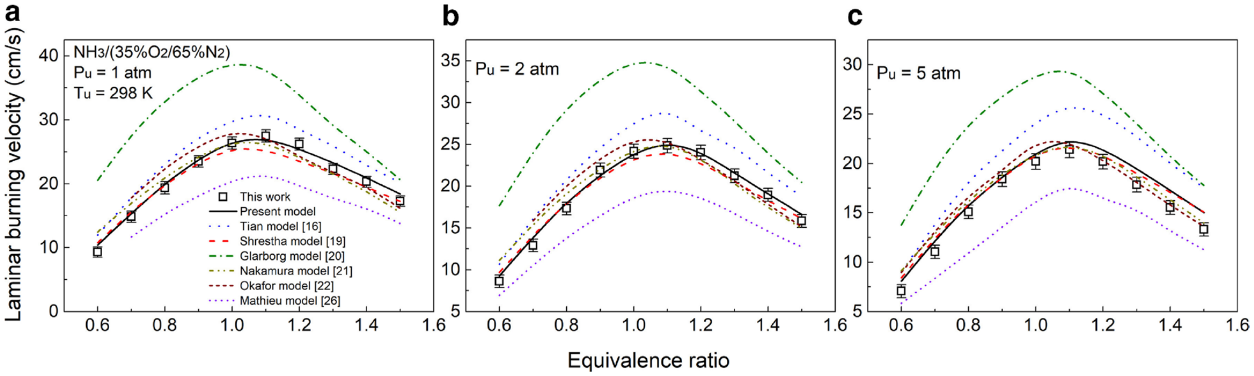

Fig. 14 LBVs of NH3/(35%O2/65%N2) mixtures at Tu = 298 K and Pu = 1–5 atm [80]. (Reprinted from Combustion and Flame, 2019, Vol 210, Mei, B., et al., Experimental and kinetic modelling investigation on the laminar flame propagation of ammonia under oxygen enrichment and elevated pressure conditions, Pages No.236-246, Copyright (2023), with permission from Elsevier)

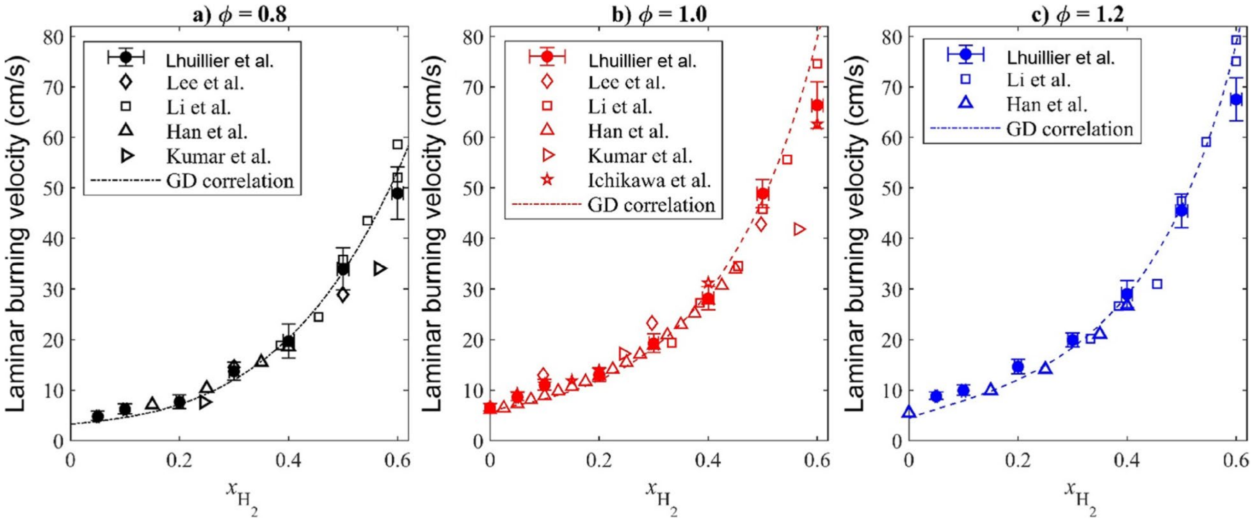

Fig. 15 Variations of ammonia LBVs with hydrogen mole fraction regarding different equivalence ratios (T=298 K, p=1 bar) [107] (Reprinted from Fuel, 2020, Vol 263, Lhuillier, C., et al., Experimental investigation on laminar burning velocities of ammonia/hydrogen/air mixtures at elevated temperatures, Pages No.116653, Copyright (2023), with permission from Elsevier)

Other findings [99] have shown that the LBV of ammonia can reach levels comparable to methane (CH4) as the hydrogen ratio of NH_3/H_2 flames increases from 33.3% to 60.0%. This phenomenon was explained based on the numerical study by Li et al. [59], which resulted from the reduction of chemical activation energy and transport effect caused by the high mobility of H_2. Three reactions contribute to these effects: $O+H_2 \leftrightarrow OH+H$, $H+O_2 \leftrightarrow OH+O$, and $H_2+OH \leftrightarrow H_2O+H$. These reactions can significantly increase the concentration of free radicals and accelerate the peak of H, O, and OH radicals. Consequently, adding hydrogen to ammonia flames is claimed to improve engine performance [108] and gas turbine applications [109, 110].

Ammonia mixture with methane Berwal et al. [111] observed a linear decrease in LBV with increasing NH3 concentrations across different mixture equivalence ratios and temperatures, as shown in Fig. 16. They particularly noted that the LBVs at rich conditions tended to align with those at lean conditions when the mixture temperature was increased. In a separate study, Okafor et al. [101] measured the unstretched LBV of premixed NH_3/CH_4 flame in a constant volume chamber. The ammonia concentration varied from 0 to 0.3, and the equivalent ratio ranged from 0.8 to 1.3. The results showed that a shift from a linear to a nonlinear relationship between flame velocity and flame stretching rate with increased ammonia concentration and equivalent ratio. This line

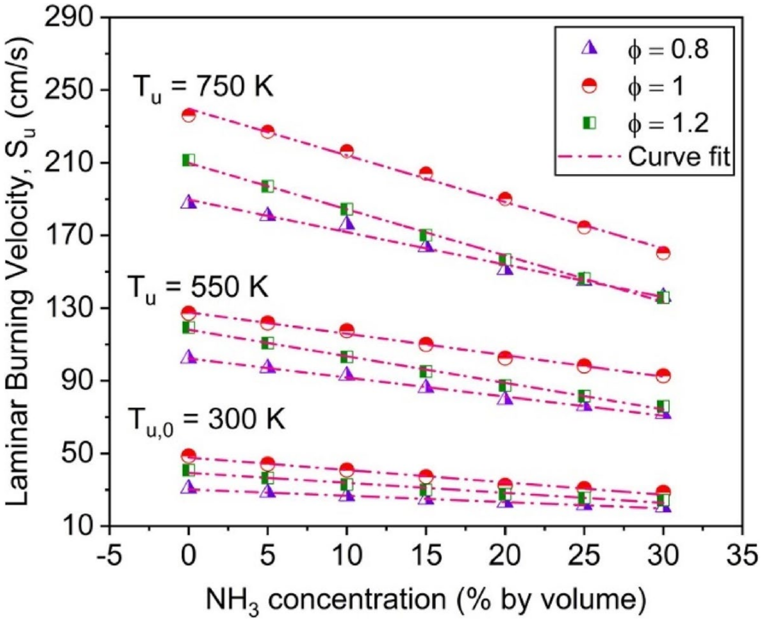

Fig. 16 Variation of laminar burning velocity with NH3 concentration at different mixture temperatures (T = 300 K, 550 K and 750 K. P = 0.1 MPa) [111]. (Reprinted from Fuel, 2023, Vol 331, Berwal, P., Shawnam, and S. Kumar, Laminar burning velocity measurement of CH4/H2/NH3-air premixed flames at high mixture temperatures, Pages No.125809, Copyright (2023), with permission from Elsevier)

of research was further advanced by Okafor et al. [85], who extended their investigations to pressures of 3 and 5 bar (see Fig. 17). Ku et al. [112] explored the potential of methane ammonia blends in expanding spherical premixed flame, measuring Markstein numbers, flame structures and LBVs. Their results showed good alignment with the findings of Okafor et al. [101]. Further, Shu et al. [113] expanded the scope of these parameters to higher pressures, up to 5 bar. They observed a strong linear correlation between LBV and methane volume fraction in ammonia-methane mixtures with the same equivalence ratio. Rocha et al. [114] employed a cone flame to study

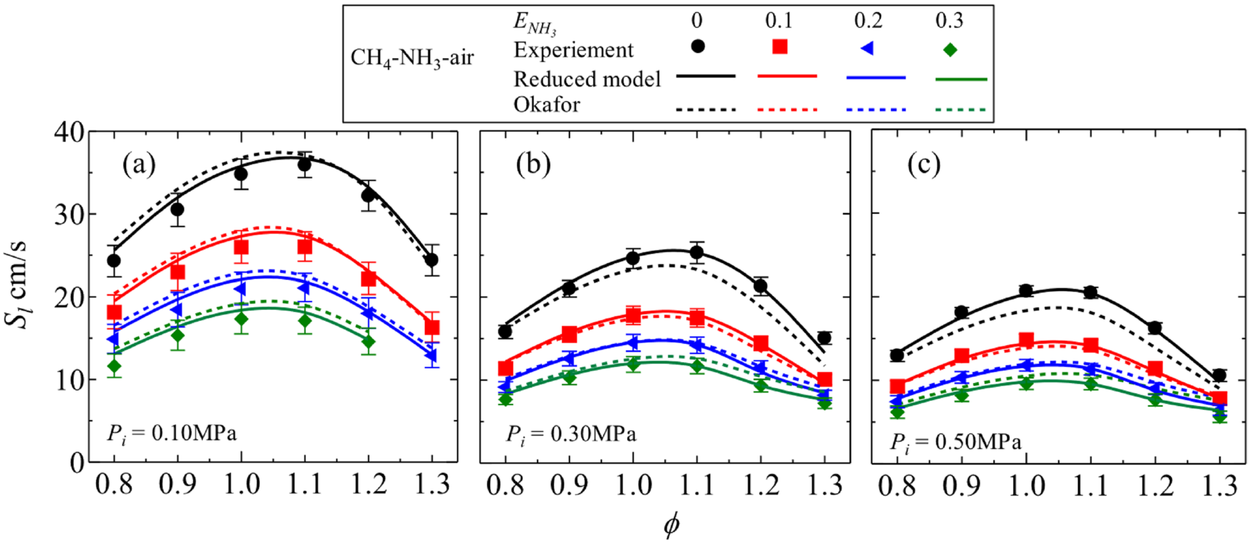

Fig. 17 Comparison of LBV of CH4 –NH3 -air flames to simulated values at (**a**) 0.10 MPa, (**b**) 0.30 MPa, (**c**) 0.50 MPa [85, 101]. (Reprinted from Combustion and Flame, 2019, Vol 204, Okafor, E.C., et al., Measurement and modelling of the laminar burning velocity of methane-ammonia-air flames at high pressures using a reduced reaction mechanism, Pages No.162-175, Copyright (2023), with permission from Elsevier). (Reprinted from Combustion and flame, 2018, Vol 187, Okafor, E.C., et al., Experimental and numerical study of the laminar burning velocity of CH4–NH3–air premixed flames, Pages No.185-198, Copyright (2023), with permission from Elsevier)

methane ammonia flames under higher pressures (up to 3 bar) and compared their experiments to numerical DNS simulations. While the simulations provided better agreement than most reaction mechanisms, both methods were sufficiently accurate in predicting trends. Lubrano Lavadera et al. [115] conducted both experimental and numerical investigations on the addition of CH_4, alongside n-heptane and iso-octan. They found discrepancies between results obtained using a spherical chamber and a heat flux burner, particularly with higher hydrocarbons, higher equivalence ratios, and higher temperatures, where the chamber values exceeded those of the burner by more than 5 cm/s. Their study indicated that the $S_L/S_{L,0}$ ratio followed a linear trend for all NH_3 mass fractions, while the temperature displayed a nonlinear behaviour in the mixtures of methane and NH_3. Furthermore, Liu et al. [116] investigated these trends under oxyfuel conditions, using CO_2 dilution (25–65%) in the oxidiser. They found an increase in burning velocity at a higher methane concentration across all equivalence ratios.

Ammonia mixture with carbon monoxide Han et al. [84] investigated how the addition of carbon monoxide (CO) affects the LBV of ammonia. They found that incorporating CO into ammonia increases its burning velocity across all mixture ratios. The maximum LBV appears to decrease starting from an equivalence ratio (ϕ) of 1 for mixtures containing 80% admixtures, as illustrated in Fig. 18. This phenomenon also includes a shift in the peak LBV: with higher CO admixtures, the point of maximum LBV moves from stoichiometric to richer conditions. This trend aligns with observations made in studies involving hydrogen admixtures.

Ammonia mixture with Dimethyl ether (DME) The widespread adoption of pure ammonia as an energy source faces challenges, including its low reactivity and flame speed, which compromise the combustion efficiency of ammonia-fuelled powertrains. Dimethyl Ether (DME) is gaining attention as a potential combustion enhancer for ammonia, given its carbon−neutral properties and superior combustion performance. Remarkably, DME is cost-effective and can be produced from CO2 and water using renewable energy sources [117]. In their study, Cai and Zhao [104] utilised a 1D freely propagating flame model to evaluate the SL of NH3/DME flames. They found that adding DME significantly boosts the laminar flame speed of ammonia, bringing it to levels comparable to hydrocarbon fuels. This improvement is attributed to both thermal and chemical effects. Additionally, they noted that the impact of pressure on ammonia flame propagation becomes more significant when DME is blended, as it changes the primary termination reactions and their sensitivities.

Similarly, Tao and Dan [103] also investigated the LBV of premixed NH3/DME/air flames, confirming a monotonic relationship between ammonia laminar flame speed on the DME blending ratio. Kinetic and sensitivity

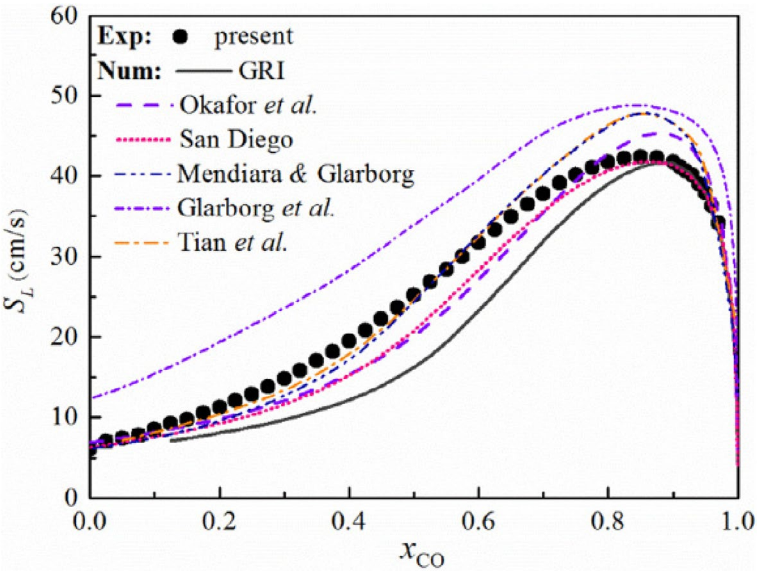

Fig. 18 Laminar burning velocities of stoichiometric NH3 /CO/air flames, compared to simulations (adopted from Han et al. [84]) (Reprinted from Combustion and Flame, 2019, Vol 206, Han, X., et al., Experimental and kinetic modelling study of laminar burning velocities of NH3/air, NH3/H2/air, NH3/CO/air and NH3/CH4/air premixed flames, Pages No.214-226, Copyright (2023), with permission from Elsevier)

analyses indicated that DME substantially modifies the major branching and termination reactions in the flame. Sun et al. [97] conducted a numerical study on the combustion characteristics and chemical kinetic mechanism of NH3/H2/DME ternary fuel, finding that DME enhances ammonia flame stability by generating more CH2O, an active precursor, in the inner flame region. Liu et al. [116] identified two reactions, N + NO = N2 + O and N + OH = NO + H, as crucial in influencing the SL of rich ammonia mixtures under oxy-fuel combustion conditions. Recently, Issayev et al. [64] studied the NH3/DME combustion at different DME blending ratios (e.g., 17.9%, 36.8% and 46.7%) and reported that the DME addition accelerated the flame propagation. Results showed the SL peaked at 26.5 cm/s (see Fig. 19) with introducing 47% of DME at 1 bar and Φ = 1.1. Similarly, Yin et al. [118] further extended the DME ratio to 50% at ambient conditions and reported that the SL was increased remarkably compared to that of pure ammonia.

The addition of oxygenated alternative fuels such as methanol and ethanol is increasingly viewed as a vital strategy to improve the reactivity and combustion efficiency of ammonia. These mixtures are becoming more popular in current combustion equipment due to their enhanced performance. In addition, methanol and ethanol can be produced from biomass, supporting carbon neutrality, and offer the advantage of high energy density storage in liquid form.

