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Plasma assisted NH_3 combustion and NOx reduction technologies: Principles, challenges and prospective

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ABSTRACT

Combustion is the cornerstone for generating energy in various sectors and applications. The predominance of hydrocarbons combustion causes several environmental and human health issues. The ever-increasing demand for decarbonization of combustion technologies in order to reduce or eliminate undesirable emissions has created a major challenge, putting tremendous pressure on the international research community to develop alternative fuels. Ammonia has recently been explored as a promising carbon-free fuel and a great alternative to hydrocarbon fuels to achieve zero-carbon emissions. However, despite its advantages over hydrogen, ammonia combustion has some drawbacks including low burning velocity, flame stability, and high NOx emissions. This review begins with energy and environmental issues before moving on to hydrogen and ammonia as promising potential solutions, their advantages and drawbacks, and ammonia combustion kinetics. It summarises the principles of chemical kinetics of both ammonia combustion and NOx reduction coupled with the chemistry, kinetic mechanisms and impact of plasma on ammonia combustion. The effects of plasma on ammonia combustion and NOx reduction are also discussed, with a comparison of NOx reduction mechanisms and plasma approach. Finally, the review discusses the perspectives and challenges of using plasma in ammonia combustion.

1. Introduction

1.1. *Energy crises and climate change*

Energy is the vital requirement for industrial application development, economic growth, and human life enhancement, thus its consumption is increasing rapidly [1]. The industrial and economic evolutions have been led by using hydrocarbon-based fossil fuels [2,3]. Ritchie and Roser [1] reported that the highest point of fossil fuels consumption was at 81.5% and inevitably depleting at a faster rate. Moreover, climate change is a severe global problem resulting from pollutants such as CO_2, CO, NOx, Sox and among others, occurring from the utilisation of hydrocarbon fuels [4]. The combustion of fossil fuels and global electricity generation based on the survey made in 2018 suggest that 61% of electricity was produced from hydrocarbon fuels, as shown in Fig. 1. This has resulted in elevating CO_2 levels, negatively impacting on the world's climate by increasing the global temperature [5]. Fig. 2 shows the global total CO_2 emissions from 1990 to 2020 [6]. The remarkable decrease in energy demand led to the astonishing decrease in global CO_2 emissions. The worldwide CO_2 emissions in the first quarter of 2020 were 5% lower than the first quarter of 2019 due to emission drop from coal by 8%, from natural gas by 2.3%, and from oil by 4.5%. The emissions of CO_2 declined in the countries that endured the initial and major impacts of COVID-19, such as the EU (−8%), China (−8%), and the US (−9%), with slighter climate conditions effect on the emissions falloff in the US [5].

Several processes have been investigated for carbon capture, separation, storage and utilisations for other applications. Absorption and adsorption are the most common and commercial techniques for carbon separation and reduction. NOx and SOx are considered as the main pollutants responsible for environmental issues such as photochemical smog, regional haze, acid rain, and tropospheric ozone. Fig. 3 shows the global total GHG emissions from 1990 to 2018 [6]. There are three approaches which have been widely studied in order to mitigate the harmful effect of NOx and SOx emissions, these are known as: pre-combustion, control-combustion and post-combustion methods [7]. From this perspective, the nations have been making initiatives towards mitigating the global warming and the Conference of Parties (COP) is conducted by the United Nations every year to discuss alternative solutions to reduce GHG emissions [1]. In the Paris COP 2015, it was concluded that governments should reduce GHG emissions by applying

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Nomenclature and abbreviations:

CO	Carbon monoxide
CO_2	Carbon dioxide
NOx	Nitrogen oxides
SOx	Sulphur oxides
GHG	Greenhouse gas
COP	Conference of parties
α	Branching ratio
EB	Electron beam
DNS	Direct numerical simulation
ICE	Internal combustion engines
NH_3	Ammonia
H_2	Hydrogen
ϕ	Equivalence ratio
SI	Spark ignition
NRP	Nanosecond repetitively pulsed
NTP	Non-thermal Plasma
GA	Gliding arc
LBO	Lean blow off
CP	Constant pressure
PLIF	Planar laser-induced fluorescence
LBV	Laminar burning velocity
NPD	Nanosecond pulsed discharges
GPSA	Global pathway selection analysis
DeNOx	De-Nitrogen Oxides
SLM	Standard litre per minute
E/N	Electric field reduction
SCR	Selective catalytic reduction
SNCR	Selective non-catalytic reduction
PM	Porous medium
ppm	Parts per million
MILD	Moderate and intense low-oxygen dilution
R1	Chain-branching reaction
R2	Chain termination reaction
PRFs	Pulse repetition frequencies
GAP	Gliding arc plasma
DBD	Dielectric barrier discharge
RBO	Rich blow off
λ	Lambda

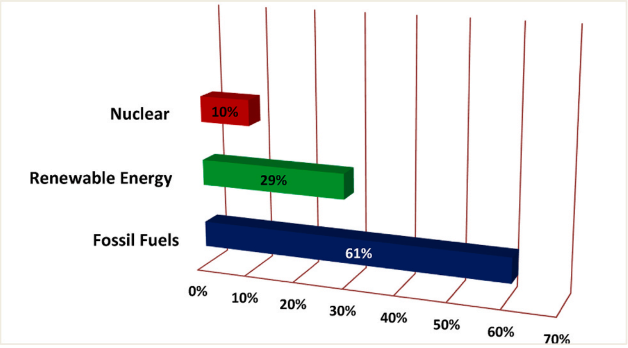


Fig. 1. Electricity generation from various sources of fuel. Reproduced from Ref. [5].

long term strategies for sustainable energy development [8].

1.2. Hydrogen and ammonia

The reduction of GHG emissions can be achieved through various alternative solutions like solar, wind, biomass, tidal and hydro energies, which can be stored in batteries and/or utilised directly [9]. Chemical energy is a more favourable continuous form of energy, and hydrogen (carbon-free fuel) is one of the most promising solutions for achieving carbon neutrality due to its remarkable physicochemical characteristics and potential applications [10]. Remarkable research has been done on hydrogen utilisation. The coupling of hydrogen blending with high energy ignition were examined by multi-point high energy ignition means and double direct injection of gasoline/hydrogen, through a suitable stratified mixture. Results indicated that hydrogen blending can increase the indicated thermal energy by 6.5% and expansion of lean burn limit by 66.2% compared to pure gasoline engine. NOx emission will decrease by increasing the excess air ratio, but it will be much lower than of the equivalence ratio condition when $\lambda > 1.6$. The emissions of CO, CO_2 and hydrocarbons were lower than the equivalent ratio conditions [11]. An experimental investigation was conducted by powering multifuel burner for industrial applications with hydrogen contents between 0 and 100%, where the hydrogen input varied between 100 and 140 kW. Results illustrated that thermal load increased at constant calorific energy input by 16.3%. All blends showed high potential for decarbonization of industrial heating [12].

The role of syngas chemical composition on the instabilities of laminar diffusion flame was numerically investigated. The obtained results indicated that a higher level of instability was for low H_2:CO and did not increase any further with a ratio less than 1 [13]. Furthermore, the heat generation with less fluctuation was encouraged by CO while H_2 increased the flame temperature and its fluctuation. CH_4 dilution into syngas raised the level of instability and heat generation fluctuation [13]. The composition of fuel effect was studied for a deep understanding of heat generation and emission of the laminar diffusion flame of syngas/producer gas. The obtained results illustrated that at H_2:CO = 3:1, 1:1, and 1:3 the generated heat flux was 222, 432, and 538 W/m^2. Furthermore, adding CH_4 to syngas/producer gas raised the heat generation remarkably (614 W/m^2 at 20%) and decreased the emission formation [14]. However, the addition of 20% CO_2 and N_2 to the syngas/producer gas declined the heat generation from 432 W/m^2 to 364 W/m^2 and 290 W/m^2, respectively [14]. The effect of hydrogen content and co-flow air on the flame characteristics of a H_2/N_2 laminar diffusion flame was numerically investigated. The escalation of either hydrogen content or fuel velocity and co-flow air affected the flame temperature, chemistry heat release, flame size, and formation of NOx [15]. Higher hydrogen content in fuel had a remarkable effect on NOx emission and flame temperature while increasing fuel velocity and co-flow air affected the flame dimension and heat release [15]. Numerical investigation was conducted on the effect of methane/syngas/nitrogen/hydrogen and methane-ammonia-hydrogen and fuel blend on combustion performance and emissions. The obtained results indicated that addition of 15% of hydrogen to methane increase the maximum temperature of combustion chamber by 100 K [16]. However, the temperature dropped by 200 K by adding 15% of ammonia and the location of temperature shifted away from the point of injection. The thermal NOx increased by 28% with hydrogen content enhancement by 10%. Moreover, adding 15% of hydrogen to methane and adding and 5% hydrogen plus 15% ammonia to methane declined the emission of CO_2 by 30.7% and 14%, respectively compared to pure methane [16].

A numerical analysis of NH_3–H_2 blend (10%–50%) was performed in a high swirl gas turbine combustor with different cooling angles (15°, 30°, 45°). As a result of fuel blend the maximum temperature levels reduced in the combustion chamber due to the low burning rate of ammonia. The angle of 45° enables more homogeneous temperature distribution compared to the other angles at the outlet [17]. A numerical investigation was conducted on the premixed combustion of hydrogen/air in lean to ultra-lean conditions using platinum (Pt) catalyst in a planar channel burner. The non-catalytic burner showed no flame at $\phi \leq 0.16$ over the Re number investigated. Furthermore, at Re < 571 and Re < 381 flame extinction occurred for $\phi = 0.18$ and 0.20, respectively. Also, in all cases a significant amount of unburned hydrogen was found in the burner. However, a complete combustion of H_2 is achieved for $0.10 \leq \phi \leq 0.20$ by coating the walls with Pt catalyst, where the contribution of homogeneous reaction (gas phase) increased with Re. Moreover, NOx emission showed an increase with ϕ but had a negligible effect with the inflow Re number [18]. However, several drawbacks are associated with hydrogen utilisation due to its low boiling point (−252.9 °C) and high thermal and mass diffusivities [1]. Moreover, hydrogen has a wide range of flammability limits with air and oxygen which pose many safety issues, particularly in transportation and storage. Also, hydrogen flame is non-luminous, making the flame monitoring visually and optically difficult [19]. Therefore, using hydrogen as a fuel in energy and transportation applications poses additional challenges.

There are several chemical fuel carriers for hydrogen that have been examined for varying applications [20]. Ammonia has been found to be one of the most promising energy carriers compared to other possible fuels [21]. Ammonia can act as a green fuel, as it does not contain carbon resulting in zero-carbon emissions, and is relatively hydrogen dense, possessing 17.8% hydrogen by weight. Importantly, ammonia can be liquified either at 8.5 atm at room temperature or at −33 °C and atmospheric pressure, which makes it much easier for storage compared to hydrogen [22]. On the other hand, the requirement of ammonia storage is similar to another commercial: propane. Ammonia has a wide range of applications, and its industry has been stable over the past 100 years. Steam reforming process of natural gas contributes about 85% of the world's ammonia production through the Haber-Bosch process. However, the conventional ammonia production process elevates NOx levels, which could be decreased through selective non-catalytic reduction [23]. Furthermore, carbon-free ammonia can be achieved through optimising the Haber-Bosch process and using renewable electricity [24]. The simplicity of liquefying ammonia reduces the cost of transportation, which is associated with the cost of development [25].

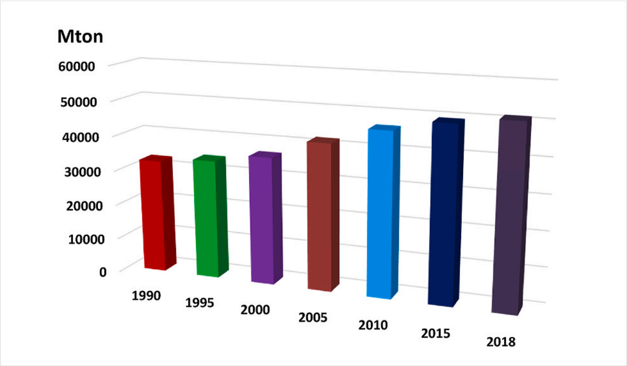


Fig. 3. Global total GHG emissions, 1990–2018 [6].

Furthermore, a considerable amount of transportation and storage infrastructure already exists, as well as an established safety handling procedure, therefore a significant investment reduction will be achieved in further infrastructure development for ammonia and training costs [26].

Numerous researchers have examined the utilisation of ammonia in different combustion systems, including internal combustion engines, thermal power generation (gas turbines and coal-fired boilers) [27]. The concept of powering internal combustion engines (ICE) with ammonia from hydrogen was put forth by Norsk Hydro in 1905 [9]. Ammonia was first used in trucks in Norway in 1930, and later enhanced in Belgium in 1943. During the 2010s, cars powered with ammonia have seen the latest developments, with sports cars being the most recent application in the transportation sector [28]. A comprehensive investigation was made on fuelling spark ignition and compression ignition engines with ammonia [29]. Roughly 10% of hydrogen as a mass fraction is required for an ammonia/hydrogen fuelled engine to achieve 30% thermal efficiency and favourable engine performance. However, further studies are required to optimise engine parameters to increase the mass fraction of hydrogen. Heat release rate for a compression engine was increased by almost 10% increasing aqueous ammonia at a full load condition. Nevertheless, engine noise levels were elevated with prolonged ignition

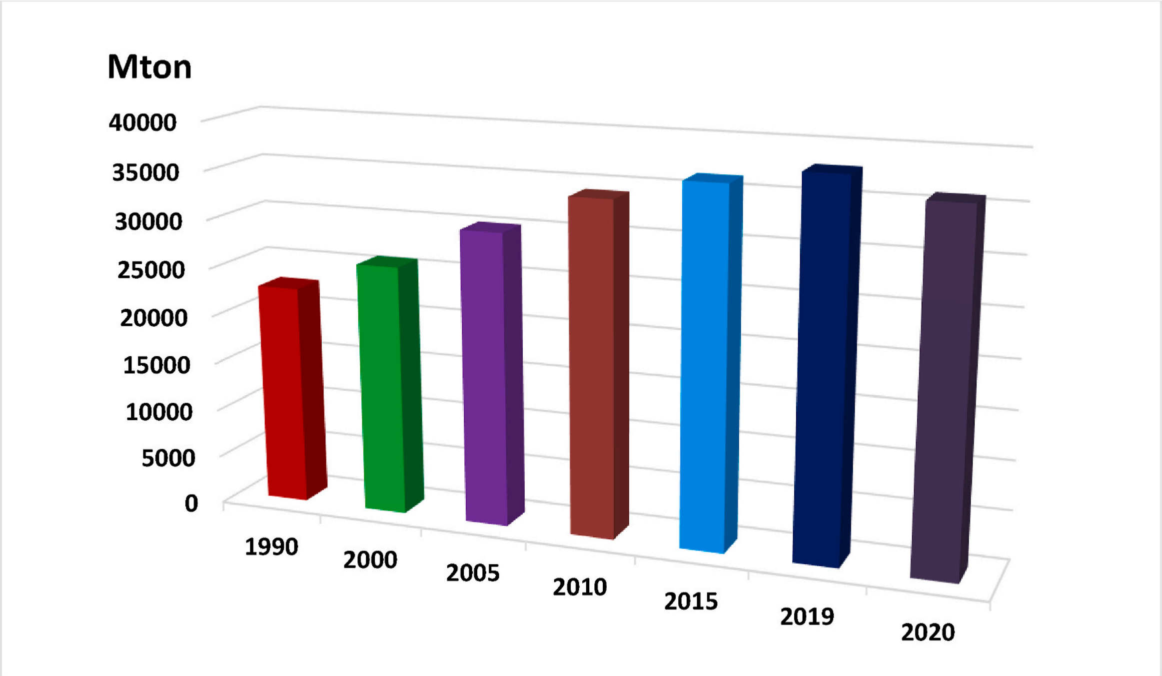


Fig. 2. Global total CO_2 emissions, 1990–2020 [6].

delay, and researchers have found that optimising multiple fuel injection of a compression ignition engine is a potential solution for improving the engine performance [9]. Experimental investigation was carried out on the effect of premixed ammonia/hydrogen/air mixture on the performance of a spark ignition engine. Cyclic stability and stochiometric efficiencies along with avoiding misfires was achieved by blending 20% volumetric percentage of hydrogen with ammonia. However, higher percentage of hydrogen leads to reduce the efficiency due to higher wall heat losses. Moreover, hydrogen has promising characteristics for accelerating the combustion, as the results show that the accelerating effect has particular impact on the primary stages of the reaction. The results indicate that NH_3 could be used as a fuel for SI engines with little to no design modifications [30].

Considerable work was conducted on the development of gas turbines during the 1960s. Currently, various approaches of using ammonia in gas turbines have been examined, with the main barrier being NOx emissions associated with combustion of ammonia directly. Recently, partial premixed combustion research in a gas turbine powered by hydrogen/ammonia is gaining a considerable interest, as it can operate at a low equivalence ratio of 0.4, and in the regime of a slight fuel-rich mixture. NOx concentration was reduced by a factor of about 5.9 at an equivalence ratio of 1.05 as compared with the stoichiometric condition [9]. An experimental investigation on a gas turbine environment using 70%NH_3–30%H_2 volumetric percentage was performed to determine its feasibility. The obtained results demonstrated that the blend has the potential to achieve combustion stability and reduce emissions for the without bypass air. However, produced emissions are above the requirement for gas turbines, therefore, new injection techniques are required to reduce NOx/unburned NH_3 in flue gases while increasing the power output [31].

1.3. Ammonia combustion drawbacks and solutions

Despite the many advantages of using ammonia as a fuel, it faces major challenges due primarily to poor combustion characteristics such as a narrow flammability range, low burning velocity because of low reactivity of ammonia, and high ignition energy [8]. Furthermore, the flame temperature and radiation heat transfer of ammonia/air mixture are lower than fossil fuels due to CO_2 deficiency in the flame [23]. High NOx formation coupled with ammonia combustion is another major drawback because of NOx emission mechanism, characterised by lower temperature and higher rate of reaction when compared to conventional thermal NOx [32]. There is a possibility of unburned ammonia in the exhaust for rich conditions (i.e. ammonia slip phenomenon) [33]. In terms of narrow flame stabilization range, high ignition and low flame speed, it is of paramount importance to increase ammonia combustion rate. In fact, various solutions have been developed to overcome this issue, including blending ammonia with more reactive fuels of high burning velocity, oxygen enrichment, and fuel preheating with an oxidizer [34]. NOx emission in ammonia combustion can be potentially controlled and reduced by using selective and non-selective catalytic reduction methods [7,35]. A swirl injector could be used to recirculate premixed gases for further reduction of NOx emissions, as swirling gases inside a combustion chamber can lower unburned ammonia and NOx emission [8,35–37]. Two stage combustion and distributed combustion are effective methods to reduce NOx emissions, achieving NOx emission standards [35,38]. Recent studies on chemical kinetics mechanisms of ammonia have suggested that increasing the inlet pressure is another potential method for reducing NOx emissions [8,39]. Ammonia slip problem could be resolved through two promising solutions: regulation of exhaust gas and dual-stage combustion [23,35,40].