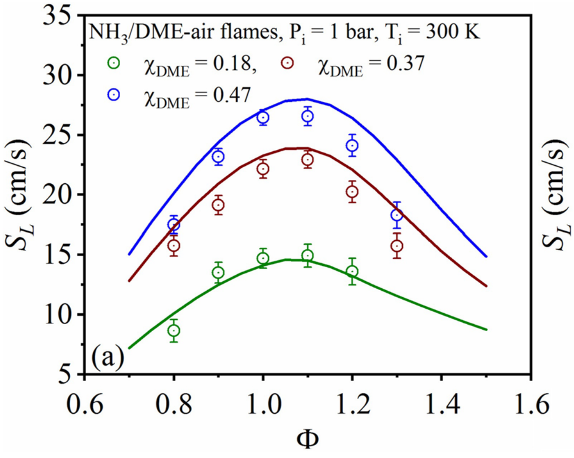

Fig. 19 Comparison of measured laminar burning velocity (open circles) and model predictions for various DME blending ratios (χ_DME) at different equivalence ratios (φ) . [64]. (Reprinted from Renewable Energy, 2022, Vol 181, Issayev, G., et al., Ignition delay time and laminar flame speed measurements of ammonia blended with dimethyl ether: A promising low carbon fuel blend, Pages No. 1353-1370, Copyright (2023), with permission from Elsevier)

Ronan et al. [119] conducted experimental investigation on the influence of ethanol addition, ranging from 0 to 100% in NH3. They used the spherical expanding flame technique under constant pressure conditions, enabling measurements at equivalence ratio from 0.8 to 1.3, at 423 K and 1 bar. Their findings indicated a linear relationship between the LBV and ethanol addition, with a more noticeable increase in LBV under lean conditions compared to rich ones. In a similar work, Wang et al. [120] used an Heat flux burner setup for the investigation of methanol and ethanol addition from 0-100% in NH3. Their experiments spanned four temperatures (298, 348, 398, and 448 K) at 1 bar. The results exhibited a linear trend for methanol at stoichiometric conditions but a nonlinear trend for ethanol, which was consistent with the predictions of five reaction mechanisms, as shown in Fig. 20. They also employed the temperature coefficient to validate the consistency of their experimental data.

2.3.5 The buoyancy effect

Ichikawa et al. [121] provided an example by visualising the spherically propagating of ammonia's laminar flame, as shown in Fig. 21. It is hypothesised that the observed orange chemiluminescence originates from the NH_2 ammonia α band spectra and the spectra of superheated H_2O vapour. This study also highlighted the buoyancy effect of the already burned hot gases, a consequence of ammonia's low burning velocity. Such buoyancy complicates the analysis of spherical flames, making it a challenging aspect to account for. In another research endeavour [82], Schlieren imaging was employed to examine the LBV of ammonia under various pressure conditions, with the results presented in Fig. 22. It can be seen that at higher pressure conditions the circular shape of the flames becomes more compressed. Shu et al. [113] further investigated these buoyancy effects, utilising Schlieren method with varying ignition power to establish the stability criteria for methane/ammonia flames. Their results indicated that the lower flammability limit remained consistent irrespective of pressure. However, the experiments revealed that the upper flammability limit expanded at higher pressures, such as 5 bar.

2.3.6 Turbulent flame speed

Turbulence in combustion is typically characterised by the turbulence intensity and the integral length scale, which signifies the average size of large eddies [122]. In real engine conditions involving turbulent eddies cause the flame surface to wrinkle, thereby expanding the flame area, enhancing local reactant transport, and altering the flame structure [123]. Consequently, turbulent flame speed (TFS) is considerably higher than laminar flame velocity (LFS), primarily due to the effects of

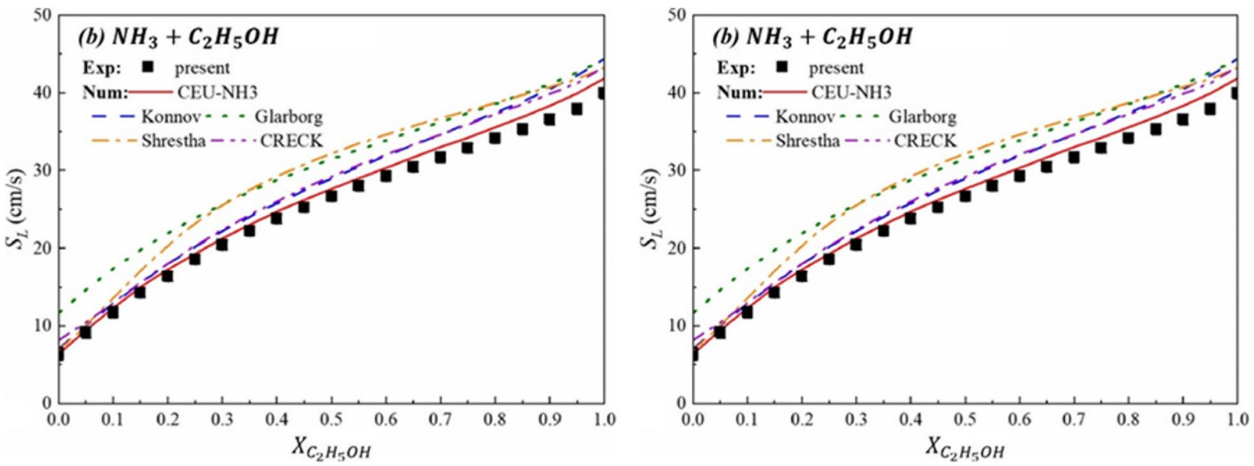

Fig. 20 Laminar burning velocity of stoichiometric of **a**) NH3/CH3OH/air, and **b**) NH3/C2H5OH/air flames, at 298 K and 1 atm [120]. (Reprinted from Combustion and Flame, 2021, Vol 229, Wang, Z., et al., Experimental and kinetic study on the laminar burning velocities of NH3 mixing with CH3OH and C2H5OH in premixed flames, Pages No. 111392, Copyright (2023), with permission from Elsevier)

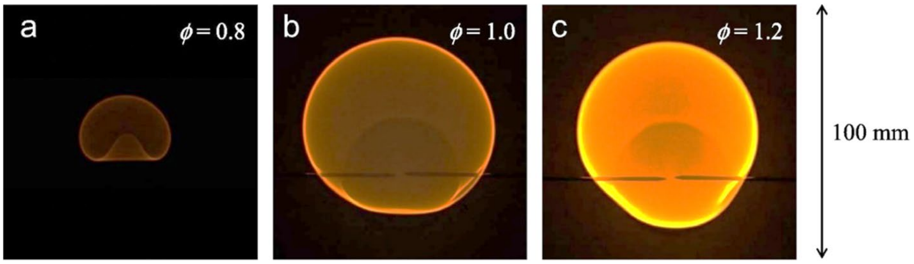

Fig. 21 Direct photographs of ammonia/air premixed flames of Pi = 0.1 MPa at t = 100 ms. (**a**) φ=0.8, (**b**) φ=1.0, (**c**) φ=1.2 [121]. (Reprinted from International Journal of Hydrogen Energy, 2015, Vol 40(30), Ichikawa, A., et al., Laminar burning velocity and Markstein length of ammonia/hydrogen/air premixed flames at elevated pressures, Pages No. 9570-9578, Copyright (2023), with permission from Elsevier)

flame wrinkling and stretching caused by turbulence [32]. TFS, a vital parameter in understanding combustion characteristics, aids in elucidating the interplay between chemical dynamics and turbulence [124] and is essential in designing combustion chamber and validating numerical models. Dating back to Damkohler's pioneering work in 1940 [125], turbulent combustion was initially conceptualised as a planar, quasi-one-dimensional structure propagating in a stable, homogeneous, and isotropic background turbulence [126]. Thus, TFS is defined as the mean velocity at which a premixed flame propagates into a statistically homogeneous mixture with zero average velocity and identical properties [127]. Various experimental studies, such as those by Plessing et al. [128], Kobayashi et al. [129] and Soika et al. [130], have focused on evaluating TFS under different conditions, including changes in pressure and its impact on flame curvature. Bradley et al. [131, 132] also contributed significant TFS data for various air/fuel mixtures in a spherical bomb, establishing that TFS is more influenced by pressure than by the mixture's Markstein length.

Recently, the study of TFS in ammonia combustion has garnered renewed interest, especially due to ammonia's zero carbon emissions. Ammonia presents a unique case for premixed turbulent combustion research because of its lower LBV, thicker flame, and higher Karlovitz number (Ka), making it suitable for non-invasive optical and laser diagnostic techniques. Xia et al. [123] visualised the time-sequential Schlieren images of a fuel-lean NH3/O2/N2 flame ($\phi = 0.6$) from 1 to 6 ms with laminar flow ($u\prime = 0$) and four different turbulence intensities. Fig. 23 illustrates how the flame becomes increasingly wrinkled and accelerates with greater turbulence intensity, with smaller patterns of flame surface deformation becoming more evident. In addition, the study observed that the flame's speed and the inhomogeneity caused by the vortex folding effect are amplified with increased turbulence intensity. The research also measured TFS and LFS across

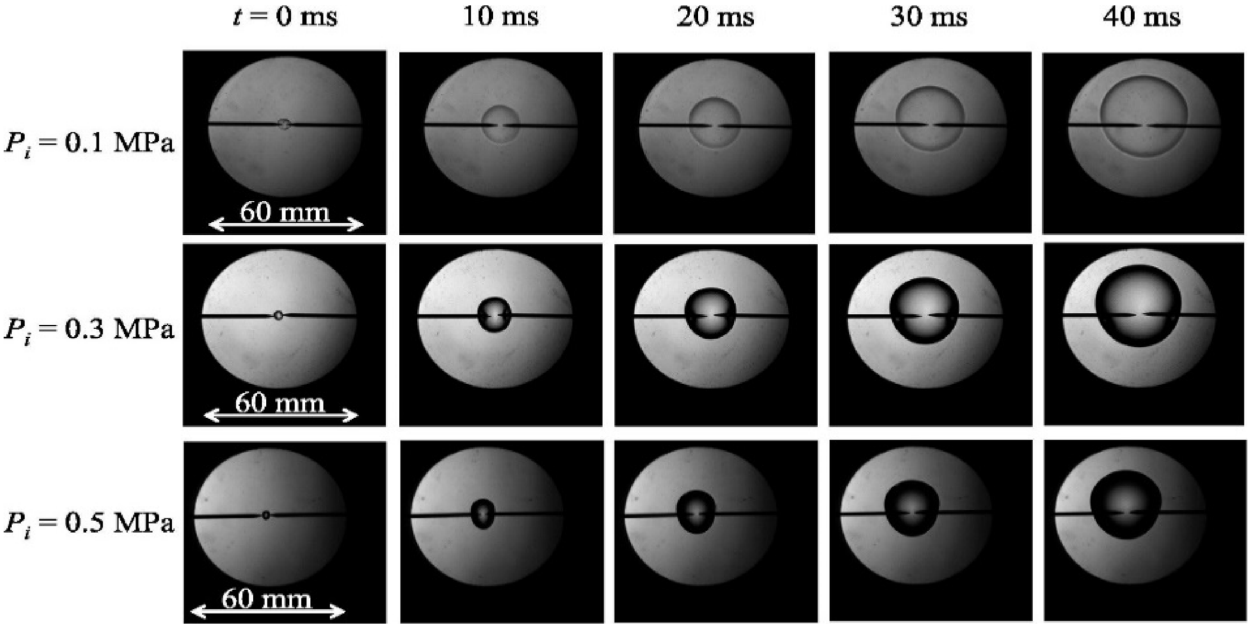

Fig. 22 Schlieren images of ammonia/air premixed flames at different pressure conditions (∅=1) [82]

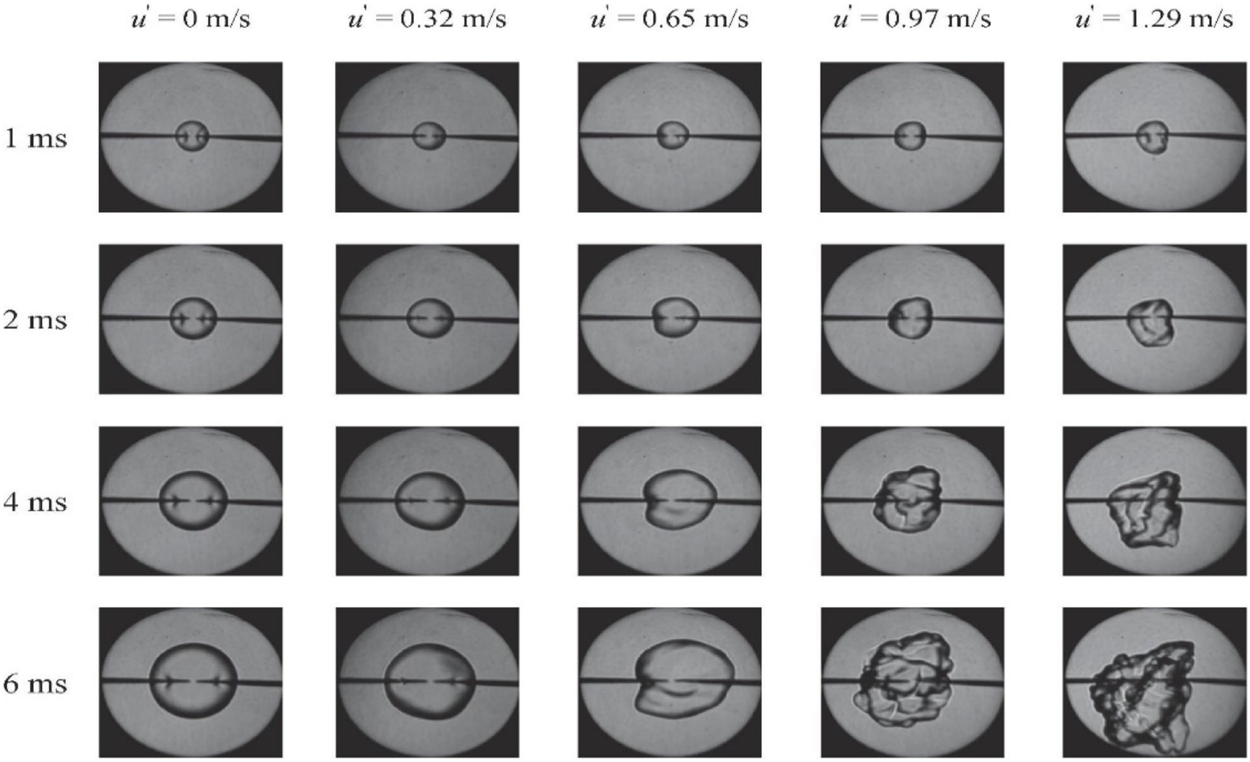

Fig. 23 Schlieren images of NH3/O2/N2 mixtures at various turbulence intensities (∅=0.6) [123]. (Reprinted from Fuel, 2020, Vol 268, Xia, Y., et al., Turbulent burning velocity of ammonia/oxygen/nitrogen premixed flame in O2-enriched air condition, Pages No. 117383, Copyright (2023), with permission from Elsevier)

various equivalence ratios, finding that both peaked around ϕ = 1.0, as shown in Fig. 24.

Hashimoto et al. [133, 134] observed that adding methane to ammonia extends the flammability limit of the ammonia flame. While ammonia's LBV peaks at slightly richer conditions (ϕ ~ 1.1), the diluted ammonia/air mixture is capable of propagating even under high turbulence intensity. This enhanced propagation is attributed to the Lewis number (Le) being less than one under lean conditions, which elevates TFS and broadens the extinction limit. Ichikawa et al. [135] conducted experiments with ammonia/methane/air mixtures in a nozzle burner at 0.5 MPa and 298 K, measuring the TFS across various NH3 mole fractions up to 39%. Dai et al. [122] further evaluated the TFS of stoichiometric ammonia/methane/air mixtures and confirmed the validity of a turbulent combustion model for flames at initial pressures up to 3 bar. Their study also detailed the development process of NH3/CH4/Air flames under different turbulence conditions, highlighting the increase in flame wrinkling and decrease in TFS as the NH3 mole fraction rose from 20 to 60%, primarily due to the influence of flame chemistry as shown in Fig. 25.

Lhuillier et al. [93] compared TFS in three mixtures (NH_3/H_2, NH_3/CH_4, CH_4/H_2) under various engine conditions. They reported that the ratio of turbulent to laminar flame speed increased with the addition of H_2 but decreased with the addition of CH_4. This is explained by the opposite thermal-diffusion-chemical properties of H_2 and CH_4, along with their different responses to flame stretching. Similarly, Ichikawa et al. [135] found that higher NH3 fraction led to lower flame surface density and a reduced speed ratio between turbulent and laminar flames. Wang et al. [124] studied the combustion of stoichiometric NH3/O2/N2 mixtures under oxygen-enriched conditions, revealing that increased O2 fraction enhanced TFS across all pressures and turbulence intensities, while lowering the turbulent/laminar flame speed ratio. TFS was also noted to rise with increasing temperature and turbulence intensity, particularly at high O2 concentrations. In addition, Yang et al. [136] implemented the direct numerical simulation to study the turbulent combustion of NH3 and NH3/H2 mixtures under engine conditions. They concluded that the addition of H2 promoted the reactivity of the mixture and enhanced turbulent combustion. In particular, the flame surface shows more wrinkled structures with lean mixtures than with stoichiometric and rich mixtures. Thus, the highest TFS was found in the lean case because a more furrowed flame surface led to a lower effective Le, which can be explained by the interaction between turbulence and thermal diffusion instability. More experimental and numerical studies of the turbulent combustion of ammonia and its fuel mixtures can be found in refs. [137, 138].