1.4. Contribution of this review article

Conventional methods for enhancing ammonia combustion have the potential to improve the combustion drawbacks of ammonia and reduce NOx emissions to some extent. However, blending ammonia with hydrocarbon fuels will not avoid carbon emissions. Oxygen enrichment does not fit industrial application due to the high production cost of pure oxygen. Hydrogen, as a free carbon fuel, could be added to ammonia resulting in some improvement in laminar burning velocity, yet this approach requires a large start-up time. Therefore, it is a requirement to have an instantaneous ammonia combustion approach for practical and industrial applications [41]. Plasma assisted ammonia combustion is one of the most promising solutions to enhance the combustion performance of ammonia and to reduce NOx emission owing to the ability to control combustion instability, change transport processes, produce heat and active species. Plasma is expected to be a novel solution to control the mode of combustion and enhance engine performance [42]. Many reviews have discussed hydrogen and ammonia production as well as combustion, with a wide scale of utilisation and chemical kinetics mechanisms, providing a comprehensive study on demonstrating the conventional methods for enhancing ammonia combustion performance and reduction of NOx emissions. Conversely, this review focuses particularly on the roles of plasma in ammonia combustion kinetics, flammability, ignition, flame enhancement, NOx reduction, and its kinetics compared to the traditional NOx reduction technologies.

2. Plasma assisted ammonia combustion characteristics

2.1. Roles of ammonia combustion kinetics

To develop clean combustion technology and define flame propagation, emission characteristics and chemical reactions occurring throughout the combustion of ammonia, it is necessary to fully understand and improve the detailed chemical kinetics models [43]. The chemical kinetics of fuels help to illustrate the species generated throughout the combustion process. The Bunsen burner methodology was used to determine the flame speed and other parameters of ammonia-air mixture at 423 K, in the 19th century [44]. The development of combustion kinetic models is based on two pillars: the most authoritative thermodynamic characteristics and kinetic parameters and the updated datasets of experiments obtained through a vast range of conditions [45]. There are three different radicals H, OH, and O that are responsible for ammonia breakdown in various environments: lean, stoichiometric, and rich flames. O radicals breakdown ammonia in lean condition, OH radicals act upon ammonia in a stoichiometric condition, and H radicals breakdown ammonia molecule in the rich condition. Furthermore, researchers found that HNO, NH, and N radicals are responsible for the formation of NO, where HNO radicals have a contribution of 70% to form NO, while 10%–15% is the contribution of N radical in NO production [25,46,47]. Also, NH radicals can lead to the formation of N_2O. It was proposed that $NH_3+NH_2=N_2H_3+H_2$ was the optimal reaction to avoid NH and NH_2 radicals deteriorating at temperature above 2800 K [1,25]. It was found that the reaction of HNO + OH has the largest contribution rate during lean combustion, where HNO + H is the most import reaction pathway during rich combustion.

The H percentage in the O/H radicals is increased in rich combustion compared to lean combustion, enriching H to increase the tendency of NHx ($x = 1,2,3$) to be oxidized. The bonding reaction, NHx + NHx, is the dominant kinetic mechanism in the rich flame. The intermediate channel of NNH could generate N_2 without involving NO generation [25,48]. Therefore, NO production is reduced after the oxidation of large amount of NHx by H atoms and the reaction of self-bonding. It was elucidated that NNH dissociation is the most important reaction for generating N_2, where N_2H_2 reaction is responsible for most NNH, whereas NH + NH_2 is the main source of N_2H_2. Furthermore, results showed that a large amount of NO can be produced from the reaction of O atoms with NNH intermediates through the NHx binding pathway [49,50]. Subsequent studies found that the most important intermediate for ammoxidation and responsible for all combustion products formation is NH_2. A significant enhancement in N_2O, NO, NH_2 and other radicals prediction was recorded by improving the predicted radical profile of NH_2 [25,51]. Several studies have released various detailed chemical kinetics models for ammonia oxidation but contain some carbon species like methane. These models differ from each other in structure due to the number of reactions involved. Various studies highlighted a high complexity in the sequence of reactions for ammonia consumption and product formation [45]. Temperature has a significant impact on the branching between various possible reaction pathways. The refinement of chemical kinetic parameters and a deep understanding of the chemistry will significantly improve the prediction of ammonia oxidation behaviour in terms of combustion, emissions, and practical applications.

2.2. Principles of ammonia combustion with plasma

Plasma assisted ammonia combustion is a field which cannot be strictly classified as an additive. The utilisation of plasma could enhance the flammability limits, reduce the ignition delay time, thus has the potential to overcome the combustion and emission drawbacks of ammonia and control the combustion instabilities, further promoting the utilisation of ammonia as a fuel in practical applications [41]. This technology could modify the ammonia combustion characteristics without any waste heat recovery requirement or the limitation of cold start [52].

Various researchers have demonstrated plasma assisted combustion through different numerical models and experimental studies to understand the kinetics and physiochemical process of ammonia combustion using plasma [53,54]. A numerical investigation was performed on the oxidation of ammonia/oxygen/helium mixtures using non-thermal pulsed plasma discharge. Kinetic model assembly was utilised to simulate the effect of non-thermal plasma on ignition delay time by varying the number of pulses and the frequencies of pulsed discharges at temperatures of 600–1500 K and atmospheric pressure. The detailed chemical kinetic model was developed to include the reaction processes of all relevant electron collisions. An integration of the plasma chemistry solver and the neutral chemical kinetics solver was made to represent the pulsed discharge. The effect of nanosecond repetitively pulsed (NRP) discharge on the ignition delay was compared with the thermal ignition at active conditions. Furthermore, an investigation has been performed to explore the minimum initial temperature to ignite a mixture at a given frequency of a train of pulses, where a pulse energy of 0.05 mJ/cm^3 is used to remain in the regime of non-thermal discharge [55]. The obtained results found that the ignition delay time was reduced by 40–60% at a moderate number of pulses compared to the deposition of instantaneous thermal energy, where the higher pulse recurrence frequencies (at 1 MHz pulsation frequency) yielded shorter ignition delay times up to 90% [55].

Time evolution of OH radical analysis interestingly elucidated that higher pulse repetition frequencies (PRFs) increase the radical pool at low temperature, where radicals recombine at lower PRFs in the prolonged time between the pulses resulting in the limitation of ignition advancement [55]. Results further showed that higher pulse frequencies will support a lower initial temperature resulting in ignition, as a 500 KHz frequency resulting in mixture ignition at initial temperature of 500 K. Furthermore, it would supply high number of radicals which can alter the ignition hysteresis, extinction, and support, at all temperatures, smooth transition and reactions [56]. Moreover, the radical formed were prevalent in the conventional chain branching reaction in the thermal phase, where the formed OH radical in the discharged pulses were less consequential approaching the ignition time. The obtained results indicated that in the ignition final stages, fast pulsing combined with the diminished effect of pulses is more effective compared to the high frequency pulses resulting in ending the discharge before the thermal phase begins [55].

Experimental investigation was carried out on the effect of gliding arc plasma (GAP) and the use of a swirl burner to enhance ammonia combustion efficiency. The effects of gas flow rate, gas discharge medium, and equivalence ratio (ϕ) on the flame speed and flame stability were experimentally examined. The results from the case of injecting ammonia through the burner swirl ring and with air GAP showed that the atomic spectrum for O* and H_β along with the components of the molecular spectrum NH* and OH* dominated the optical emission spectrum of the ammonia flame, resulting in enhanced ammonia combustion [57]. Moreover, experimental results indicated that the limit of combustion for the ammonia flame expanded significantly when the air is the plasma discharge gas compared to ammonia combustion without plasma, where it would increase simultaneously with the air flow rate increment. Ammonia GAP assisted combustion led to a narrower limit of combustion for the ammonia flame as compared to air GAP [57].

The ability of using non-thermal plasma (NTP) technology such as microwave discharge, pulsed corona discharge, and dielectric barrier discharge (DBD) in the combustion systems has been investigated due to low power consumption compared to thermal plasma [58,59]. The effect of NTP generated effectively between the nozzle and flame by a novel ring-shaped nozzle head through a DBD reactor on the stability of ammonia/air turbulent and lean premixed mixtures. Non-thermal plasma effect studied by varying the applied AC voltage, the mean mixture velocity, and frequency in a modified model gas turbine swirl combustor. The NTP assisted combustion of ammonia/air flames extended the regime of stable flame to a lower equivalence ratio and boost the flame stability under both conditions of fuel-lean or fuel-rich. Columnar flame, oscillating flame, and lifted flame are the regimes of the lean and rich premixed ammonia/air flames without applying NTP. Oscillating flame could be effectively stabilised by NTP as it enhances the combustion performance resulting in extending columnar flame regime to lower equivalence ratio in the whole domain of the mean mixture velocity. However, the oscillating flame and lifted flame regimes were slightly extended at low mean mixture velocity of 4–8 m/s, where NTP did not affect the lean blow off (LBO) limit at high mean mixture velocity of 10–16 m/s as the oscillating flame with its rapid wide characteristics could not generate streamers. On the contrary, rich blow off (RBO) limit in the whole range of mean mixture velocity did not affect NTP. Furthermore, NTP extended the rich limits of the columnar flame and oscillating flame regimes at low mean mixture velocity of 4–10 m/s to higher equivalence ratio [59].

Flammability of ammonia/air combustion in a well-designed rapidly mixed swirl burner was experimentally enhanced by applying AC-powered gliding arcs (GA). Thermal and non-thermal plasma sustained in a 50 SLM of an air flow were enhanced using GA, as nanosecond pulsed discharges (NPD) would cause severe electromagnetic interference. In-situ optical diagnostics including planar laser-induced fluorescence (PLIF) imaging of OH radicals, spectrometer inspection, high-speed photography, and measurement of NH_2* chemiluminescence were performed to examine the intermediate chemistry of combustion and plasma, due to the flame and discharge complexity. The obtained results indicated that AC gliding arc discharges extended the lean blow off limit from 0.7 to 0.8 to 0.3–0.4 [60]. The effect of non-equilibrium plasma on ammonia combustion, particularly laminar burning velocity, was investigated through simulation. Simulation results indicated a small rise in the gas temperature after applying discharge of non-equilibrium plasma, resulting in increased adiabatic flame temperature. The analysis of reaction path further clarified that OH radicals generated by non-equilibrium discharge accelerates ammonia decomposition at low temperatures through a chain of ammonia combustion initiation reactions. Furthermore, the increase of decomposed ammonia amount in the unburned gas is proportional to the increase of electric field, where the laminar burning velocity (LBV) significantly increased as the non-equilibrium plasma discharge increased the H atom which has a strong impact on increasing the LBV. Also, the non-equilibrium plasma had a higher effect on increasing the mass flux and more reduction rate of flame thickness compared to preheating combustion [61].

Flame enhancement was experimentally investigated through applying non-equilibrium plasma with a nano second high voltage pulse generator with voltages and frequency ranges of 6–15 KV and 2–26 KHz, respectively. Results indicated that the ammonia/air flames were always stable at the burner exit through applying plasma. Lean blow off (LBO) limit was extended significantly at plasma voltage, frequency, discharge power, thermal load, and equivalence ratio of 11 KV, 7 KHz, 39 W, 1.9%, and 0.94, respectively. LBO limit is further extended by applying the same discharge power of 39 W and raising the voltage to 15 KV with a frequency of 4 KHz. On the contrary, decreasing voltage to 6 KV with a frequency of 26 KHz made the plasma effect negligible. It was indicated a kinetic effect took place through observing the LBO sensitivity to the voltage corresponding to electric field reduction. Furthermore, results suggested that plasma increased the number of reactive species and reactant temperature, where the discharge power is proportional to them [62]. NH_2* chemiluminescence and OH planar laser-induced fluorescence (PLIF) were utilised to observe the effect of nano-second pulsed non-equilibrium plasma on premixed ammonia/air flames stabilization. The obtained results found that plasma activation could extend the regime of attached flame to a lower equivalence ratio with more intense signals of the NH_2* chemiluminescence and OH PLIF near the region of discharge at ϕ of 0.94. It was observed from the signals of NH_2* chemiluminescence and OH PLIF that plasma generates NH_2* and OH radicals, where these signals increase with the increment of power and discharge voltage. Results showed that plasma activation generates NH_2* and OH radicals at very low equivalence ratio between 0.48 and 0.57 that can be observed from the signals of OH PLIF and NH_2* chemiluminescence. Furthermore, the production of NH_2* was found to be related to the generation of OH or oxygen-related species produced by plasma, where the impact of electron could be secondary [63].

Global pathway selection analysis (GPSA) was studied to identify the global pathways that dominate and govern the combustion chemical kinetics of ammonia under various conditions. The obtained results found that the plasma reactivity is controlled by nitrogen excitation to its electronic and vibrational states and the posterior de-excitation to its ground state in the reaction cycles. The study identified and verified the dominant pathways of plasma assisted ammonia combustion, and excitation and deexcitation loops along with their importance [64]. A gas turbine model coupled with nano-second pulsed plasma was utilised to study the lean blowoff limits as a major drawback of ammonia combustion. Results indicated that plasma activation increased the formation of NH_2 resulting in a lean blowoff limits extension [65]. Flame stability of ammonia/air premixed swirling flame was enhanced by using rotating gliding arc discharge plasma along with extending the limits of blow-off (LBO) to about 0.43–0.57. Furthermore, plasma activation removed the hysteresis phenomenon of the transition between the lift-off and attached flames [66].

The impact of non-thermal plasma operating conditions on the ignition delays of ammonia/air combustion was computationally investigated. It was observed that ignition for a low equivalence ratio $\phi = 0.5$ (fuel-lean mixture) was faster as compared to a high equivalence ratio $\phi = 1.2$ (fuel-rich mixture) due to lower OH radical consumption between pulses of plasma through the inhibiting of reactivity reaction for leaner mixtures $NH_3 + OH \rightarrow NH_2 + H_2O$. However, higher pressure at 3 atm delayed the ignition through the pressure dependent recombination reaction $H + O_2 \rightarrow HO_2 + M$ which limits the reactive radicals as compared to a low pressure at 1 atm. Also, plasma discharges improved the generation of OH radicals and NH_3 reforming which had a significant impact for faster ignition as compared to deposition of thermal energy [67].

3. Plasma NOx reduction technology

3.1. Roles of ammonia - NOx emission

By considering the combustion products from the Gibbs free energy from the ammonia reaction $4NH_3 + 3O_2 \rightarrow 2N_2 + 6H_2O$, NOx does not appear as a final product. Hence, NOx emissions will inevitably be generated through ammonia combustion in real conditions. Therefore, researchers are focusing on reducing NOx emissions whether from the environment of combustion or from the mechanisms of reaction [25,68]. It was found that for ammonia/air combustion NO increases with increasing the equivalence ratio reaching its peak of 0.9, then NO decreases with further increase in the equivalence ratio until it became negligible at ϕ of 1.3. Although, burning at a rich condition will decrease NOx, unburned NH_3 will increase rapidly resulting in reduction in combustion efficiency [69,70]. According to the literature, the formation of NO is through the radicals of HNO, NH, and N, and the intermediate reactions according to the flame types and radicals are [1,46]:

For HNO:

$$HNO + M = NO + H + M \ (\text{stoichiometric flame } NH_3\text{–}O_2)$$

$$HNO + H = NO + H_2 \ (\text{rich flames } H_2\text{–}O_2 \text{ with doped } NH_3)$$

$$HNO + OH = NO + H_2O \ (\text{lean flame } NH_3\text{–}O_2)$$

$$HNO + O_2 = NO + HO_2 \ (\text{lean flames } H_2\text{–}O_2 \text{ with doped } NH_3)$$

For NH:

$$NH + O = NO + H \ (\text{stoichiometric and rich flames } H_2\text{–}O_2 \text{ with doped } NH_3)$$

For N:

$$NH + O_2 = NO + OH \ (\text{lean flames with doped } NH_3)$$

$$N + O_2 = NO + O \ (\text{lean flame } NH_3\text{–}O_2)$$

$$N + OH = NO + H \ (\text{stoichiometric flame } NH_3\text{–}O_2)$$

Researchers found that HNO contributes to 70% of NO formation, where N contributes to 10–15% of total NO production due to the slow formation rate of N atom. Large amount of O and OH radical pools are existing in fuel-lean equivalence ratios which convert NHi to NO. NOx formation is mainly through the chemical reaction between NH_2, NH, and N radicals with O_2, O, OH, and HO_2 to produce NO or by the intermediate species HNO and H_2NO [40]. One of the most NOx formation pathway is $NH_2 + O{=}HNO + H$, which has also a less favourable branching reaction at high temperatures $NH_2 + O{=}NH + H_2O$. Therefore, it is responsible for the consumption of low percentage of NH_2. Results indicated that this reaction has weak temperature and pressure dependences, as the experimental results are only available at low temperatures [40,71]. NH_2 goes to NH by H abstraction, the chemical reaction pathway of NH with O_2, O, OH to produce NO include [72]:

$$NH + O_2 = HNO + O \tag{1}$$

$$NH + O_2 = NO + OH \tag{2}$$

$$NH + OH = HNO + H \tag{3}$$

$$NH + O = NO + H \tag{4}$$

The rate constant of reactions (1) is recently modified by Elishav et al. Reactions (3) and (4) have their computing reactions: $NH + OH{=}N + H_2O$ and $NH + O{=}N + OH$, respectively [73]. HNO is converted to NO through direct decomposition as suggested by Stagni et al. ($HNO{=}NO + H$) or bimolecular reaction with H ($HNO + H{=}NO + H_2$) and the reactions of three-body collision ($HNO + OH{=}NO + H_2O$) as used by Glarborg and Shrestha Mechanisms [74]. The adopted rate constant of HNO + H and HNO + OH are similar and offered by different mechanisms, except for the Stagni mechanism that utilised the rate constant of Chen et al. of branching reactions $HNO + OH{=}NO + H_2O$ and $HNO + OH{=}HONO + H$, that are lower than other mechanisms [75].

The extended Zeldovich mechanism states that NO produced through ammonia combustion is classified into two types: thermal NO and fuel NO. Thermal NO is generated by oxidising N_2 at temperature exceeding 1800 K, where fuel NO is produced through nitrogen atoms being oxidized during oxidation of ammonia. Three main reactions of thermal NO formation through the Zeldovich mechanism: $N_2+O{=}NO + N$, $N + O_2{=}NO + O$, and $N + OH{=}NO + H$. It was elucidated that $N_2+O{=}NO + N$ is a rate-limiting reaction occurring at high temperature of 1800 K and is considered as a primary NO reduction stage in NH_3/air rich flames, as it generates more N atoms that promoting the reaction in the reverse or reduction direction [76]. It was observed from $N + OH{=}NO + H$ reaction that OH has a significant effect on the process of NO production [77,78].