2.4 Extinction processes

While ammonia is recognised as a promising carbon-free fuel that can be produced by renewable energy sources,

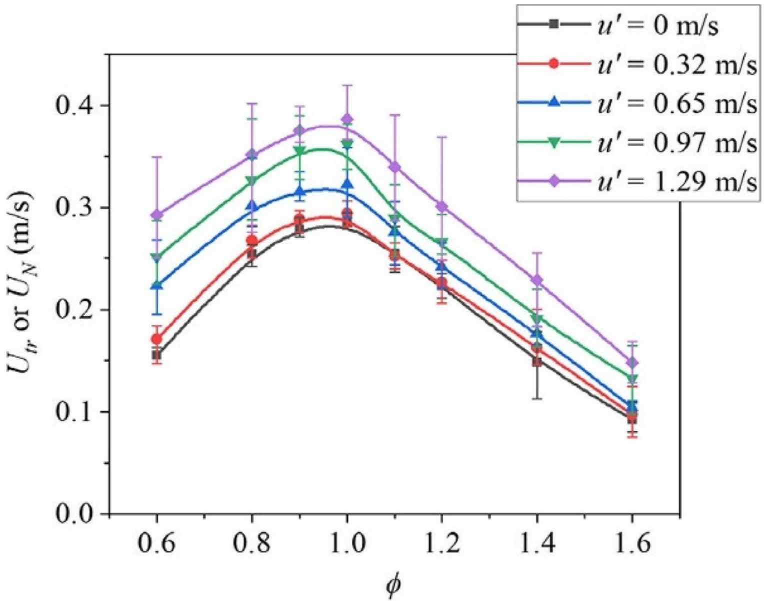

Fig. 24 Turbulent burning velocity (U_N) and stretched laminar burning velocity (U_tr) as a function of equivalence ratio [123]. (Reprinted from Fuel, 2020, Vol 268, Xia, Y., et al., Turbulent burning velocity of ammonia/oxygen/nitrogen premixed flame in O2-enriched air condition, Pages No. 117383, Copyright (2023), with permission from Elsevier)

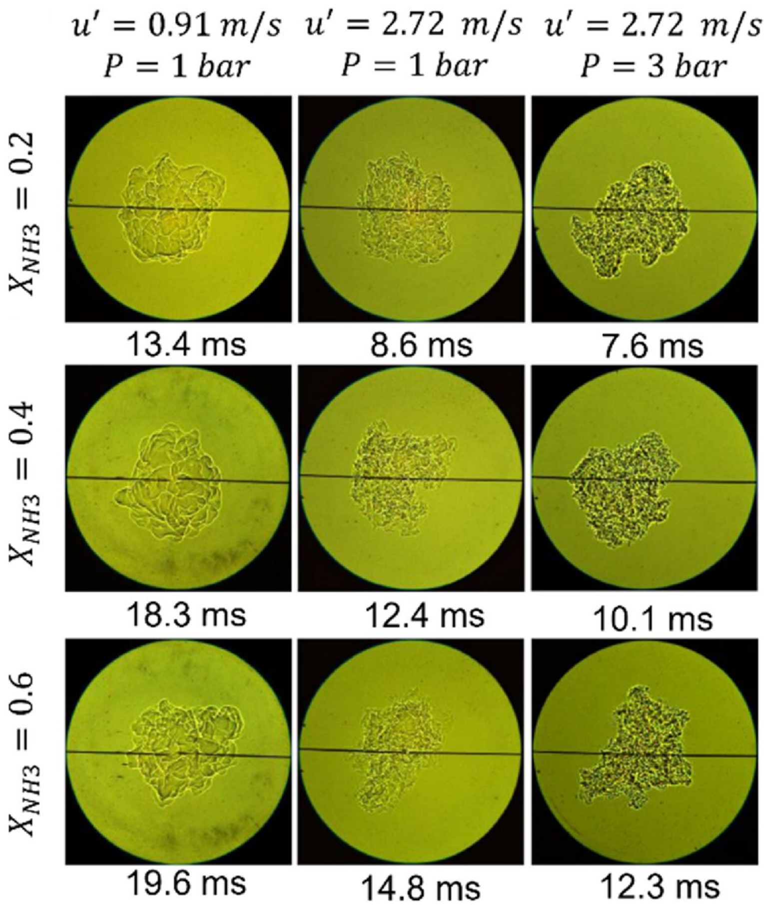

Fig. 25 Flame morphology under different ammonia molar contents, pressures, and turbulence intensities. (adjusted from [122]) (Reprinted from Combustion and Flame, 2022, Vol 242, Dai, H., et al., Measurement and scaling of turbulent burning velocity of ammonia/methane/air propagating spherical flames at elevated pressure, Pages No. 112183, Copyright (2023), with permission from Elsevier)

its inferior combustion properties have hindered widespread adoption in thermal power plants. A notable issue is the tendency of ammonia flames to blow off under turbulent conditions, making the investigation of their extinction limits crucial. An early work implemented by Zakaznov et al. [139] showed that the critical extinction diameter of ammonia flame at stoichiometric ratio and constant pressure conditions varies depending on the nature of flame propagation and equipment variables, such as whether the chamber is open or closed. This was further elucidated by some following studies on ammonia/air flame extinction limits. For example, Ichimura et al. [134] revealed that the fuel-lean flames are more sustainable than fuel-rich ones at high-turbulence conditions. This can be explained by the Lewis number (Le), which is less than unity, and the local burning velocity could be promoted by the diffusional-thermal instability. In contrast, the local burning velocity of fuel-rich NH3/air mixture does not increase with Le > 1, leading to a low extinction limit. Moreover, hydrogen addition to ammonia/air mixtures has been identified as an effective means of enhancing flame stability. Wiseman et al. [140] demonstrated that the NH3/H2/N2/air flames are less tendency towards extinction than CH4/air flames for identical strain rates. Similarly, a recent work conducted by Shen et al. [141] examined the blow-out limits of NH3/H2/N2/air flames in an axisymmetric unconfined bluff-body burner, finding that these flames had higher blow-out velocities than CH4/air flames, with a significant difference in order of magnitude. Their DNS results indicated that the rapid hydrogen diffusion into the preheat layers of the turbulent flame front was critical for preventing the blow-out behaviour.

Furthermore, Colson et al. [142] investigated how pressure influences ammonia flame extinction characteristics, showing that the extinction stretch rate of NH3/air flame increases with pressure and is comparatively higher than that of CH4/air flame. They also noted a more significant reduction in characteristic reaction time with pressure in ammonia/air flames. Xiao et al. [143] investigated stretched NH3/air flames with methane addition, observing improved sustainability at high stretch rates with increased methane blending. For non-premixed ammonia flame, Choi et al. [144] studied the blow-off limits, flame temperature and morphology of the counterflow non-premixed NH3/H2/air flames at normal pressure and elevated temperatures. They concluded that the extinction limits and the maximum flame temperature improve with higher hydrogen blending ratios and air temperatures, while decrease at higher strain rates. Ku et al. [112] measured the extinction limits and structure of the non-premixed NH3/CH4/air flames and demonstrated that the flames could sustain less NH3 at high strain rates. Moreover, Lee and Kwon [145] numerically investigated the non-premixed NH3/H2/air flames at normal pressure conditions, and concluded that the radiation effect did not affect the extinction limits and flame structure remarkably especially at high stretch rates.

2.5 Ammonia emissions

In addition to its low flame speed, limited flammability range, and high ignition energy requirements, the use of ammonia as a fuel is also challenged by its significant NOx emissions. These emissions pose serious risks to both human health and the environment. Another undesirable byproduct of ammonia combustion is N2O, which has a global warming potential 300 times greater than CO2 [146]. Therefore, it is crucial to thoroughly understand the mechanisms behind NOx formation in ammonia flames and develop strategies to reduce these harmful emissions. NOx emissions primarily consist of NO, NO2, and N2O, with NO constituting about 90% of these emissions. Thus, reducing NO is a key objective in achieving low-NOx combustion in ammonia flames. The formation of NOx in ammonia combustion occurs predominantly through two pathways: fuel NOx and thermal NOx. Fuel NOx is generated by the oxidation of nitrogen atoms present in ammonia, while thermal NOx forms when atmospheric nitrogen (N2) is oxidised into NOx at temperatures exceeding 1800 K.

The conversion of fuel-bound nitrogen to NOx emissions is influenced by local fuel concentration and combustion characteristics. In the combustion process, various radicals like NH3, CN, N, HCN, and NH are produced, which either contribute to the formation of fuel NOx or assist in its decomposition. A simple model proposed by some researchers [147] explains this phenomenon through a dual competitive reaction pathway, highlighting the complexities in balancing NOx formation and reduction during ammonia combustion:

$$Fuelnitrogen \rightarrow\ N\,intermediates(NH_3, CN, N, HCN, NH) \rightarrow NO(O_2\ oxidation), or \rightarrow N_2(NO\,reduction)$$

According to the Zeldovich mechanism [148], the primary reactions of thermal NO are listed by (1) – (3). Also, the temperature plays an important role in thermal NO production, since the reaction (1) is rate-limited and mostly occurs at high temperatures (usually above 1800 K) [149].

$$N_2 + O = NO + N \tag{1}$$

$$N + O_2 = NO + O \tag{2}$$

$$N + OH = NO + H \tag{3}$$

Fenimore [150] introduced an additional pathway for NO generation, known as the prompt NOx mechanism, detailed through reactions (4) – (7). This mechanism operates primarily at lower temperatures (1373 K) in fuel-rich zones. Moreover, the CH_i radicals react with N2, forming intermediates such as HCN, NH, CN and N. These intermediates are subsequently oxidised to NO during the combustion process. To mitigate the production of prompt NOx, maintaining the fuel mixture in a lean state and minimising the local formation of additional CH_i species is recommended [147].

$$CH + N_2 = HCN + N \tag{4}$$

$$N + O_2 = NO + O \tag{5}$$

$$HCN + OH = CN + H_2O \tag{6}$$

$$CN + O_2 = NO + CO \tag{7}$$

In NH3/air mixture, the NO concentration gradually increases with the equivalent ratio, peaking at an equivalent ratio of 0.9. Byond this point, as the mixture becomes increasingly fuel-rich, the NO mole fraction sharply decreases [147]. However, this shift towards a fuel-rich combustion leads to a significant increase in unburned NH3, adversely affecting combustion efficiency. Studies [17] have indicated a tradeoff between NO and unburnt NH3 emissions, with the total emissions of NO and NH3 reaches a minimum around the equivalence ratio of 1.1. Thus, opting for rich combustion emerges as a viable strategy to mitigate NOx emissions. Mashruk et al. [151, 152] demonstrated the formation and reburn routes of fuel NOx in 70/30VOL.% NH3/H2 swirling turbulent flames under both fuel-lean ($\Phi=0.65$) and rich ($\Phi=1.20$) conditions. The involved pathways are illustrated in Fig. 26 and Fig. 27, respectively. In these scenarios, NH3 is

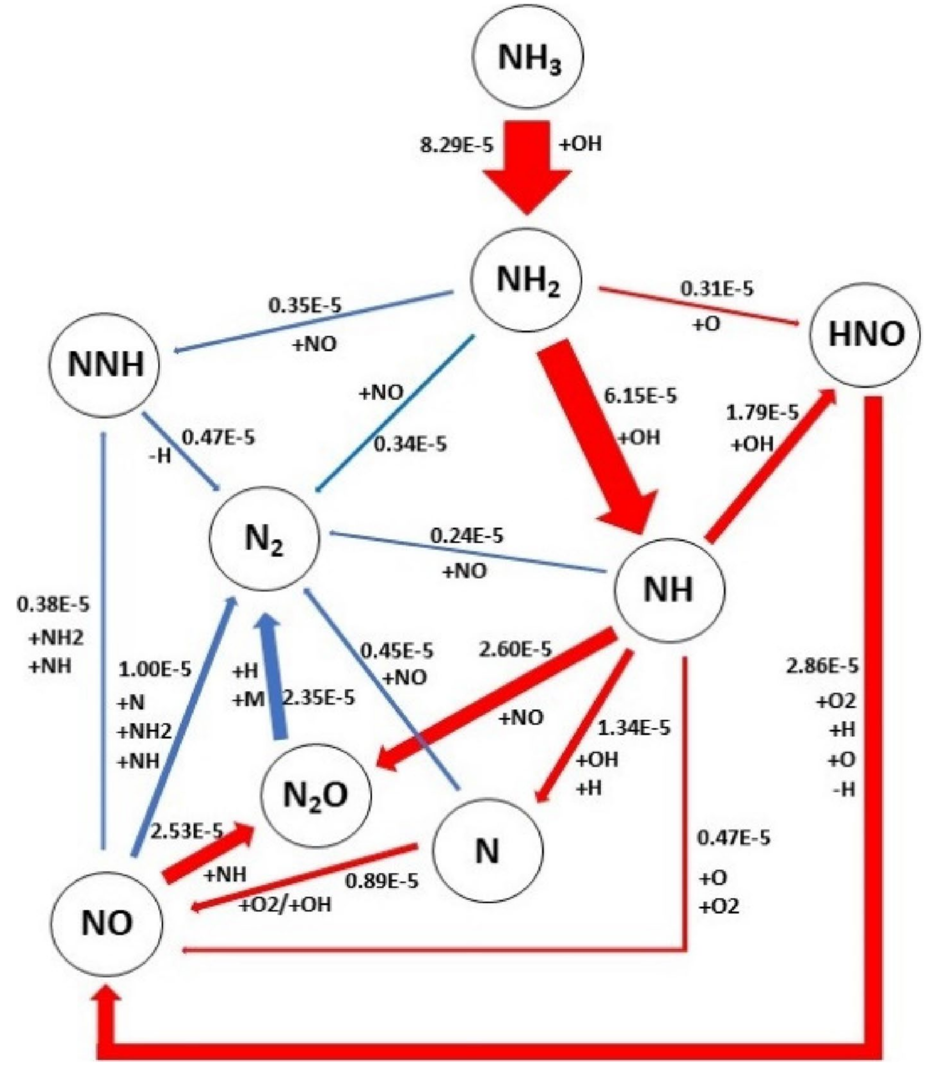

Fig. 26 Fuel NOX formation and reburn pathways at lean condition (Φ = 0.65). Reproduced from[151]

converted into NH2 and NH radicals through reactions with OH, a process common to both lean and rich burns. These radicals then react with O radicals to form Nitroxyl (HNO), a primary source of NO in ammonia flames. In addition, the produced NO reacts with NH radicals, leading to the formation of N2O, which can subsequently be converted to N2 via a third-body reaction: N2O(+ M) ↔ N2 + O(+ M). Other significant pathways for NO reduction include the chain branching reaction NH2 + NO ↔ NNH + OH and the terminating reaction NH2 + NO ↔ N2 + H2O [153, 154].