Another study found that the reduction of NO and oxidation was equivalent, where they cancelled each other by increasing the temperature in the reactions of $N_2+O{=}NO + N$ and $N + OH{=}NO + H$. Therefore, it was indicated that the Zeldovich mechanism did not have a significant effect on changing the NO amount generated in the reaction [8,25]. Nevertheless, the chemical kinetics of NO reduction in which NO reacts with NHx to produce N_2 depends on the environmental conditions that inhibit or promote the reaction. NH radical used for NO reduction mainly produces N_2O subsequently consumed by H atom as in the reaction $N_2O + H{=}N_2 + OH$. The De-NOx mechanism exist through oxidation of ammonia specially in rich conditions, where NHi reacted with NO and NO_2 to form NNH and N_2O to produce eventually N_2 [40]. Thermal DeNOx is a mechanism in which NOx could be reduced effectively via NH_2 through chain-branching reaction R5: $NH_2+ NO{=}NNH + OH$ and chain termination reaction R6: $NH_2 + NO{=}N_2 + H_2O$ in a certain range of temperature and concentration of oxygen, which are the most important reactions and are sensitive in flame speed predicting and NOx formation [79,80]. The De-NOx reactions include:

$$NH_2 + NO = NNH + OH \tag{5}$$

$$NH_2 + NO = N_2 + H_2O \tag{6}$$

$$NH_2 + NO_2 = NO + H_2NO \tag{7}$$

$$NH_2 + NO_2 = N_2O + H_2O \tag{8}$$

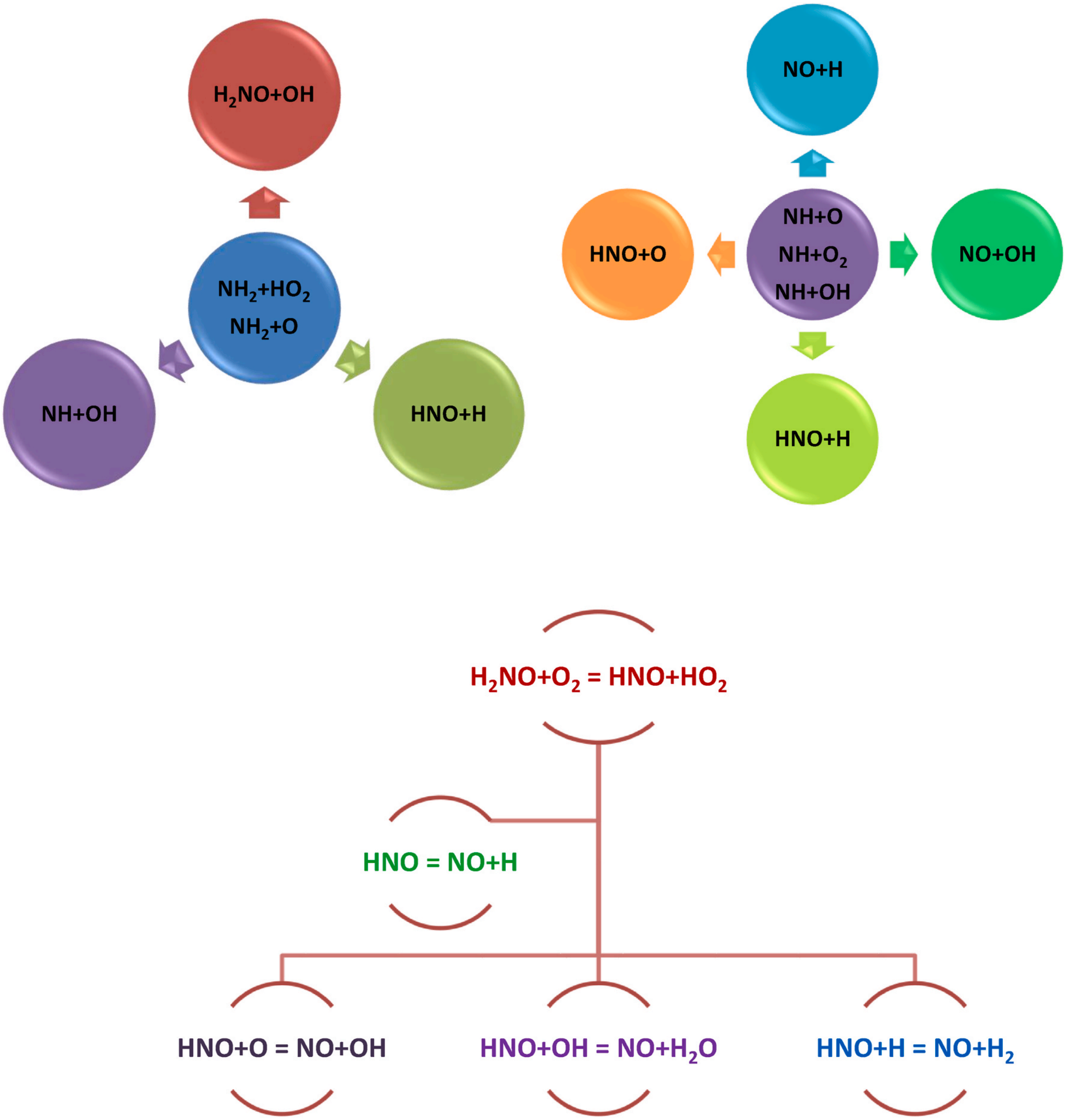


Fig. 4. NOx generation pathways with its important reactions. Reproduced from Ref. [84].

$$NH + NO = N_2O + H \tag{9}$$

$$NH + NO = N_2 + OH \tag{10}$$

Thermal DeNOx is also characterised by the self-sustaining property because of O and OH productions from R5, where the branching ratio α $= \frac{K5}{K5+K6}$ controlled the production ratio. It was also found that the ammonia reactivity control by α is a key parameter, where α depends on temperature and it has a value between 0.3 and 0.4 at a temperature range of 1100–1400 K [25,40]. Fig. 4 Illustrates the NOx generation pathways including its important reactions, where Fig. 5 shows the important De-NOx pathway reactions. Large ratios of excess air could form HO_2 radical promoting NO oxidation to NO_2, as the mutual transformation of NO mainly causes the formation of NO_2 which follow the same behaviour of NO in the measurement of emissions and favourably formed in lean conditions [81]. According to the evaluation of Glarborg mechanism the branching ratio $\frac{K8}{K7+K8}$ of reaction (8) is about 20% adopted in most mechanisms. The branching reactions (9) and (10) adopted the Baulch et al. rate constants in most mechanisms [82]. The reaction pathway of N_2O is one of the most important De-NOx reactions as N_2O comes from the oxidation of NH which eventually will react with H or form N_2 by contribute to the reactions of three-body collision $N_2O + H \equiv N_2 + OH$ and $N_2O + M = N_2 + O + M$ [40]. The branching ratios of N_2O reactions with H and O control the selectivity to form NO or N_2 from N_2O. The prediction of NO formation from N_2O is sensitive to the branching ratio of the $N_2O + O$ reaction between NO + NO and $N_2 + O_2$. The activation energy play an important role causing $N_2 + O_2$ formation at lower temperatures where the formation of NO dominates at temperatures above 1900 K [83].

3.2. Roles of ammonia - NOx reduction with plasma

Simultaneous NOx reduction from ammonia combustion can be reached effectively through the utilisation of plasma technology. NOx emissions have been shown to be reduced via non-equilibrium gliding arc plasma assisted ammonia combustion in a swirl burner. NOx

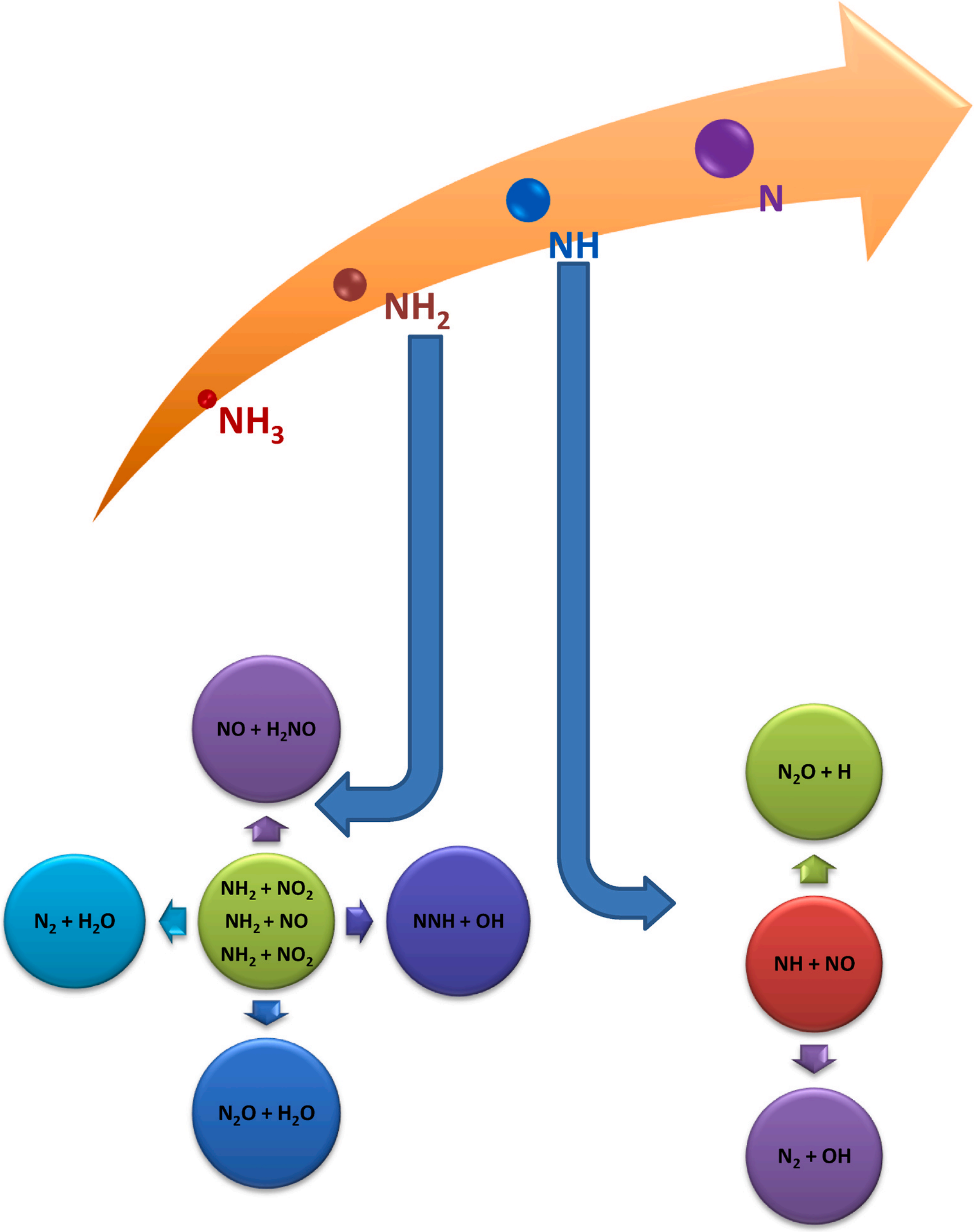


Fig. 5. Important De-NOx pathway reactions. Reproduced from Ref. [84].

emissions could also be reduced via ammonia GAP and air GAP-assisted combustion [57]. The study indicated that NOx emission was less than 100 ppm for 10 SLM ammonia flow rate and lean flame, and near zero at an equivalence ratio larger than 1.6. NOx emission was also found to be less than 100 ppm for different equivalence ratios and ammonia flow rate of 10 SLM with ammonia GAP and injection of air, where the decomposition of ammonia produced online hydrogen by ammonia GAP [57].

Another study examined the effect of non-thermal plasma (NTP) on NOx emission in a swirl combustor [59]. It was found that NOx emission is higher in lean premixed NH_3/air flames (ϕ between 0.75 and 0.95) as compared with rich premixed flames (ϕ between 1.1 and 1.3) regardless of NTP existence. Results further indicated that non thermal plasma with high discharge power and flow resistance time reduction reduced NOx emission significantly. NH_2* chemiluminescence measurements found that NTP generates more NH_2 in the ammonia/air flames resulting in lower NO concentration level as demonstrated through $NO + NH_2 \rightarrow NNH + OH$ and $NH_2 + NO \rightarrow N_2 + H_2O$ [59]. Flue gas analyser was used to measure the NOx emission generated in a swirling flame of ammonia/air assisted by AC-powered gliding arc. Obtained results indicated that plasma discharges had a low global effect on NOx reduction at high equivalence ratios. However, NO emission reached a value below 100 ppm and NO_2 was nearly zero because of the mechanism of thermal DeNOx at ϕ of 0.57. The measurements of NH_2* chemiluminescence and PLIF imagining elucidated that the discharges of gliding arc generated NH_2* and OH with stronger signals. Furthermore, unburned NH_3 that is located in a region away from plasma could reduce NOx emission in the downstream hot gas by entering the recirculation zones [60].

A significant reduction in NOx emissions was recorded by using non-equilibrium plasma generated by high voltage pulse generator at an assumed plasma power of 59 W and adiabatic flame temperature of 2009 K. NOx emission was 2645 ppm at equivalence ratio of 0.94 and calculated adiabatic flame temperature of 2007 K [62]. Results showed that increasing the voltage and discharge power had a remarkable effect on decreasing NOx emission. Furthermore, NOx decreased with increasing plasma discharge voltage with constant discharge power and vice versa, it decreases at constant discharge voltage with increasing discharge power through increasing the frequency. The study indicated that there are possible reaction pathways to decrease NOx via the formation of large quantity of HO_2 in the plasma region consuming NO and NO_2 as shown: $NO + HO_2 \rightarrow OH + NO_2$ and $NO_2 + HO_2 \rightarrow HONO + O_2$. The other pathway could be through NH_2 to consume NO through the reactions: $NO + NH_2 \rightarrow NNH + OH$ and $NH_2 + NO \rightarrow N_2 + H_2O$, both are essential for thermal DeNOx mechanism [62,85,86].

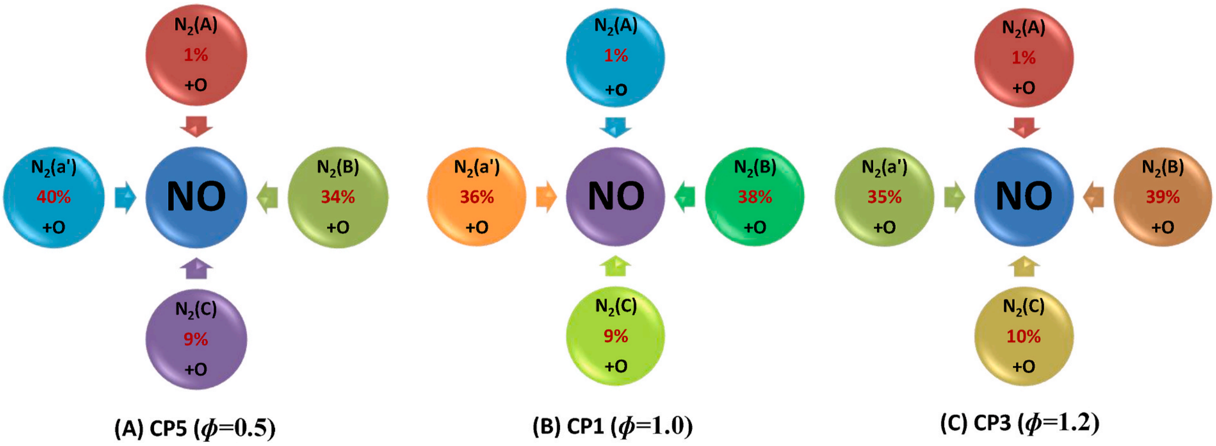
The effect of nanosecond pulsed, plasma-assisted ammonia combustion operating conditions on NOx emissions were computationally examined in reactors of constant volume and pressure. The study focused on electric field reduction (E/N), energy density, equivalence ratio, pulse frequency, and pressure as operating conditions [67]. The obtained results indicated that the relaxation of vibrational-to-translational of NH_3 and N_2 vibrational states has a significant effect on heating of gas process at lower E/N as compared to higher contribution of vibrational energy. It was found that in a lean mixture, plasma generates oxygen radical due to the presence of high content of O_2 and the posterior reactions of electronic excited N_2 states with O to form NO during the pulses resulting in higher NOx emissions of fuel-lean mixture comparing to stoichiometric and fuel-rich mixtures. NH_2 and NH fuel radicals play an important role in NO reduction when the number densities of NO reaches the maximum point for stoichiometric and rich mixtures. The NO reduction pathways: $NO + N \rightarrow N_2 + O$, $H_2 + NO \rightarrow H + HNO$, and $H_2 + NO \rightarrow NH + OH$ are due to de-oxygenation. Higher pressures also played a dominant rule in collisional quenching with higher rates during the inter-pulse gaps resulting in reducing the NOx air-bonds during the pulses because of the lower amount of N_2 and O_2 electronically excited states [67]. Plasma enhanced the production of OH radicals and NH_3 reforming that produces N_2 resulting in lower NOx generation as compared to the deposition of thermal energy. Plasma discharges helped in accelerate NH_3 consumption to produce N_2 in turn reducing the NH_3 and NH_2 radical availability at ignition to generate NOx.

Fig. 6 shows the NO formation pathway at different equivalence ratios and constant pressure (CP) conditions, while Fig. 7 illustrates the NO consumption pathway at different ϕ and CP, and finally Fig. 8 displays the analysis of consumption and formation of NO through and after the 3rd pulse, at different pressures, 1 atm (CP1) and 3 atm (CP2). The conditions for different cases of CP 0D simulations: initial temperature of 800 K and pulse frequency of 100 kHz. The E/N for CP1, CP2, CP3, and CP5 is 150 for all cases, ϕ equals to 1.0, 1.0, 1.2, and 0.5, Pressure (atm) equals to 1.0, 3.0, 1.0, and 1.0, Energy density per pulse (J/cm^3) equals to 0.05 for all cases, and number of pulses equals to 10, 20, 10, and 10, respectively [67]. The impact of nano-second pulsed plasma on NOx emissions of ammonia combustion in a gas turbine dump combustor model was examined. Plasma activation was found to increase NH_2 formation as measured by NH_2* chemiluminescence resulting in the reduction of NOx [65]. Table 1 summarises the roles of plasma technology in NOx reduction.

3.3. NOx reduction technologies

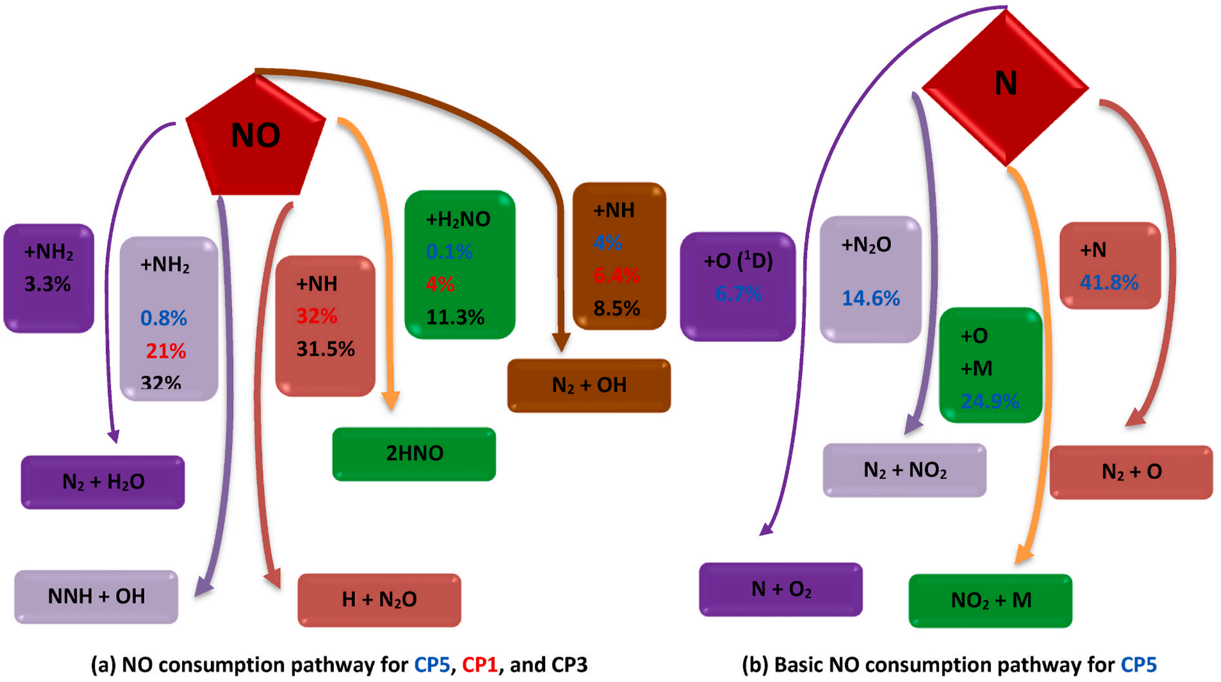
NOx is considered as one of the most dangerous pollutants and is responsible for many environmental and health problems. NOx can exist theoretically in various forms such as N_2O, NO_2, N_2O_4, NO, N_2O_5, N_2O_3, and NO_3, where it could be found in the form of >95% NO and <5% NO_2 in the exhaust gases depending on the atmosphere and temperature [5,7]. It can be classified according to the mechanisms of formation into fuel NOx, thermal NOx, and prompt NOx. Thermal NOx formation is depending on high temperature >1450 °C and is proportional to the oxygen concentration and gas residence time [4,87]. The reaction between N_2 and hydrocarbon radicals forming the prompt NOx, where nitrogen-containing species oxidation would form fuel NOx such as HCN and NH_3 [88]. The compounds of intermediate nitrogen such as CN, NH, NH_2, and N (produced by rapid conversion of fuel nitrogen compounds) can be converted to NO by the attack of oxygen species or reacted with NO to form N_2 [5,7,89].