As illustrated in Fig. 28, the research by Somarathne et al. [137, 138] involved 3D numerical computations on ammonia/air flames. Their results revealed that applying high initial pressure to NH3 flames under rich conditions leads to a significant reduction in fuel NO production. This effect is attributed to the decreased concentration of OH radicals and the enhanced combination of NH_i $(i = 1, 2)$ radicals under these conditions. The same study [138] also demonstrated that high pressure can diminish unburnt NH3 emissions, a result of the shortened characteristic reaction time for ammonia. Chai et al. [147] further support these findings, noting that increased pressure accelerates three-body reactions such as H + OH + M = H2O + M and H + O2 + M = HO2 + M. The enhancement of these reactions under high-pressure conditions leads to more efficient ammonia oxidation pathways, thereby suppressing NOx emissions.

$$NH_3 \overset{+OH}{\rightarrow} NH_2 \overset{+HO_2,NO_2}{\rightarrow} H_2NO \overset{+O_2}{\rightarrow} HNO \overset{+O_2}{\rightarrow} NO \overset{+NH_2}{\rightarrow} N_2$$

Owing to its high ignition energy requirement, low flame speed, and combustion instabilities, ammonia is often blended with other fuels, such as hydrogen

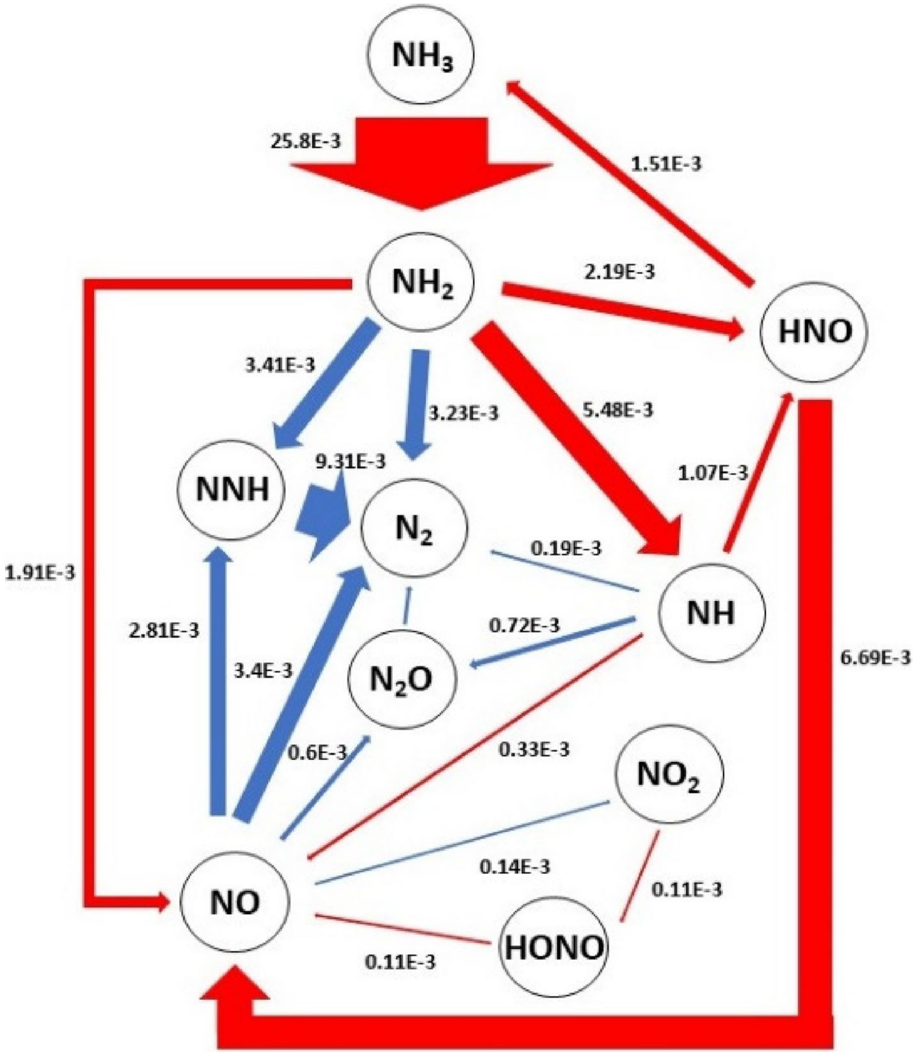

Fig. 27 Fuel NOX formation and reburn pathways at rich condition (Φ = 1.20). Reproduced from [152]. (Reprinted from International Journal of Hydrogen Energy, 2021, Vol 242, Mashruk, S., H. Xiao, and A. Valera-Medina, Rich-Quench-Lean model comparison for the clean use of humidified ammonia/hydrogen combustion systems, Pages No. 4472-4484, Copyright (2023), with permission from Elsevier)

or methane, to serve as an 'ignition promoter', as discussed early. While this strategy effectively improves combustion characteristics, it can exacerbate NOx emissions due to the resultant higher combustion temperatures. Consequently, the NOx emissions resulting from ammonia fuel mixtures have garnered significant attention from researchers. Among various studies, Okafor et al. [101] developed a mechanism to predict the NO_X emissions from NH_3/ CH_4 flames. This mechanism was formulated by adapting the GRI-Mech 3.0 mechanism and incorporating the Tian mechanism [149]. Their research focused on a stoichiometric 70/30VOL.% CH4/NH3 flame, with the reaction pathways detailed in Fig. 29.

Early work by Valera-Medina et al. [13] on premixed $61/39_{VOL.\%}$ NH3/CH4 swirl flames showed a decrease in both NO_X and CO_2 emissions with increasing the equivalence ratio. However, this led to a rise in unburned NH3, hydrocarbon and CO emissions. Likewise, Ramos et al. [155] studied the NH3/CH4 flame with different blending ratios (X_{NH3}=0—0.7) and equivalence ratios (0.8, 0.9 and 1.0). The results showed that the NO_X production was promoted when the NH3 fraction rose from 0 to 0.5, then dropped when the X_{NH3} went up continuously. Besides, adding more ammonia resulted in higher CO concentration at lean conditions. Xiao et al. [143] concluded that NH2 + NO = N2 + H2O and NH + NO = N2O + H

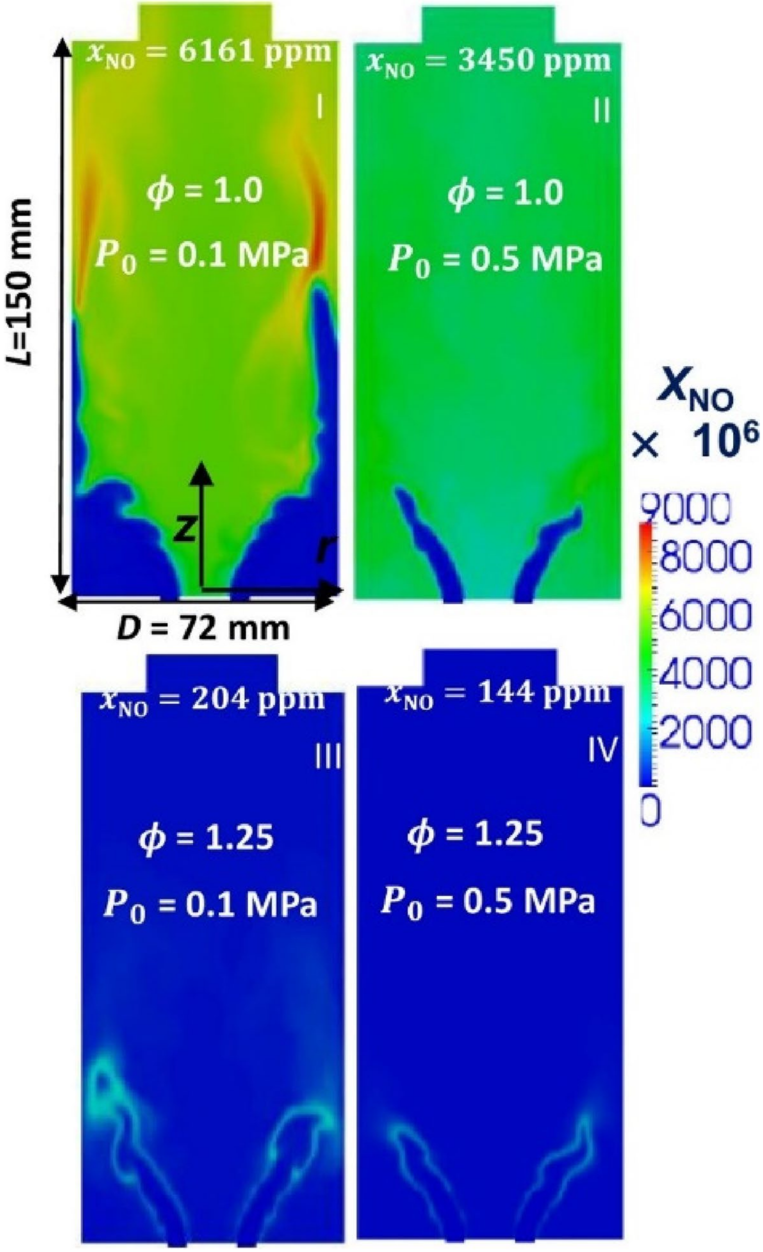

Fig. 28 NO distributions of turbulent premixed ammonia flames regarding different equivalence ratios and initial pressure (T0=500 K,U_in=39.1 m/s, swirl number=0.68). Adjusted from [17]

were two main pathways to decrease NO production for CH4/NH3 flame. With the addition of methane, HCO = H + CO and CH2OH + H = CH3 + OH exhibited higher sensitivities, indicating a more prominent role in hydrocarbon species and reactions. The NO generation is insensitive to hydrocarbon reactions with rich burning, during which NH2 + NO = NNH + OH becomes more effective for NO suppression. Woo et al. [156] also analysed the NOx generation of NH3/CH4 flame at different conditions, they concluded that the NOx emissions increased monotonously with adding more ammonia, and the maximum NOx concentration was obtained at an oxygen ratio of 0.7. Moreover, studies [99, 121] have demonstrated that hydrogen is superior to hydrocarbon fuels in promoting ammonia combustion, since it has a much higher burning velocity and lower ignition energy. In particular, there is no pollutant produced by itself. However, in some cases, the

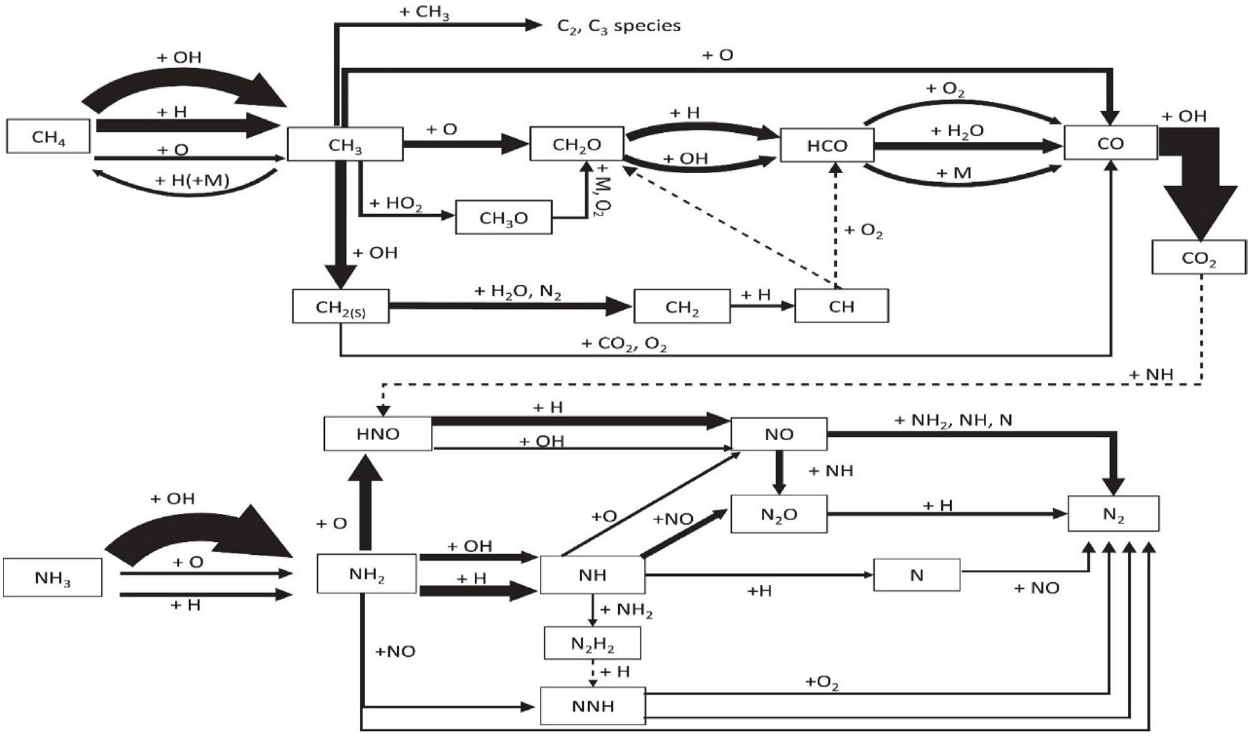

Fig. 29 Reaction pathways of stoichiometric 70/30VOL.% CH4/NH3 flame. Reproduced from [101]. (Reprinted from Combustion and flame, 2018, Vol 187, Okafor, E.C., et al., Experimental and numerical study of the laminar burning velocity of CH4–NH3–air premixed flames, Pages No.185-198, Copyright (2023), with permission from Elsevier)

hydrogen may promote NOx emissions when mixed with ammonia because of its high flame temperature and rich radical pool of O/H. As shown in Fig. 30, the NOx emission increases gradually with the hydrogen fraction for the stoichiometric NH3/H2 flame, owing to more generated OH and O radicals being generated at high temperatures. The NOx concentration peaks when the molar fraction of H2 reaches around 0.8, then it decreases fast by continuously adding more H2, as less fuel NOx is produced because of lower NH3 content in the mixture [157]. Lee et al. [105] illustrated that the ammonia substitution with hydrogen contributed to much lower NOx and N2O emissions under fuel-rich than lean conditions.

The incorporation of hydrogen into NH3/air flames significantly boosts combustion, primarily through key reactions like O + H2 = OH + H, H + O2 = OH + O, and H2 + OH = H2O + H [147]. Table 2 provides a summary of the principal reaction pathways for both NH3/air flames and NH3/H2/air flames. Research by Joo et al. [158] emphasised that hydrogen addition is particularly effective when its mole fraction is below 0.75. As the equivalence ratio increases, the significance of the H2 + OH = H2O + H reaction diminishes, whereas the NH3 + OH = NH2 + H2O reaction becomes more prominent. This shift favours the thermal deNOx process, especially under fuel-rich conditions, contributing to more efficient NOx reduction.

Recent research has focused on deNOx strategies for NH3/H2/air flames in lean combustion scenarios. Valera-Medina et al. [16] conducted a study on lean premixed NH3/H2 flames with a 50/50VOL.% blending ratio. They observed that NO emissions could be reduced to as low as ~ 100 ppm at an equivalence ratio (ϕ) of 0.4. Building upon this, Mashruk et al. [159] examined NH3/H2/air flames across a broad range of equivalence ratios (0.55 – 1.40) under atmospheric conditions. Their findings suggested an optimal equivalence ratio of 1.20 for a two-stage burner configuration, which balances minimal emissions with high combustion efficiency.

3 Applications

This section addresses various technologies employed for ammonia combustion, including both those in the development phase and those that have been established through extensive research and development programmes. Despite the advancements, established methods or technologies still face obstacles, including issues like ammonia slip, carbon monoxide formation (especially when blended with fossil fuels), and

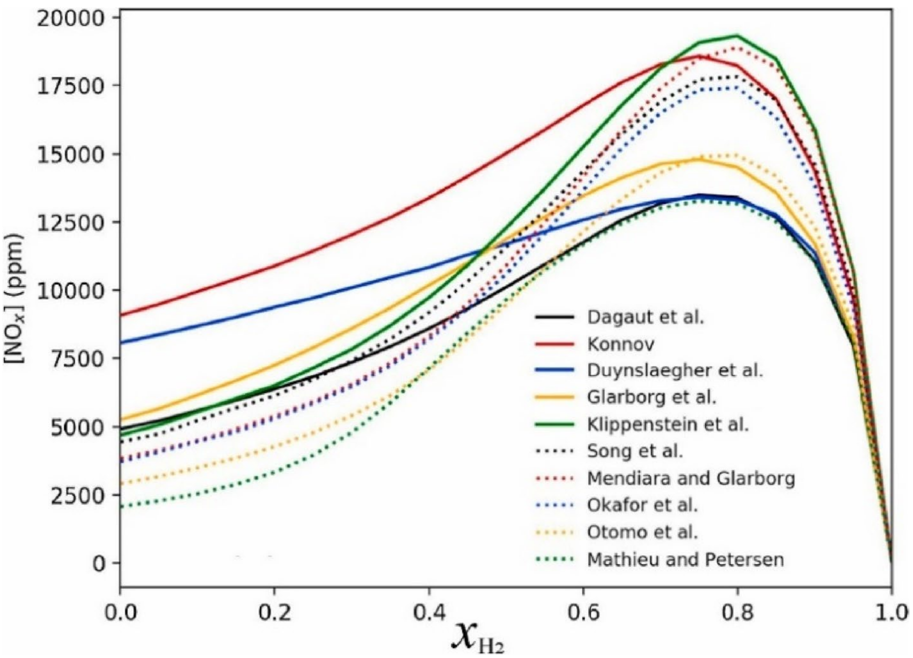

Fig. 30 NOx emissions of NH3/H2 flame as a function of H2 mole fraction (∅=1,P_0=1 bar,T_0=298 K). Reproduced from [157]. (Reprinted from Fuel, 2019, Vol 246, da Rocha, R.C., M. Costa, and X.-S. Bai, Chemical kinetic modelling of ammonia/hydrogen/air ignition, premixed flame propagation and NO emission, Pages No. 24-33, Copyright (2023), with permission from Elsevier).

Table 2 Main reactions of NH3/air and NH3/H2/air flames [157]

NH3/air	NH3/H2/air
$H + O_2 = O + OH$	$H + O_2 = O + OH$
$H + NO + M = HNO + M$	$NH_2 + NO = NNH + OH$
$NH_2 + NO = NNH + OH$	

maintaining flame stability. The discussion extends to the application of ammonia in boilers/furnaces, internal combustion engines, and gas turbines. It also offers deeper insights into the challenges associated with delivering and injecting ammonia into these combustion systems.

3.1 Boiler and furnaces

The basic principle of furnaces is to convert the chemical energy contained in fuels into heat energy. This heat is then utilised for various purposes, such as warming environments, evaporation liquids, or production of superheated conditions within these systems. Boilers, a specific type or function of furnaces, primarily focus on heating fluids (like water). They are designed to provide space heating or generate steam at specified pressures, temperatures, and flow rates. This steam is then employed for heating applications or power generation. Although these systems generally operate close to stoichiometric (ideal) conditions, their combustion principles are largely similar. The distinctions between furnaces and boilers are relatively minor and are outlined in Table 3.