Numerous researchers have found effective methods to reduce NOx emissions which could be classified as pre-combustion, control-combustion, and post combustion. It was found that pre-combustion method is not effective for NOx reduction by selecting fuel with low nitrogen content, where optimising the combustion process and enhancing the efficiency of fuel utilisation is used to apply control-combustion methods that could be classified into nitrogen-free combustion and low-NOx combustion [7,90,91]. The reaction temperature of flame-free gas–solid is considered as one potential solution, where limiting the peak temperature of combustion flame, residence time, and oxygen concentration in the reaction zone is another solution of control-combustion. Low-NOx combustion technologies have been developed and applied that can achieve 30–50% of maximum removal efficiency [7,92,93]. Post-combustion methods have been applied to achieve higher NOx reduction to meet the emissions standard as the pre-combustion and control combustion had low efficiency removal. The most promising technologies for NOx reduction for post-combustion are selective catalytic reduction (SCR) and selective non-catalytic reduction (SNCR) in the gas flue from stationary sources. Hydrocarbons, CO, NH_3, and H_2 are used as reductant for SCR process that react with NOx in the presence of catalyst to generate N_2 and H_2O at 260–455 °C [7,94,95]. Ammonia or urea solutions are utilised without catalyst for SNCR process to reduce NOx at a temperature of 850–1100 °C. However, ammonia injection in SNCR process tends to oxidise, forming NOx at temperatures >1100 °C. Therefore, the maximum reduction value for SNCR process at molar ratio of NH_3 to NOx between 1.5 and 2.5 lies in the range of 30–70%. On contrary, SCR



(A) CP5 (ϕ=0.5) (B) CP1 (ϕ=1.0) (C) CP3 (ϕ=1.2)

Fig. 6. NO formation pathways at (A) $\phi = 0.5$ (CP5), (B) $\phi = 1.0$ (CP1), and (C) $\phi = 1.2$ (CP3) in a constant pressure 0D reactor through the final pulse. The calculation of percentage of flux is relative to the NO species formed. Reproduced from Ref. [67].



(a) NO consumption pathway for CP5, CP1, and CP3 (b) Basic NO consumption pathway for CP5

Fig. 7. NO consumption pathways at (A) $\phi = 0.5$ (CP5), (B) $\phi = 1.0$ (CP1), and (C) $\phi = 1.2$ (CP3) at the number density of NO maximum point (when the consumption rates of NO surpass the production rates). NO consumption is represented by the percentage of flux. Reproduced from Ref. [67].

shown higher reduction value of 80%–95% at optimal molar ratio of NH_3 to NOx lies in the range of 0.9–1. However, NH_3 in both the processes could not react completely with NOx because of insufficient mixing, concentration profiles, and inappropriate temperature [7,96,97]. Numerous methods have been investigated to reduce NOx emissions such as electron beam (EB) irradiation, alkali absorption, complex absorption, catalytic oxidation absorption, and bioprocesses. Table 2 illustrates the results of several studies of reduction of NOx emissions from ammonia oxidation.

4. Prospective and challenges

The continuous and rapid progress in various industries, technologies and transportation continues to have a significant impact on daily life. Therefore, combustion in different applications such as burners, gas turbines, and internal combustion engines continues to be used globally in factories, aircraft, automobiles etc.

Plasma assisted ammonia combustion is a research field that still has challenges and research gaps and thus requires further research and improvement. Development of novel modelling, theoretical calculations and experimental methods are all essential for filling the gaps, enabling to more robust analysis and forming less complicated and suitable reaction. More experimental results are required to be carried out for laboratory as well as industrial scales, while the reaction kinetic schemes should be further reduced to capture the important reactions and require validation against experimental results. Low computational cost and improvement in the accuracy of emissions prediction could be reached by using reduced reaction models. High pressure impact on the combustion process and NOx emissions needs further studies. Also, further experimental and computational research are required to capture the chemical and reaction kinetics of plasma species in laboratory scale and real combustion systems for deep understanding of plasma impact on ammonia combustion to be applicable in practical applications.

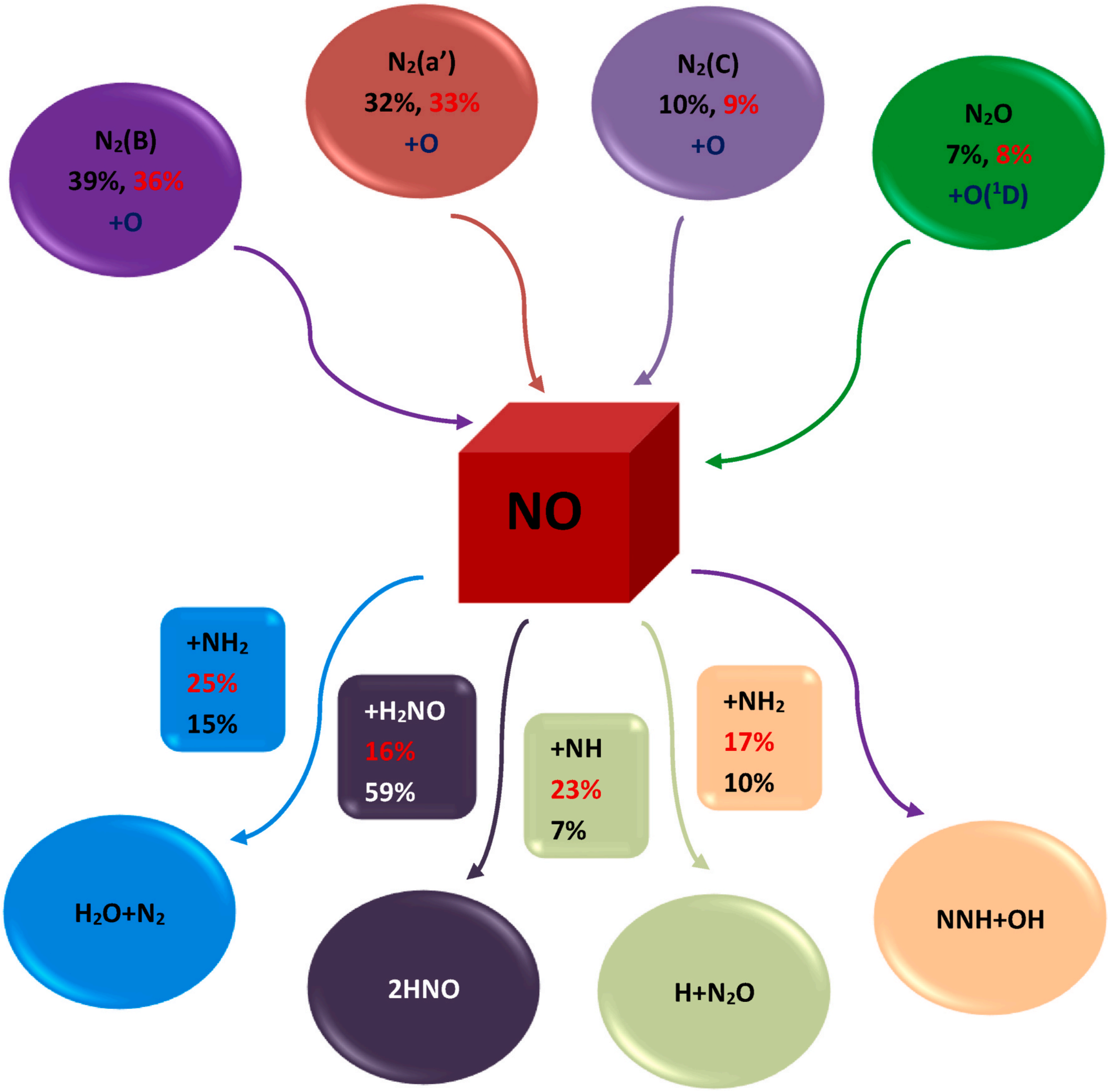


Fig. 8. The analysis of consumption and formation of NO through and after the 3rd pulse, at pressures of 1 atm CP1 and 3 atm CP2 in a constant pressure 0D reactor. Reproduced from Ref. [67].

Furthermore, more research is required to understand the effect of discharge energy, value of reduced electric field, mixture compositions, discharge duration, and different pressures. Further research is required to have detailed kinetic models for qualifying the effect of plasma under various conditions on ammonia combustion. The kinetic pathways of plasma generated vibrationally and electronically excited species with multi-dimensional models and detailed chemistry are rarely discussed as well as the impact of electrode geometry on the dynamics of discharge and ignition needs further research. Moreover, the chemical kinetics and the reaction pathways of plasma species that is responsible for NOx reduction of ammonia combustion requires further experimental and computational examination. Plasma assisted ammonia combustion is a growing research field where new discoveries could bring the field closer to practical applications.