Furnaces and boilers boast several key advantages: they are robust and relatively easy to operate, typically offering longer lifespans, as detailed in Table 4. In terms of thermal efficiency, they outperform many other thermal energy production systems, such as thermal power plants. While furnaces and boilers are generally larger in size, necessitating considerable space and energy inputs, their long-term utility, particularly in the context of using alternative fuels like ammonia, is expected to sustain their relevance and application for an extended period.

Ammonia has been produced for over a century, serving various roles from being a key ingredient in fertilisers and refrigeration to acting as an alternative fuel source. While ammonia can be combusted in furnaces as part of a fuel mixture, furnace designs need to adapt to use ammonia as the primary fuel. Such systems must be carefully adjusted to the specific fuel type, its composition, and combustion characteristics to ensure optimal performance during operation [160]. As a result, there is an inevitable yet ongoing need for developing new furnace designs specifically for ammonia fuels, as indicated in references [11–13, 16]. Progress in this area is already evident at the industrial level, with companies like IHI and Duiker leading the way. For instance, Duiker's Stoichiometric Controlled Oxidation (SCO) technology represents a significant step in ammonia combustion,

Table 3 The comparison between furnaces and boilers

Difference	Furnaces	Boilers
Use	• Industrial applications– to produce and withstand higher temperatures and flowrates for processes, energy and materials production (i.e., melting of raw materials, destruction of waste products, coke oven gas, superheated steam) • Domestic applications– to coke food and/or provide hot air to interior spaces	• Industrial applications– to generate superheated steam at a specific pressure, temperature and mass flowrate for Stirling engines or Steam turbines to generate electric power or for chemical processes • Domestic applications– to provide boiling water, hot water and/or space heating for buildings
Examples	• Central heating systems in buildings • Electric resistance heaters/heat blowers • Fireplace and kitchen stove	• Heat recovery steam generators in power plants • Residential boilers • Kettle heater
Type of fuel	• Natural gas, liquefied petroleum gas, oil, wood, coal or electricity	• Waste heat from thermal cycles, coal, natural gas or electricity

Table 4 Energy production systems applicable to ammonia [4]. (SOFC: solid oxide fuel cell)

Technology (Stationary)	Efficiency (%)	Capital cost (£/kW)	Advantages
Internal combustion engines	30–40%	1000	Robust technology High power density Established technology with fuels blends
Fuel cells	40–65%	760—1300	Suitable for mobile applications Decomposes ammonia in-situ Highly tolerant of ammonia (SOFC)
Combined cycles gas turbines	55–60%	750	High power outputs (> 1MW) High power availability during peak consumption Full-cycle development (combined heat and power)
Furnaces and boilers	85–90%	150—350	Established technology Very robust technology Long life spans High power outputs (> 1MW)

applicable in commercial-size furnaces for both revamps and new designs tailored for ammonia firing. Similarly, various Japanese programmes have made considerable strides, with kilns and co-firing furnaces demonstrating ammonia's versatility as a fuel. An overview of these research and development efforts, along with commercial advancements, is presented in Fig. 31.

In smaller-scale furnace units, innovative technologies like Moderate or Intense Low-oxygen Dilution (MILD) combustion are being explored for efficient ammonia combustion. Characterised by high reactant temperatures and minimal combustion temperature increases, MILD combustion significantly reduces nitrogen oxide (NOx) thermal production [5, 161]. Ammonia's MILD combustion has been stabilised across a broad range of equivalence ratios, demonstrating markedly lower NOx emissions compared to standard combustion methods [12], as shown in Fig. 32. Further information and insights into MILD combustion are available in references [161–165]. In addition, strategies such as water injection into the oxidant stream under stoichiometric-fed (or appropriate) conditions can further diminish NOx emissions [163], potentially enhancing the utility of furnaces for small to medium-scale power applications using ammonia [5].

Integrating ammonia-fuelled furnaces and boilers with Rankine or combined power cycles is another promising approach. Industrial utilisation, particularly in oil and gas facilities, has shown that ammonia combustion can effectively produce heat or superheated steam for power generation. A research study conducted at Cardiff University [27] demonstrated the viability of ammonia furnaces and boilers for the production of sufficient steam/heat for these power cycles. Further studies implemented under industrial conditions showed that co-firing ammonia with other fuel blends (i.e., coal, methane) can reduce CO_2 emissions or could mitigate NO_x emissions in the case of oxygen-enriched combustion, as discussed in refs [11, 166–168]. Ongoing research investigations are aiming to demonstrate the techno-economic feasibility of furnaces/boilers fuelled with ammonia-based fuels. The National Institute of Advanced Industrial Science and Technology (AIST) in Japan has shown promising results with direct liquid injection in combustion systems [12], a technique initially intended for micro gas turbines but also applicable to furnaces and boilers, suggesting the

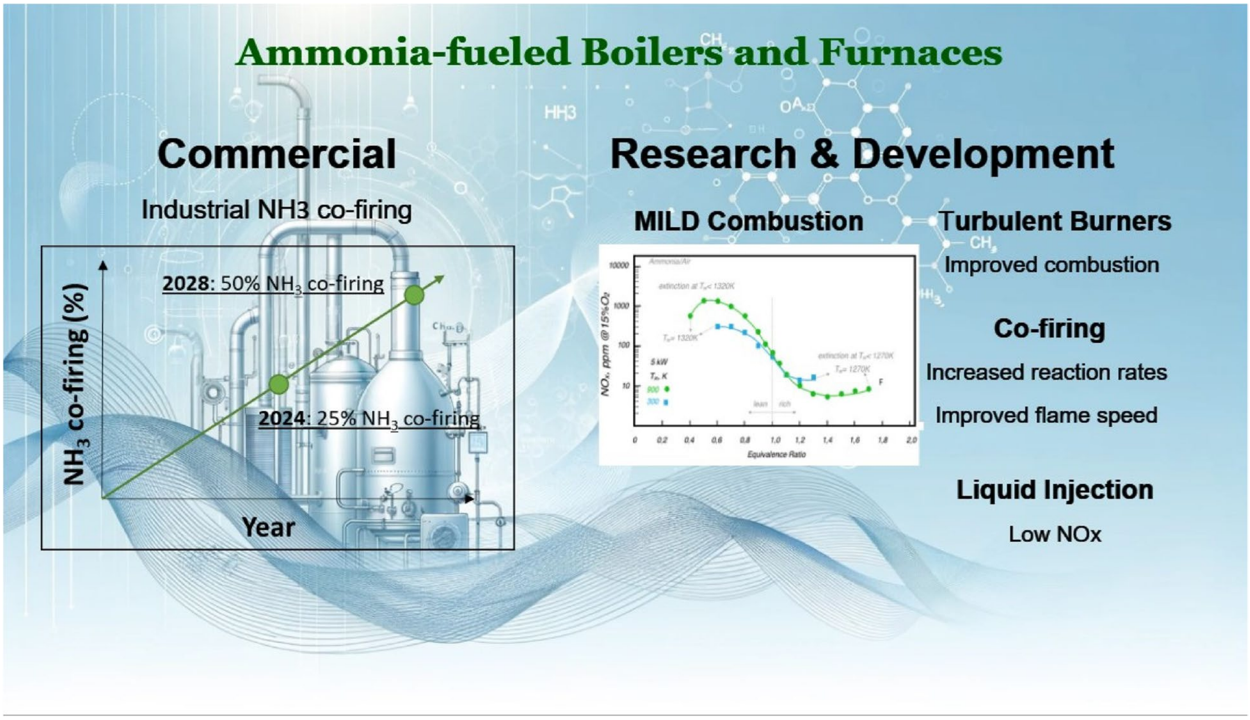

Fig. 31 Commercialisation and Research & Development of ammonia-fuelled boilers and furnaces

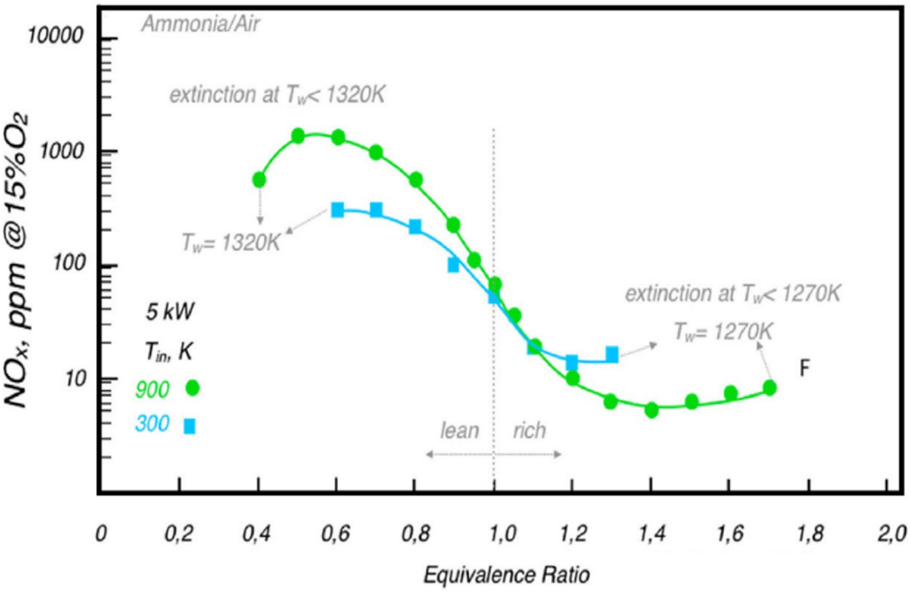

Fig. 32 NOx emissions from ammonia MILD combustion [166]. Equivalence ratio is Air-Fuel ratio

feasibility of 100% ammonia diffusion burners in these systems.

However, a challenge in many furnace and boiler systems is the lack of highly pressurised chambers. High pressure has been found to mitigate some unwanted emissions from the combustion of ammonia blends [11], and it is hypothesised that increased pressure could further reduce thermal and prompt nitrogen oxides [169]. To address this, innovations like Stoichiometric Controlled Oxidation (SCO) technology have been proposed, which do not require high pressures and leverage multi-stage combustion to achieve desirable NOx emissions. The combustion of ammonia poses challenges due to its lower laminar burning velocity compared to fuels like hydrogen or methane, as discussed early. Blending ammonia is particularly of interest for large-scale applications, but it is important to note that high hydrogen content can increase NO emissions and other pollutants like N2O, NO2, unburned ammonia, and potentially cyanide, formaldehyde, carbon monoxide, and sulphur oxides when mixed with fossil fuels [11]. Thus, robust exhaust gas analysers are essential for monitoring these emissions.

Outstanding industrial efforts include JERA and IHI's collaboration on ammonia-coal co-firing at the Hekinan power plant, aiming for a 50% ammonia co-firing rate by 2028 [170]. Similarly, SCO Duiker's technology has successfully combusted ammonia-rich streams in commercial applications. Mitsubishi Power is developing a 100% ammonia-fed boiler for industrial heat applications, with verification planned at JERA's coal power stations in 2028 [170]. These developments suggest a competitive approach in ammonia combustion technology R&D. Finally, current flue gas monitoring based on mg NOx per Nm3 may need reconsideration due to increased water content and reduced CO2 content in ammonia combustion. This could lead to regulatory challenges unless addressed properly. Industries have called for practical solutions, such as recalculating emissions based on energy content (mg NOx per GJ) or shifting to absolute emission metrics. The use of catalytic converters at exhausts, although not covered in this work, could also be an effective measure to control NOx emissions.

3.2 Internal combustion engines

Internal combustion (IC) engines are categorised broadly into two types based on their fuel ignition techniques: compression ignition (CI) and spark ignition (SI) engines. CI engines, typically utilising highly reactive fuels like diesel, achieve autoignition in lean fuel mixtures at thermodynamic conditions near the peak of the compression stroke. In contrast, SI engines ignite and propagate flames in less reactive fuels, such as natural gas or gasoline/petrol, premixed with air in near-stoichiometric amounts. Research and development efforts, along with the commercialisation of ammonia-fuelled IC engines, are illustrated in Fig. 33.

Ammonia, with its lower reactivity indicated by a high autoignition temperature of 651°C (compared to diesel's 210°C) and a high-octane number of 130 (versus diesel's 15–25), poses challenges for combustion in CI engines. The octane number represents a fuel's resistance to detonation under compression in an engine. Early attempts at ammonia CI, such as those by Pearsall and Garabedian

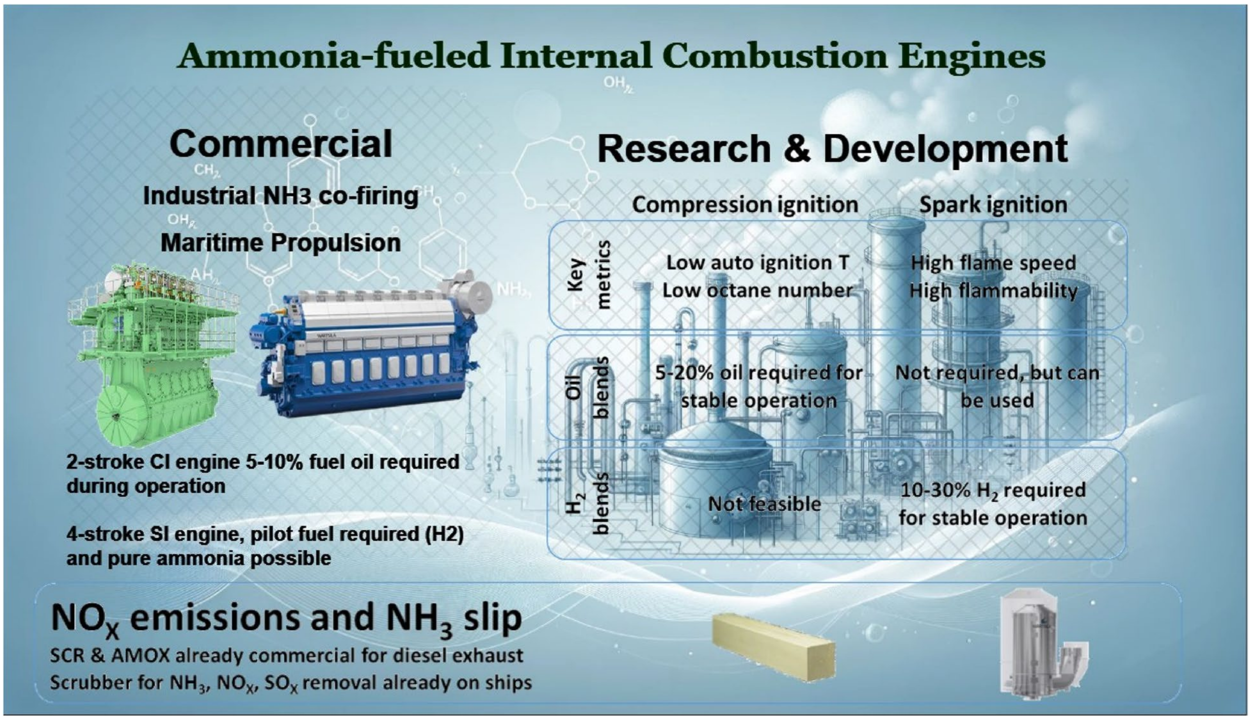

Fig. 33 Research and development into Ammonia-fuelled internal combustion engines

in 1967 [171], faced partial combustion and subsequent failures due to refrigeration effects from liquid ammonia injection, even at high compression ratios of 30:1. Similar difficulties were encountered by Gray et al. [172], who managed only marginal engine operation with dual fueling, intake air heating, and a compression ratio of 35:1. Successful ammonia combustion in standard CI engines is often achieved through dual-fuel operation. This involves injecting warm ammonia vapour into the intake air and igniting the mixture with a small amount of diesel [173–178]. Direct injection and ignition of diesel after compression help ignite the ammonia, allowing both fuels to burn simultaneously. Previous studies on dual-fuel setups using diesel and ammonia have demonstrated that ammonia can replace up to 95% of diesel for stable operation and up to 80% while maintaining efficient engine performance.

In the maritime industry, companies like MAN Energy and Wärtsilä are developing engines capable of running on ammonia, with commercialisation expected as early as 2023–2024 [179]. These marine engines, including two-stroke and four-stroke variants, require some fuel oil for ignition and stability. The fuel oil fraction typically ranges between 5 and 20% by energy content for steady operation in CI mode, similar to liquefied natural gas (LNG) two-stroke engines that use a mix of natural gas and fuel oil [180]. However, the goal is to develop engines that run solely on ammonia, particularly feasible in four-stroke SI engines [181]. In SI engines, ammonia's lower laminar flame speed compared to hydrocarbon fuels and hydrogen necessitates additional turbulence or fuel enhancement for stable operation. Studies have shown that SI engines can run on pure ammonia, but the addition of hydrogen or hydrocarbons improves emissions and efficiency. Mixed effects on stability and emissions have been observed, with pioneering work by Grannell et al. [182] on ammonia-gasoline blends in SI engines.

3.2.1 Hydrogen blending

Ammonia's high molecular stability, compared to hydrocarbons, results in a reluctance to react and a slow flame propagation speed once ignited [82]. An effective strategy to facilitate rapid and efficient ammonia combustion in IC engines is the incorporation of secondary fuels with more reactive combustion properties. Hydrogen, with its extremely high laminar flame speed (SL) and high adiabatic flame temperature, serves as an ideal promoter. It can be directly produced from ammonia through catalytic decomposition and, importantly, does not contribute to CO2 emissions in the exhaust. Even at low substitution rates, hydrogen has been shown to significantly enhance ammonia's combustion properties in spark ignition (SI) engines [17, 83, 183–187]. Adding as little as 30% hydrogen to ammonia by volume can elevate the SL of ammonia to levels comparable to gasoline [17]. This blend not only raises combustion temperatures and efficiency but also reduces unburned fuel emissions. While engines can function on pure ammonia, even modest hydrogen blending markedly improves combustion and emissions. However, higher hydrogen concentrations can lead to increased NOx emissions due to elevated combustion temperatures. Research by Gill et al. [173] involving a compression ignition (CI) engine tested with pure ammonia, hydrogen, and intermediate mixtures showed varying impacts on efficiency. Pure ammonia led to the least reduction in brake thermal efficiency, while pure hydrogen and blends had a more significant efficiency penalty, understandable given hydrogen's high-octane number (well over 130) and an autoignition temperature of 585°C. Pochet et al. [187] experimented with a homogeneous charge compression (HCCI) engine, using ammonia and hydrogen blends, ranging from pure hydrogen to 69% ammonia by volume (74% by lower heating value). At a compression ratio (CR) of 16:1 and without high-reactivity fuel like diesel for ignition, intake air heating was necessary for autoignition, requiring more heat as ammonia content increased. Mørch et al. [188]found that blending 10% hydrogen with ammonia in a spark ignition (SI) cooperative fuel research (CFR) engine offered the best balance of efficiency and power. In contrast, Lhuillier et al. [189] reported that a 5% H2 and 95% ammonia mixture was insufficient for stable operation in spark ignition engines, with optimal indicated efficiency near a 10% H2 blend.

3.2.2 Unburned ammonia emissions

When ammonia is used as a direct substitute for conventional fuels in internal combustion engines, its inferior flame properties can lead to significant emissions of unburned ammonia. This is concerning not only because of ammonia's toxicity but also due to its detrimental environmental effects [190, 191]. In the atmosphere, ammonia undergoes photocatalytic reactions, transforming into NOx and contributing to smog formation. In addition, recent findings suggest that atmospheric ammonia can create secondary organic aerosols and particulate matter through mechanisms akin to those of NOx [190–193]. Generally, engines running solely on ammonia, without hydrogen or hydrocarbon blending, exhibit the highest levels of unburned ammonia emissions due to lower flame temperatures. However, engine performance and ammonia emissions can be improved by adjusting engine calibration, such as advancing fuel injection and ignition timing [194]. Studies indicate that adding hydrogen to the fuel mixture significantly reduces ammonia emissions, with measured concentrations ranging from 0.8 to

1.75% by volume. Under rich fuel conditions, hydrogen and ammonia exhibit similar levels of unburned exhaust concentrations, varying between 0.5 to 2% by volume. A subsequent study by Mounaïm-Rousselle et al. [195] on a spark-assisted compression ignition (CI) engine reported comparable emission profiles, even under high compression ratios up to 17:1, but without hydrogen supplementation. Considering ammonia's extremely high-octane number, operating CI engines at very high compression ratios using ammonia or ammonia-hydrogen mixtures could be more beneficial, potentially leading to more complete combustion and reduced emissions.

3.2.3 NOx and N2O emissions

Nitrogen oxides (NO_X) emissions from ammonia combustion in engines are derived from conventional thermal mechanisms found in hydrocarbon-air combustion, and directly from the ammonia fuel itself. The thermal or Zeldovich Mechanism generates NO and NO_2 emissions from the air participating in the combustion process at temperatures greater than 1800 K. At these elevated temperatures, vibrational excitations in N_2 molecules facilitate the thermal formation of NO, which is subsequently converted to NO_2 due a thermodynamic equilibrium favouring NO_2 formation over NO at elevated temperatures [196]. Fuel-borne reactions rely on concentrations of atomic nitrogen from ammonia decomposition in the reaction zone.

In engines using high ammonia fuel fractions, both in compression ignition (CI) and spark ignition (SI) modes, combustion temperatures are typically lower, thus reducing thermal NOx formation. For instance, maintaining a low hydrogen content in SI engines can keep peak combustion temperatures in check, subsequently curbing thermal NOx production [188]. With a 10% H_2 blend in ammonia, NO_X emissions are reportedly comparable to those from gasoline combustion. Similarly, CI engines fueled 100% with ammonia in dual-fuel configurations exhibit lower NOx emissions than those using diesel alone or hydrogen-ammonia fumigation [173]. However, pure ammonia fumigation is known to increase nitrous oxide (N_2O) emissions compared to hydrogen-ammonia mixtures.

N2O and NOx can also form directly from ammonia fuel via non-thermal processes. Although higher ammonia fractions lead to decreased thermal NOx, they often elevate the direct formation of NO and N2O. A study by Pochet et al. [197] showed a temperature-dependent balance between NO and N2O, where conditions eliminating one species might enhance the other. Intriguingly, fuel-rich combustion in ammonia-partly fueled engines tends to decrease both NOx and N2O emissions. For example, fuel-lean SI operation results in significantly higher N2O emissions compared to a fuel-rich mode [198]. In fuel-rich scenarios, unburned hydrogen from partial ammonia combustion is common. Hayakawa et al. [199] identified a crossover air–fuel ratio around 1.05, where unburned ammonia, hydrogen, and residual NO are minimised. Operating richer in fuel leads to excess NH3 and H2 in the exhaust, while stoichiometric or leaner conditions tend to produce NO emissions exceeding 1000 ppm. This trend in fuel-rich operation is particularly notable with ammonia, as unlike hydrocarbon fuels, soot particle formation is not a concern. These findings suggest that high NOx emissions are not an inevitable outcome of ammonia use in engines. Further research is essential to understand the interplay between thermal and fuel-borne NOx and N2O formation across various fuel stoichiometries and mixtures in both CI and SI engines.

3.2.4 Emissions mitigation strategies

While blending fuels or employing in-situ ammonia cracking to generate hydrogen can reduce emissions, ammonia combustion in dual-fuel modes with hydrocarbons may increase NOx, N2O, hydrocarbon, and particulate emissions. Nevertheless, exhaust after-treatment, a common feature in most compression ignition (CI) engines, offers numerous options for clean ammonia CI engine operation. These methods include exhaust gas recirculation (EGR) for in-cylinder NOx management, wet scrubbers, and ammonia oxidation (AMOX) catalysts for unburned ammonia mitigation, and selective catalytic reduction (SCR) for NOx conversion. Exhaust gas recirculation (EGR) effectively lowers combustion temperatures and thereby reduces thermal NOx. This method is particularly effective in ammonia-fueled engines, as demonstrated by Pochet et al. [187]. In CI engines without hydrocarbon blending, EGR does not present the usual soot-NOx tradeoff due to the absence of carbon. However, excessive use of EGR can reduce the reactivity of ammonia fuel, leading to combustion stability issues.

Ammonia, being basic and water-soluble, can be effectively removed from exhaust gases using wet scrubbers, which also capture particulates and reactive acid gases like NO2 or SO2 [200–202]. If ammonia concentration in the exhaust is low, selective catalytic reduction (SCR) and AMOX catalysts can oxidise it to N2 via NOx intermediates. Unburned ammonia emissions can be advantageous in SCR systems, eliminating the need for additional ammonia or urea injection as in modern diesel engines. The ammonia to NOx ratio (ANR) in the exhaust is crucial for SCR efficiency, with a stoichiometric ANR of 1.0 ensuring complete NOx reduction and no ammonia slip. Catalytic ammonia oxidation (AMOX) and selective catalytic reduction (SCR) technologies, already matured in diesel systems, require minimal adjustments for ammonia engine applications, depending on stoichiometry and hydrocarbon fuel blending. The presence of ammonia slip at the exhaust increases with the increase in ammonia content in the fuel mix, as the reaction becomes colder and slower, as previously mentioned. Therefore, the trade-off between slip ammonia and NOx is still a topic of active research.

N2O emissions pose a unique after-treatment challenge due to their inert nature. Although not directly harmful as a pollutant, N2O is a potent greenhouse gas, undermining the primary goal of ammonia combustion for greenhouse gas reduction. N2O can be reduced through ammonia or metal-based zeolite catalysts in SCR processes [203, 204]. However, N2O production in the SCR of NOx with ammonia [205] presents a paradox in controlling this greenhouse gas. Pathways for both NO and NO2 to form N2O vary depending on temperature and catalyst formulation [206, 207]. Interestingly, N2O emission mitigation technologies, like those developed for European nitric acid plants [208], have shown significant reductions and may offer viable solutions for engine exhaust treatment.

3.3 Gas turbine engines

Gas turbines, alongside steam and hydropower turbines, constitute one of the three primary classes of turbines. Their operation is governed by the Brayton cycle, a single-phase thermodynamic cycle involving a gaseous working fluid, also referred to as the simple cycle. This cycle underpins the working principles of gas turbines. There are two notable variants of the gas turbine Brayton cycle: (1) the open cycle, which employs an internal-combustion process, and (2) the closed cycle, which relies on heat-transfer processes. These variations are systematically illustrated in Fig. 34. Several design concepts exist for combustion chambers in gas turbines, such as tubular, annular, or multiple designs [209]. The primary objective of these chambers is to supply the turbine with an optimal combustion profile, aiming for the lowest emission rates and the longest operational lifespan.

Nowadays, gas turbines are a major source of global power generation. Despite their capacity to generate substantial power outputs with a relatively small physical footprint per unit of power, they are more complex compared to other power generation technologies. Nevertheless, integrating ammonia-fuelling systems into gas turbines aligns with governmental strategies for transitioning to net-zero emissions by 2050. However, as mentioned earlier, the use of ammonia in these systems tends to result in high levels of NOx emissions and other pollutants. This challenge has prompted various groups worldwide to develop new systems that balance low pollution profiles with high efficiency and low maintenance requirements. While research and development are ongoing to realise an ultra-low polluting ammonia-fuelled gas turbine, current progress and initiatives reveal the promising potential of these technologies. There are discernible pathways for further enhancement in this area. A comprehensive overview of the research, development, and commercial advancements in ammonia-fuelled gas turbines is presented in Fig. 35.

In the 1960s, pioneers in the field like Verkamp et al. [210], Pratt [211], and Newhall and Starkman [212] embarked on the initial exploration of using ammonia in gas turbines. Their research shed light on the challenges posed by ammonia's higher ignition energy and its slower reaction rate with air compared to other fuels. They also identified the potential of enhancing flame speeds with hydrogen-embedded ammonia. The U.S Army Engineer Research and Development Laboratories, in collaboration with the Solar Division of International Harvester Company [41], further investigated ammonia as a viable

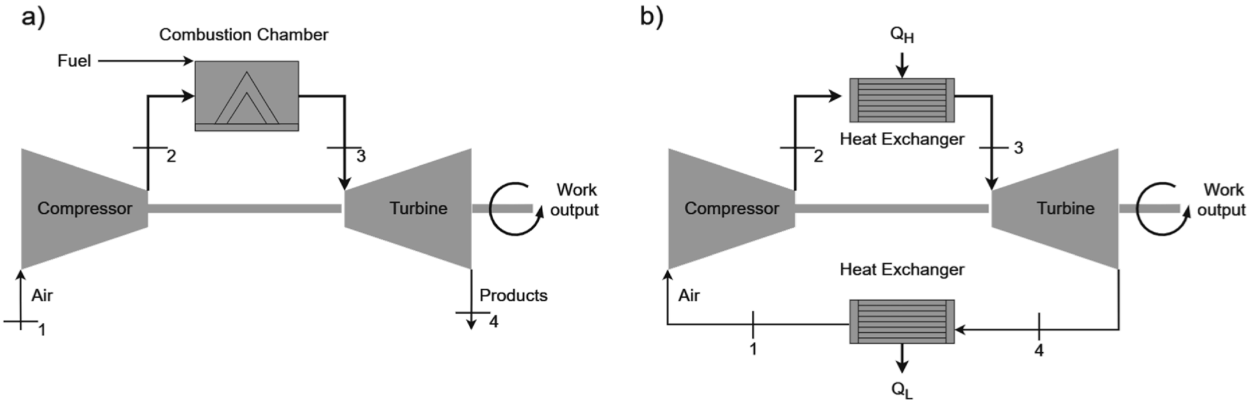

Fig. 34 A gas turbine Brayton power cycle (**a**) Open cycle and (**b**) Closed cycle

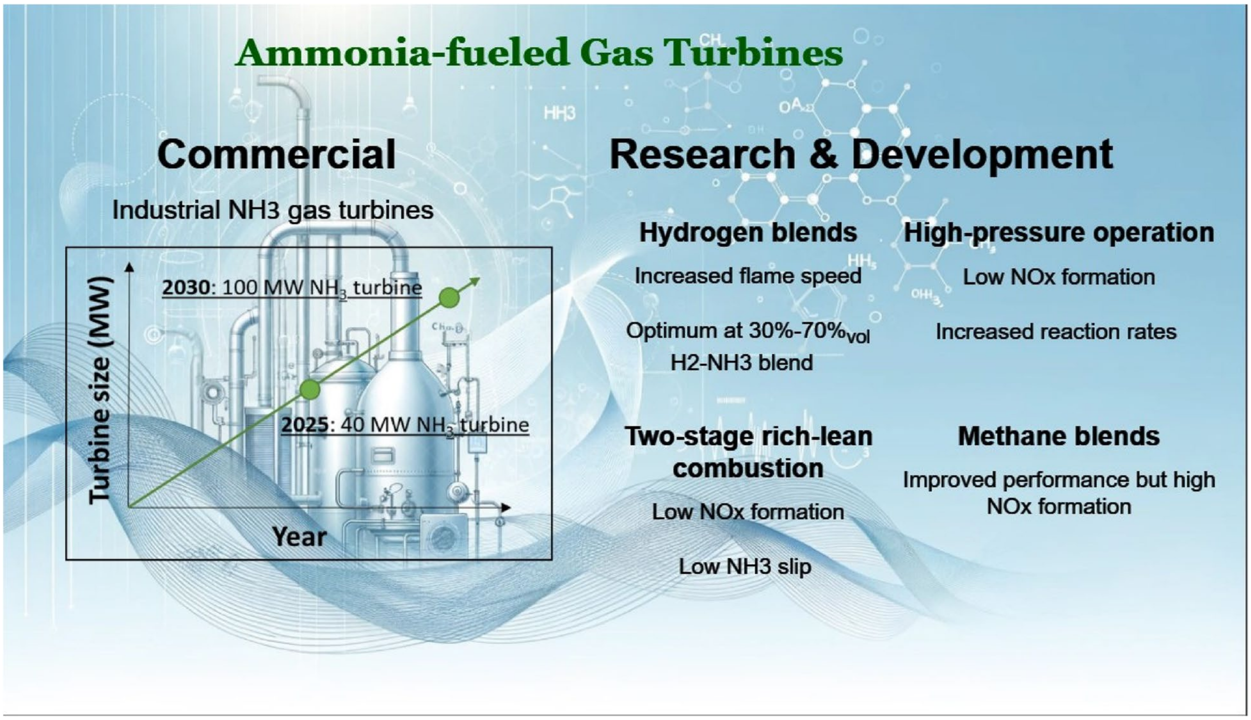

Fig. 35 Research and development into Ammonia-fuelled gas turbine power generations

fuel for gas turbines. This joint effort resulted in successful operation of gas turbines powered by ammonia, without significantly increasing cost or design complexity. However, the findings also highlighted the need for further research to advance this technology [41]. Another U.S project aimed at assessing ammonia-fuelled gas turbines for UH-1D helicopters and CV-7A fixed-wing aircraft also took place [213]. While the results validated the functionality of ammonia in these jet engines, the associated costs deemed the use of ammonia unfeasible for such systems, leading to the discontinuation of the programme. In addition, in the aerospace sector, ammonia technology found its way into propulsion systems. NACA Langley Aeronautical Laboratory (now NASA Langley Research Centre), in collaboration with the Reaction Motors Division of Thiokol Chemical Corporation [214], developed the Thiokol XLR99-RM-1 rocket engine for the X-15 research aircraft. Despite technical challenges that eventually halted the programme, the X-15 completed 199 successful missions, reaching a maximum speed of 6.7 Ma. More information on these developments can be found in references [214–217].

3.3.1 Ammonia and hydrogen in gas turbines

In recent years, ambitious research programmes have been launched to enhance the use of ammonia in gas turbines. Collaborative efforts by Tohoku University, the National Institute of Advanced Industrial Science and Technology, and Toyota Energy Solutions Inc. [37, 137, 218] have successfully demonstrated that air-ammonia mixtures can be effectively used for micro gas turbine power applications. These mixtures exhibit stability over a wide range of air–fuel ratios, pressures, and turbulence intensities. The concept of two-stage rich-lean combustion has been proposed to control ammonia/air combustion emissions, characterised by low NOx but with the trade-off of high unburned ammonia and hydrogen production from ammonia decomposition. Okafor et al. [218] developed a prototype low-NOx combustor for pure ammonia using this concept, showing promising results in a 50-kW-type micro gas turbine [219]. Their results showed that the optimum air–fuel ratio for the primary combustion zone is about 1.1 at which low NO_x emissions are ~ 40 ppm (vol%) with a combustion efficiency of 97% at 0.3 MPa. However, Keller et al. [220] reported that reducing NOx emissions using two-stage combustion or altering combustor designs remains uncertain, prompting further exploration of alternative NOx reduction methods, such as catalytic systems.

The interest in ammonia combustion in gas turbines has grown significantly [15, 16, 35]. Current trends in gas turbine development emphasise the importance of inlet parameters, such as compression ratios, air–fuel ratios, and inlet temperatures, in achieving higher thermal efficiencies with lower emissions. Otomo et al. [83] and Xiao et al. [169] found that excessive NOx can be mitigated by operating gas turbines under fuel-rich conditions with an excess of ammonia. However, this often leads to reduced thermal efficiency due to uncombusted hydrogen. The use of a secondary chamber could address this issue. Keller et al. [220] proposed a novel combined-cycle configuration using ammonia fuel to manage high NOx levels. In this system, hydrogen from ammonia decomposition, not combusted in the primary chamber, is used in a Heat Recovery Steam Generator (HRSG) to enhance power output and cycle efficiency. Operating the gas turbine under fuel-rich conditions and using Exhaust Gas Recirculation (EGR) to manage inlet temperatures could balance thermal efficiency and NOx levels.

Cardiff University has been conducting ammonia combustion research since 2014, revealing the complexity of premixed ammonia blends and their combustion dynamics [5]. They observed that NOx and CO emissions were low at air–fuel ratios higher than 1.10 with methane and 1.20 with hydrogen as doping agents. Mixing ammonia with hydrogen significantly increases the Laminar Burning Velocity (LBV) as shown in Fig. 36, with optimum results observed in the 20–30%$_{vol}$ hydrogen range [98]. Lower hydrogen leads to blowoff and laminar speed loss of the flame, whilst higher values shift the chemistry towards hydrogen, leading to flashback and multi-flame scenarios. These blends were experimentally investigated using a 40-kW combustor under fuel-rich conditions [16]. However, high hydrogen concentrations led to instability and a narrow operability range [16]. Further experiments at higher pressures showed a decrease in NOx emissions with increased pressure and fuel content [221]. Recent work at Tohoku University [221] showed that two-stage rich-lean combustion for co-firing ammonia and hydrocarbons with ammonia ratios below 50%

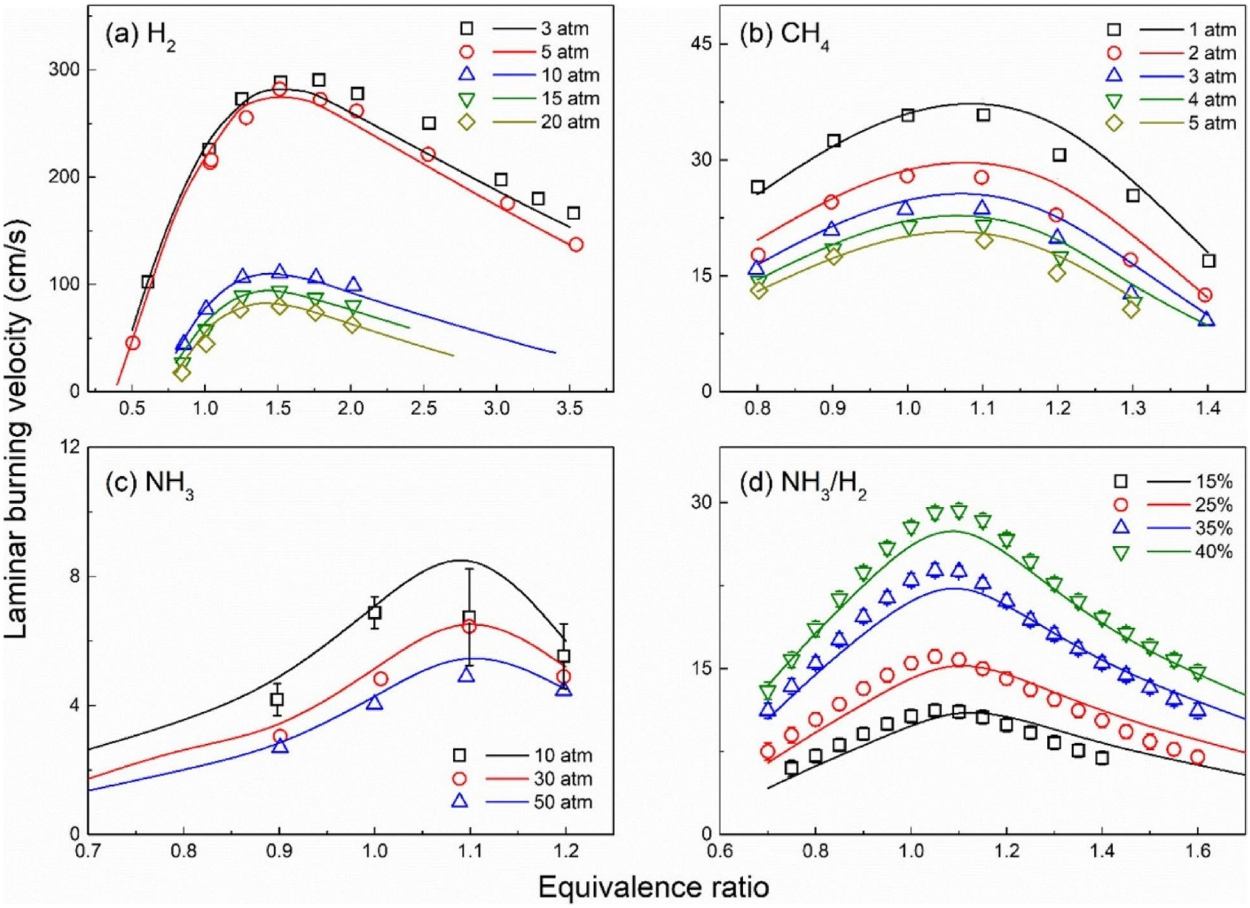

Fig. 36 Measured (symbol) and simulated (line) LBVs of (**a**) H2/O2/He mixtures at Tu = 298 K and Pu = 3–20 atm, (37) (**b**) CH4/air mixtures at Tu = 298 K and Pu = 1–5 atm, (38) (**c**) NH3/air at Tu = 298 K and Pu = 1–5 atm, (4) and (**d**) NH3/H2/air at Tu = 298 K, Pu = 1 atm, and various H2 contents (15, 25, 35, and 40%) [98]. (Reprinted from Energy & Fuels, 2022, Vol 36(15), Zhang, J., et al., Unravelling Pressure Effects in Laminar Flame Propagation of Ammonia: A Comparative Study with Hydrogen, Methane, and Ammonia/Hydrogen, Pages No.8528-8537, Copyright (2023), with permission from American Chemical Society)

could achieve combustion efficiencies of 99%. Similarly, multi-stage combustion research at Cardiff University demonstrated potential using ammonia/hydrogen blends under humidified conditions [110]. The direct benefit of humidification in gas turbine cycles includes increased power and efficiency, offering improved economics for ammonia use [15].

Another area of exploration is very fuel-lean combustion, which can lead to ultra-low NO and NH3 production [222]. However, this approach increases N2O emissions significantly, challenging the goal of climate change mitigation. Recent studies by Pugh et al. [110] suggest that increased pressure (of ~ 2.8 bar) can reduce N2O production. Combined Cooling, Heating and Power (CCHP) systems fuelled with ammonia blends represent another promising solution. Gutesa-Bozo et al. [154] demonstrated the potential of these systems to deliver power profiles at efficiencies close to 60%, competing with fossil fuel alternatives. A novel project in Norway is exploring the thermoacoustic characteristics of ammonia-hydrogen blends for gas turbine combustion systems [5], aiming to minimise retrofitting requirements in commercial turbines. Japan's Cross-Ministerial Strategic Innovation Promotion (SIP) framework [223] is facilitating research to meet the country's energy demands using ammonia. Tokyo Gas leads a consortium aiming for 20% ammonia co-firing by this decade and 100% ammonia combustion by 2050 [224, 225], as shown in Fig. 37. Mitsubishi Heavy Industries plans to operate a 40MW power unit running on pure ammonia by 2025 [226], while General Electric (GE) and IHI are collaborating on similar-sized power units [227], indicating a strong future for ammonia-fuelled gas turbine technology.

3.3.2 Emissions mitigation strategies

Nitrogen oxide emissions are a significant obstacle in developing technologies that utilise nitrogen-based fuels like ammonia for power applications. Decades of research have underscored the necessity of understanding the pathways of nitrogen oxide species production to control their formation. As mentioned earlier, ammonia combustion can be impacted by all three main mechanisms of NOx formation, with fuel NOx being particularly critical for ammonia. Understanding the chemical processes during combustion is crucial, with factors like blends, injection strategies, and operational conditions (temperature, pressure, flows) all influencing the reaction mechanisms in the combustion chamber. This area remains a vital point of research and development. Unburned ammonia emissions also significantly impact these systems' emission profiles. Regulations will likely address any ammonia slip, influencing combustor design. Ammonia in exhaust gases can lead to particle matter formation, including ammonium sulfides and nitrates, thus harming the environment. Multi-stage combustion presents a potential solution, using unburned ammonia from the first stage in a subsequent stage, thereby reducing both ammonia and NOx emissions through chemical recombination at high temperatures [229]. Xiao et al. [168] demonstrated that NO formation significantly decreases at pressures higher than atmospheric, though further validation is needed under industrial conditions.

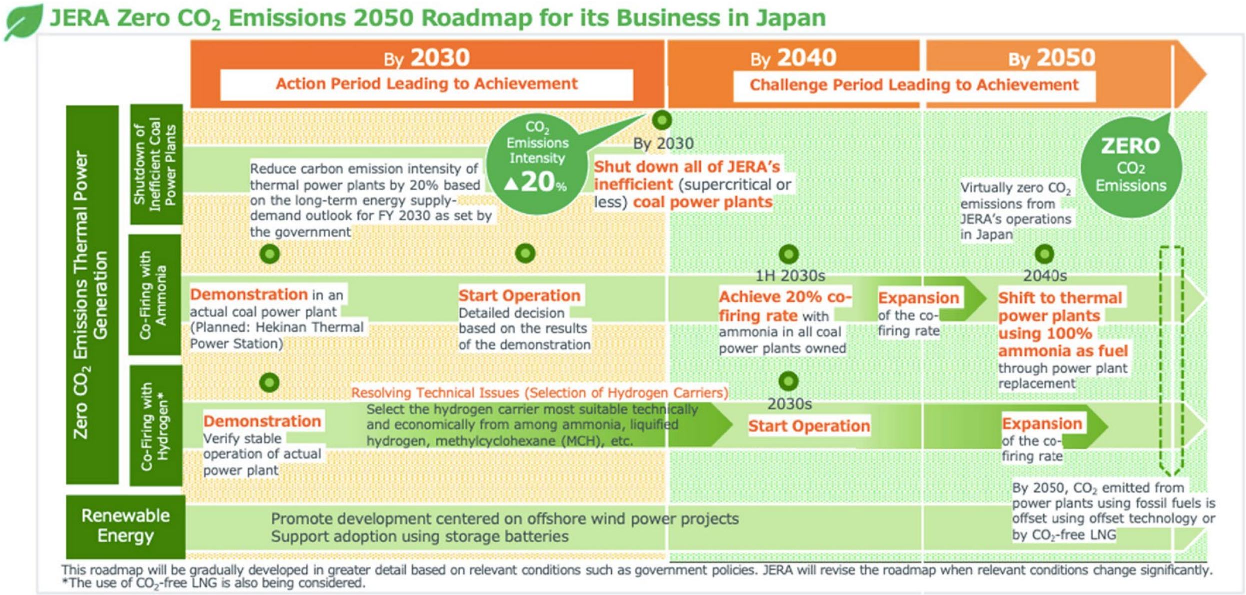

Fig. 37 Japan's 2050 Road Map for net zero emissions [257]

CO, hydrogen cyanide (HCN), and formaldehyde emissions have been observed in combustion experiments using methane and ammonia. While HCN and formaldehyde are typically decomposed post-combustion, slight traces may remain at higher power outputs, a concern that requires further investigation. The use of tertiary blends, such as methane, ammonia, and hydrogen, has been explored by Mashruk et al. [230], revealing improved combustion stability with up to $30\%_{\mathrm{vol}}$ hydrogen. However, methane leads to CO and CO2 emissions, especially at high air–fuel ratios. At low ammonia concentrations combined with high methane, CN radicals' formation occurs, potentially leading to NCN and precluding NO formation. The complexities of emissions and ignition parameters in these tertiary blends still need to be unraveled. Hydrogen is preferred over methane for doping ammonia flames due to the absence of CO and CO2 emissions. Methane/ammonia blends represent a step towards decarbonisation, but ideally, methane would be replaced by hydrogen if doping is necessary. Hydrogen can be produced via cracking, as shown in analyses by Cesaro et al. [228], which indicate the feasibility of achieving over 99% purity. However, this process incurs an efficiency penalty of about 8–10% if used in pure ammonia gas turbines. Partial cracking could enable more stable ammonia/hydrogen blends, like 70–30% or 80–20%, with lower efficiency penalties [35]. Post-combustion systems like Selective Catalytic Reduction (SCR) are also viable for emission control, as demonstrated by AIST [12]. It is likely that the first generation of large ammonia-based gas turbines will rely on SCR systems to decompose slip ammonia and produce water and N2 at the system's outlet.

3.3.3 Materials

A crucial aspect still under evaluation is how ammonia/hydrogen blends affect combustion chamber materials. Research in this area is sparse, with only a few studies detailing the potential corrosion or impact of using ammonia in gas turbine systems. An intriguing discovery from a U.S. Army programme indicated that Inconel 600 showed no deterioration when used in components like nozzles, scrolls, or liners. Other materials like Inconel X and 713C also exhibited satisfactory resistance, although transient tests did not provide conclusive comparative data [11]. Initial investigations by AIST revealed that ammonia could lead to material degradation, but applying thermal coatings made of zeolites and yttrium might counteract this issue. Preliminary findings [231] suggest these coatings effectively prevent metal degradation, a common problem in combustion operations that generate a range of species from basic (e.g., NH4+) to acidic (e.g., HNO3). More recent research by Kovaleva et al. [232] explores how ammonia/hydrogen blends can significantly impact the casing components of turbines. Issues like hydrogen embrittlement, coupled with nitridation, can reduce the ductility of materials typically resistant to high temperatures, such as Hastelloy, and Inconel. Given the infancy of this research area, further studies are necessary to fully understand these effects. Alnaeli et al. [24] conducted a comprehensive literature review on high-temperature materials for complex components in ammonia-hydrogen gas turbines. This review emphasised the importance of material selection and design in developing efficient and safe gas turbine engines fuelled by ammonia-hydrogen blends. It highlighted the challenges posed by unique combustion properties, such as ammonia corrosion and hydrogen embrittlement, and the necessity of using advanced analytical techniques for material development. The review underscored the critical need for comprehensive material characterisation and innovative design strategies to ensure the efficiency and longevity of turbines operating with ammonia-hydrogen fuel, aligning with the global shift towards sustainable energy solutions. Nevertheless, ammonia emerges as a strong contender to replace fossil fuels in gas turbines, with research on ammonia/hydrogen blends becoming increasingly prominent. To fully leverage the potential of these energy fuels, critical research and development are imperative. This includes creating novel ammonia/hydrogen energy cycles and innovating combustion systems and turbomachinery designs for efficient, cost-effective utilisation.

3.4 Technologies for fuel preparation

The following section focuses on the challenges and potential solutions associated with preparing and handling ammonia, particularly in maritime applications, where ammonia is expected to be adopted as a fuel at the forefront. The choice of fuel preparation methods will vary based on individual vessel characteristics, the type of engine utilised, and the storage mechanism employed. Therefore, this discussion is framed within the context of fuel preparation for marine use.

3.4.1 Ammonia handling

Ammonia, a colourless gas with a distinct, pungent and suffocating smell, is a key chemical in various industrial processes, particularly in the production of fertilisers. Handling ammonia safely is critical due to its potential health hazards [233]. Ammonia is caustic and it can cause severe burns to the eyes, skin and respiratory tract [234], therefore, when dealing with ammonia, it is imperative that all associated personnel to use appropriate personal protective equipment (PPE) such as rubber gloves, goggles, face shield, chemical resistance protective clothing, and respirators [235]. According to the Ohio State University [236], 80% of reported incidents result from improper procedure, lack of knowledge or training, and failure to follow proper safety precautions. The University's researcher reported that washing with water is the first emergency measure to use when skin or eyes are exposed to ammonia for at least 15 min. On the other hand, facilities for storing or using ammonia should have proper ventilation systems to avoid the accumulation of gas, which can lead to respiratory issues or even explosions in high concentrations. In case of a leak, immediate evacuation and alerting emergency services are essential [237–239]. Training personnel in safe handling procedures and emergency response is also crucial to mitigate risks associated with ammonia exposure. Regular maintenance and inspection of storage tanks and pipelines can prevent leaks and ensure the integrity of containment systems [236, 238]. Fig. 38 illustrates the classification of the hazards' identifications for ammonia.

3.4.2 Introducing ammonia into the combustion chamber

Upon deciding to use ammonia as the primary fuel in internal combustion engines, selecting the most suitable solution to meet application demands is crucial [242]. Factors such as the load curve, engine response, peak power requirements, size, carrying capacity, volume available for fuel storage, and vehicle range all play pivotal roles in designing the overall system [240, 241]. While the choice of four- stroke or two-stroke engines is independent of the fuel type and more influenced by the load curves and marine-specific considerations like gearbox removal, the decision between Spare Ignition (SI) and Compression Ignition (CI) becomes more complex with the transition to ammonia [243].

The distinction between SI and CI engines is becoming less clear. With the advancement in direct injection gasoline (GDI) and varying combustion mixture fractions, the efficiency advantages of CI engines are diminishing. It is essential to remember that the Otto cycle is inherently more efficient than the Diesel cycle, and in reality, internal combustion engines (ICEs) operate somewhere between these thermodynamic ideals [244]. Historically, SI was synonymous with pre-mixed and homogeneous combustion, but this has changed with the dominance of GDI in the automotive sector, utilising lean, stratified mixtures, and multi-injections. Similarly, CI has been associated with non-premixed, mixture-controlled combustion in a generally lean mode. When considering ammonia as the primary fuel, the focus should shift from the ignition technique to the method of introducing ammonia into the engine.

Ammonia can be introduced into the engine as a gas or liquid, similar to LPG, either via the inlet manifold (akin to port fuel injection, PFI) or directly into the combustion chamber. The choice between these methods hinges on ammonia's temperature and pressure, which also determine its storage mechanism (pressurised, cooled, or both). Gaseous ammonia introduction occurs when its pressure is below the vapour pressure, around 8 bar at room temperature. This implies that gaseous injection in the inlet manifold is more feasible than direct cylinder injection, where the ambient cylinder pressure at injection start is likely above 8 bar.

The ammonia introduction method into the engine dictates the components and system requirements for the

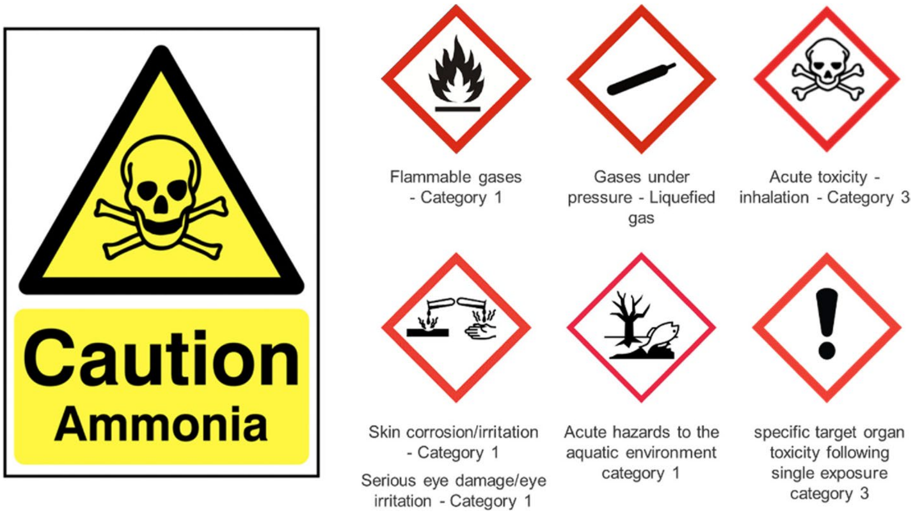

Fig. 38 Workplace Hazardous Materials Information System (WHMIS). Adjusted from [258, 259]

fuel preparation system. If gaseous ammonia is used, heat exchangers/vaporisers are needed; for liquid ammonia, low-pressure transfer pumps and high-pressure injection pumps are necessary. These specifics depend on both the introduction method and storage approach. Furthermore, certain aspects of ammonia combustion may require extra systems in the fuel preparation process. These issues, along with details on ammonia combustion, are further discussed in the subsequent sections.

3.4.3 Combustion impacts on fuel preparation

One of the main problems with ammonia is its ignition, as mentioned early. The remedy for this issue is the addition of another fuel (i.e., hydrogen) that will readily ignite. Hydrogen is a viable option as an ignition promoter, especially in spark ignition (SI) engines, due to its wide flammability range and low ignition energy. Its high diffusivity and flame speed, combined with a small flame quench distance, facilitate rapid mixture formation and more complete combustion throughout the combustion chamber, including in crevices. Adding hydrogen to the manifold where ammonia is introduced can enhance the mixture's homogeneity and combustion efficiency, reducing the likelihood of unburned ammonia emissions (ammonia slip) [245]. Hydrogen's addition increases the flame speed and decreases the quenching distance.

It could also be introduced into the inlet manifold for thorough mixing or via early direct injection. If direct injection is used, hydrogen must be pressurised above the combustion chamber's pressure. This mixed formation could be ignited either by a spark or a pilot injection of diesel fuel. Additionally, hydrogen can be produced through ammonia cracking. However, hydrogen's high auto-ignition temperature (585°C) and susceptibility to engine knock limit its use as a primary fuel in compression ignition (CI) engines, where it functions effectively only within a narrow air–fuel ratio range. 0.04–0.06 [246, 247]. Hence hydrogen should not be thought of as an ignition promotor for CI engines.

Alternatively, fuels with a higher cetane number, similar to those used in natural gas, some hydrogen, and LPG engines, can be employed as pilot fuels in CI engines. These fuels readily ignite under compression, subsequently igniting the ammonia-air mixture. This category includes diesel fuel, broadly defined as any fuel suitable for compression ignition conditions. Other more advanced ignition techniques, like lasers and microwaves, are under exploration but are not expected to be practically implemented in the near future.

In summary, hydrogen is beneficial as an ignition promoter in SI engines running on ammonia, enhancing overall combustion quality by improving flame speed and reducing quenching distance. While diesel fuel or a similar ignition promoter is necessary in CI engines using ammonia, hydrogen can still enhance overall combustion efficiency. Determining the optimal hydrogen concentration for different engine types remains an area of ongoing research.

3.4.4 Fuel preparation systems

Given ammonia's different usage methods, additional fuel requirements (like hydrogen, diesel, methane), and storage methods, fuel preparation systems will likely comprise diverse components to ensure ammonia is delivered to the engine at the right temperature, pressure, and quantity. If stored as refrigerated liquid and used as gas by the engine, ammonia will require an evaporator, potentially utilising waste exhaust heat, engine cooling water heat, or seawater heat. Cryogenic pumps will be needed for transferring refrigerated liquid, while gaseous ammonia can be managed with low-pressure transfer pumps. The emerging reality of direct liquid ammonia injection could reduce the energy demand for evaporation and offers better safety than pressurised storage. In the event of leaks, refrigerated ammonia would form pools, vaporising slowly and reducing the risk of toxic clouds, although it can still cause serious skin burns. Pressurised ammonia, on the other hand, could lead to more hazardous leaks, forming a mist that limits oxygen availability and is corrosive to respiratory tracks [248].

One method for liquid ammonia handling, akin to current liquid gas engines, involves transferring cold liquid ammonia from storage to a pressurised service tank using a heater and pump. This service tank would accommodate boil-off gas (BOG) and unused return ammonia from the engine. Ammonia from this tank could then be used as gaseous or liquid fuel, with a low flashpoint fuel supply system (LFSS) ensuring proper fuel temperature, pressure, and quality. The fuel valve train (FVT) would safely isolate the engine during fuel switch, shutdown, or maintenance, with nitrogen purging employed for safety [249]. Unused fuel will be returned to the service tank after filtration and removal of water and other contaminants.

If ammonia is used in a dual-fuel engine with a diesel pilot, the ammonia and pilot fuelling systems would be separate, with minimal interaction. If hydrogen is also used, it could significantly impact the ammonia fuelling system. Hydrogen can be generated from ammonia through catalytic cracking, potentially reducing the need for fossil-based pilot fuels and enhancing spark-ignited engine efficiency. However, the practicality and cost-effectiveness of such a system, especially at sea, remain uncertain. Most research in this area has focused on the use of some form of the catalytic reactor [31] that accepts a portion of the ammonia, (in gaseous form) and at temperatures in the 500°C range (depending on the catalyst used). There is some limited lab and field-based demonstration of this approach to hydrogen generation for an internal combustion engine fuelled with ammonia, and the current crop of ammonia engines under development do not include this in the systems, but it is hoped that some form of ammonia cracking will be used to reduce the requirement of the fossil-based pilot fuels and to use the ammonia more effectively in a spark ignited engine. The temperature for the catalytic reaction in the systems produced so far has come from the engine exhaust gases. Whether this would work on a vessel, with the limitations on the placement of the cracker near the exhaust system (ideally on deck) and the demand for heat from the exhaust for other parts of the fuel system and exhaust after treatment (SCR). BOG could also be disposed of in the cracker, or brunt in a secondary gas combustion unit to provide heat for the cracker. The real-world effectiveness of such a cracking system has not been fully established, nor has the cost of production, cost of operation or the expected lifetime of such a catalytic reactor at sea effectively assessed.

The high toxicity of ammonia necessitates stringent safety measures, including double-walled pipelines for enclosed spaces, leak detection systems, and venting or containment capabilities for leaks. Spaces like tank connection spaces (TCS) and fuel preparation rooms (FPR) should provide secondary barriers against ammonia leaks. On deck housing of fuel systems could minimise leakage risks in enclosed spaces, and airlock systems might be needed in certain areas to contain potential leaks. A continuous airflow is maintained within the outer pipe, a method already implemented in LPG engines, to enhance safety. The Tank Connection Spaces (TCS) and Fuel Preparation Rooms (FPR), which accommodate the Low Flashpoint Fuel Supply System (LFSS) and the Fuel Valve Train (FVT), should be designed to act as secondary barriers against ammonia leakages, especially in areas where implementing double pipe protection is not feasible [236]. To further minimise the risk of leakage, particularly in enclosed spaces, a significant portion of the fuel systems could be located on deck. In some cases, the installation of airlock systems might be necessary to ensure that any leakage within enclosed spaces can be effectively contained and managed.

3.4.5 Selection of materials

Ammonia is highly corrosive towards copper, zinc, nickel and their alloys (in the presence of water), some plastics and typical gasket materials (such as Viton) [250, 251]. These materials should not be used in any ammonia systems. Iron and steel are the main metals used in ammonia storage tanks, piping and fittings. Anhydrous liquid ammonia can cause stress corrosion cracking (SCC) of vessels made of carbon steel and high-strength low-alloy steel at -33°C but most stainless steels show no cracking tendencies with ammonia under any conditions [252–254]. For low-temperature storage, low temperature steels are required.

There is a requirement to design systems to avoid ammonia contamination from engine oil and vice versa [255]. Work needs to be conducted to examine the take-up of ammonia in engine oil and lubrication systems. As with LPG, some form of sacrificial oil barrier may be used to ensure ammonia does not mix with hydraulic oil in any high-pressure liquid injectors. Due to the demand for heat in the fuelling systems, crackers or SCR, there will be demand for heat exchangers utilising engine cooling water and exhaust gas heat. The heat exchangers should be designed and operated so any ammonia leakage would leak into an intermediate substrate, with ammonia detection in any coolant that operates in sensitive areas, such as that circulated around the engine [256].

4 Conclusions and future directions

The increasing demand for clean and sustainable energy motivates the exploration of alternative non-fossil fuels, such as ammonia and hydrogen. However, the large-scale utilisation of hydrogen as a fuel source meets inherent problems of storage and distribution. In comparison, ammonia is considered a good candidate for the future energy sector as a consequence of its superior properties such as zero-carbon emission, easy storage, and high energy density. This review highlights the status quo of green ammonia technologies from both academic and industrial prospectives, offering state-of-the-art knowledge and up-to-date ammonia's application progress as primary energy vector. Some conclusions are drawn to address the different topics of ammonia as follows:

- Key challenges with ammonia as its low flame speed and temperature, high combustion instability, and narrow flammability range, recommend the need for future research to focus on the combustion characteristics of novel ammonia blends and factors influencing ammonia-based flames such as fuel mix ratio and inlet pressure and temperature.
- Regarding liquid ammonia spray, increasing the injection pressure and needle lift can improve the penetration length and cross-sectional area, whereas increased chamber pressure has counteracting effect due to intensified air drag force. The Ignition Delay (ID) and Laminar Burning Velocity (LBV) of ammonia and its blends are extensively studied, noting that ammonia's ID decreases non-linearly with hydrogen

addition. This review also explored ammonia' blending with hydrocarbon fuels under various conditions.

- While the existing chemical kinetic mechanisms could effectively predict LBV variation trends, discrepancies arise, particularly in fuel-rich conditions. LBV's dependency on influential factors such as pressure, temperature, oxygen content, mixture and buoyancy effect are illustrated in this work.
- Ammonia's low LBV, high flame thickness, and high Karlovitz number (Ka) make it a candidate for fundamental studies using optical and laser diagnostics. The relationship between Turbulent Flame Speed (TFS) and LBV varies with different mixtures due to their distinct thermal-diffusion-chemical properties and flame stretch of the blending fuels. Moreover, the highest TFS was normally shown in the lean case. This is because a more furrowed flame surface led to a lower effective Le due to the interaction between turbulence and thermal diffusion instability.
- It is also found that fuel-lean ammonia flames have a higher extinction limit under high-turbulence conditions, attributed to diffusional-thermal instability. The blow-out velocity of ammonia-hydrogen–nitrogen-air flames far exceeds that of methane-air flames, owing to hydrogen diffusion. Overall, the blow-off limit increases with increased hydrogen blend ratio and air temperature yet drops higher strain rate.
- One significant challenge for ammonia's application is high NOx emissions, produced through fuel NOx, thermal NOx, and prompt NOx. There is a trade-off between NO and unburnt NH3, both reach the minimum around the equivalence ratio of 1.1. In ammonia-hydrogen turbulent flames, NO reduction is achievable through reactions with NH and NH2 radicals. Furthermore, it is demonstrated that high initial pressure could suppress fuel NO emissions due to the decrease of OH radical and enhanced NH_i (i=1,2) radical combination. While hydrogen promotes the ammonia combustion because of its higher burning velocity and low ignition energy, the side-effect of high NOx emissions may arise due to its high flame temperature and rich radical pool of O/H. Therefore, some controlling techniques, such as the equivalence ratio, fuel mixing ratio, and optimal burner design, have been investigated for deNOx of NH3/H2/air flame. It was also observed that lean ammonia conditions produce high N2O content in the flue gases, as the temperature of combustion decays consequence of heat losses.
- Despite some innovational developments in combustion systems and novel cycles that have been proposed to promote the applications of ammonia as an energy vector, there are still challenges to overcome. Ammonia slip, carbon monoxide (when blended with fossil fuels), and flame stability are just a few challenges to account for. For furnaces and boilers fuelled with ammonia, it is promising to integrate Rankine or combined power cycles. MILD combustion is also considered an effective technique to increase combustion efficiency while suppressing NOx emissions. Some other controlling strategies, such as water addition and multi-stage combustion, are also practiced.
- In ammonia-fuelled ICEs, dual-fuel modes are beneficial, with ammonia potentially replacing the majority of the fossil fuels to reduce carbon emissions while maintaining fuel efficiency. Pure ammonia combustion is achievable in SI engines, but adding a small proportion of hydrogen or hydrocarbon fuels is still recommended to improve engine performance and emissions. Control and after-treatment techniques such as ammonia cracking, Exhaust Gas Recirculation (EGR) and Selective Catalytic Reduction (SCR) are recommended for reducing thermal NOx production in ammonia-fuelled engines.
- Recent developments in ammonia-fuelled gas turbine show that some parameters like compression ratios, air–fuel ratios and inlet temperatures in achieving higher thermal efficiencies and lower pollutant emissions. Innovative operating concepts like two-stage rich-quench-lean combustion, premixed combustion, and humidification are proposed for deNOx practices.
- This review also discusses ammonia fuel preparation and handling, emphasising the need to deliver ammonia to the engine at the right temperature, pressure and quantities. They are also dependent on many factors, such as the engine type, load curve, fuel injection, peak power, and so forth. Due to ammonia's toxicity, safety measures, such as double walled pipelines, detection monitors, and ventilation air, are recommended in case of leakage from the ammonia appliances. Materials such as Iron and steel are suggested to safeguard against corrosion or cracking in ammonia systems.

Collectively, these aspects, briefly covered in this review, highlight ammonia's potential as a crucial element in the future energy mix.

Abbreviations

CFD	Computational Fluid Dynamics
GE	General Electric
H2	Hydrogen
ID	Ignition Delay
IDT	Ignition Delay Time
IHI	Ishikawajima-Harima Heavy Industries Co., Ltd"
LBV	Laminar Burning Velocity
NH3	Ammonia
NOx	Nitrogen Oxides
R&D	Research and Development
SOx	Sulphur Oxides

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Authors' contributions

MA, HS and AVM were major contributors to writing the manuscript. WN, DE, SK, PW, MA, SM, KR, CY and SE participated in the discussion of this work. MA reviewed and edited the manuscript. All authors read and approved the final manuscript.

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Declarations

Ethics approval and consent to participate

All authors certify that the manuscript is original and not submitted to other journals.

Consent for publication

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Competing interests

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