5. Conclusion

Combustion will continue to be one of the most important cornerstones in the modern world. Conventional combustion has several drawbacks from depletion of fossil fuel reserves to environmental and health issues. NOx emissions from exhaust gases generated in various industrial sectors causes severe damage in climate change and the environment. Global efforts are carried out to mitigate the global warming by attaining the target of zero-carbon emission through utilisation of carbon free-fuel in various combustion systems. In this perspective, we explored various topics related to the ammonia and hydrogen utilisation and NOx reduction with and without plasma and the main conclusions can be shown as follows.

Despite hydrogen being a promising fuel to reduce emissions contributing to climate change mitigation, it has several limitations as it requires high cost for liquifying and transportations as well as risk of potential explosion hazards. Ammonia plays an important role as a replacement, it contains higher volumetric energy density, low cost of storage and transportation compared to liquid hydrogen. Ammonia utilisation in practical applications has some limitations due to the low burning velocity and high NOx emissions. Considerable work was made to face ammonia utilisation challenges by blending ammonia with high burning velocity fuels, two stage combustion, swirl injection, gas recirculation, and reduction of NOx associated with ammonia combustion using SRC and SCNR technologies.

Various studies also investigated blending hydrogen with ammonia. A 30% of thermal efficiency could be achieved for an engine powered

Table 1
The roles of Plasma assisted ammonia combustion in NOx reduction.

Research Methodology	Key Results
Swirl burner combined with gliding arc plasma (GAP) reactor utilised to reduce NOx emission [57].	• NOx emission was less than 100 ppm for 10 SLM ammonia flow rate and lean flame and near zero at ϕ larger than 1.6. • NOx emission was always less than 100 ppm with 10 SLM ammonia flow rate for various equivalence ratios.
The effect of non-thermal plasma (NTP) induced by a dielectric barrier discharge (DBD) on NOx emission in a swirl combustor [59].	• Regardless of NTP existence, it was found that NOx emission is higher in lean premixed NH_3/air flames (ϕ between 0.75 and 0.95) as compared with rich premixed flames (ϕ between 1.1 and 1.3). • High discharge power and reduced flow resistance time of non-thermal plasma decreased NOx emission significantly. • NTP generates more NH_2 in the ammonia/air flames resulting in lower NO concentration level.
A swirling flame of ammonia/air assisted by AC-powered gliding arc was utilised to reduce NOx emission [60].	• Plasma discharges had a low global effect on NOx reduction at high equivalence ratios. • Because of thermal DeNOx mechanism at ϕ of 0.57, NO emission reached a value below 100 ppm and NO_2 was nearly zero. • Gliding arc discharges generated NH_2* and OH with stronger signals resulting in NOx reduction. • Unburned NH_3 that is located in a region away from plasma could reduce NOx emission in the downstream hot gas by entering the recirculation zones.
Non-equilibrium plasma generated by high voltage pulse generator to reduce NOx emission [62].	• A significant reduction in NOx emissions was recorded at an assumed plasma power of 59 W and adiabatic flame temperature of 2009 K. • NOx emission decreased remarkably by increasing the voltage and discharge power. • NOx decreased with increasing plasma discharge voltage with constant discharge power and vice versa.
The effect of nanosecond pulsed, plasma-assisted ammonia combustion operating conditions on NOx emissions were computationally examined in reactors of constant volume and pressure [67].	• The relaxation of vibrational-to-translational of NH_3 and N_2 vibrational states has a significant effect on heating of gas process at lower E/N as compared to higher contribution of vibrational energy. • plasma generates oxygen radical and the posterior reactions of electronic excited N_2 states with O to form NO during the pulses resulting in higher NOx emissions of fuel-lean mixture comparing to stoichiometric and fuel-rich mixtures. • NH_2 and NH fuel radicals play an important role in NO reduction when the number densities of NO reaches the maximum point for stoichiometric and rich mixtures. • Higher pressures also played a dominant rule in collisional quenching with higher rates during the inter-pulse gaps resulting in reducing the NOx air-bonds during the pulses. • Plasma enhanced the production of OH radicals and NH_3 reforming that produces N_2 resulting in lower NOx generation. • Plasma discharges helped in accelerate NH_3 consumption to produce N_2 in turn reducing the NH_3 and NH_2 radical availability at ignition to generate NOx.
The effect of nano-second pulsed plasma on NOx emissions in a gas turbine dump combustor model was examined [65].	• Plasma activation was found to increase NH_2 formation resulting in the reduction of NOx.

with 10% (mass fraction) of hydrogen blended with ammonia fuel. Furthermore, cyclic stability, stochiometric efficiencies and avoiding misfires were improved for a spark ignition engine by using 20% (volumetric percentage). Also, using 70%NH_3–30%H_2 volumetric percentage shown a good result in combustion stability for a gas turbine. Yet, NOx emissions remained higher than those required by standards.

Chemical kinetics play a significant role in enhancing ammonia combustion and NOx reduction. Researchers found that radicals of HNO, NH, and N contribute to NOx formation, where HNO radical is responsible for 70% of NO formation. Furthermore, it was summarised that $NH_3+NH_2 = N_2H_3+H_2$ is the best reaction to avoid deterioration of NH and NH_2 above 2800 K, where the important reaction in rich combustion is HNO + H. Also, in rich flame NHx + NHx is the dominant mechanism and the percentage of H and OH radicals are increased. Studies elucidated that NNH dissociation is significant in forming N_2 and NH_2 is the source to form NNH, where it can be an intermediate to generate NO through NHx binding pathway. Other studies found that NH_2 acts as an important intermediate for ammoxidation and formation of combustion products, where temperature has an important impact on the possible reaction pathways. It was found that NO reaches its peak at $\phi = 0.9$ and became negligible at $\phi = 1.3$. However, reduction in combustion efficiency will be noticed due to unburned NH_3 at rich conditions. Various studies concluded that the Zeldovich mechanism does not have a significant impact on the NO generation in the reaction, where the reaction of NO with NHx to generates N_2 depends on the environmental conditions. Thermal De-NOx also plays an important role in the NOx reduction through the branching reactions controlled by the branching ratio.

Various studies focused on the deep understanding of plasma assisted combustion mechanisms, kinetic models, chemistry, and NOx reduction mechanisms assisted by plasma. It was found that a reduction of 40–60% for the ignition delay time was achieved using non-thermal pulsed plasma discharge for ammonia/oxygen/helium mixtures combustion, where non-thermal pulsed plasma reached 90% reduction at 1 MHz pulsation frequency. Another study found that air GAP coupled with a swirl burner enhanced and expanded the limit of ammonia combustion as compared to ammonia GAP. Assisted combustion by NTP has a significant effect on ammonia/air combustion through stabilizing the under rich and lean conditions and extended the region to lower ϕ. Rotating gliding arc discharge plasma also enhanced the flammability limit and extended the LBO limit to about 0.43–0.57, where AC gliding arc discharges extended LBO limit from 0.7 to 0.8 to 0.3–0.4. It was found that LBV increased using non-equilibrium plasma discharge, which also increases the mass flux and reduced the flame thickness compared to preheating combustion.

Considerable research has been done in simultaneous NOx reduction using plasma assisted ammonia combustion. Non-equilibrium air GAP and ammonia GAP could reduce NOx less than 100 ppm at lean flame and ammonia flow rate of 10 SLM, where it reached near zero at ϕ larger than 1.6. Another study examined NO and NO_2 emissions using AC-powered gliding arc and found that NO reached a value less than 100 ppm NO_2 of near zero at ϕ of 0.57 due to thermal De-NOx mechanism. Ammonia combustion using NTP reduced NO in stoichiometric and rich

Table 2
Results of several studies for NOx reduction from ammonia oxidation.

Research Methodology	Results
Applying a prototype biofuel combustor for combustion of ammonia and low-NOx combustor by using the combustor test rig to measure the emissions [98]	• Selective catalytic reduction was utilised to reduce NOx emissions from 1000 ppm to below 10 ppm from combustion of ammonia in prototype biofuel combustor • The modified rich-lean combustor could reduce NO_2 and NO emissions to half of the prototype biofuel combustor could decrease
Porous medium (PM) partially inserted for NOx reduction of ammonia combustion in a micro combustor [99]	• NO is reduced using PM by 81.6% from 2.54 × 104 ppm to 4.67 × 103 ppm at high inlet velocity. However, N_2O increased from 1.13 × 102 ppm to 1.32 × 103 ppm • NO emissions are decreased by increasing the porous material thermal conductivity by 99.98% from 2.70 × 104 ppm–5.17 ppm, while N_2O emissions increased from 24.8 ppm to 2.52 × 103 ppm. • Increasing the porosity from 0.5 to 0.9 resulted in reduction of N_2O emissions by 71.5% from 3.29 × 103 ppm to 9.38 × 102 ppm, while increasing NO emissions from 3.53 × 102 ppm to 1.49 × 104 ppm
New design of ammonia powered micro combustor by implementing perforated plate [100]	• It was found that the diffusion effect caused by perforated plate resulted in a considerable drop atomic ratio of N/O in the recirculation zone due to O atoms increment and less availability of N atoms that affect NO generation • The flow field, the preferential diffusion, and the conjugate heat transfer affect NOx emissions by applying the perforated plate
Developing ammonia/natural co-fired gas turbine with low NOx combustor [101]	• The obtained results illustrated that the NOx emissions were 290 ppm at ammonia mixing ratio of 20%, where it reduced to 7 ppm using selective catalytic reduction method and ammonia gas as reduction agent.
NOx reduction by reburning in pressurised oxy-combustion [102]	• It was found that NO reduced from 76% to 43% by elevating the pressure from 1 atm to 15 atm. • Furthermore, increasing the pressure resulted in boosting the conversion process of NO to N_2.
De-NOx route by adding water to reactants on a MILD ammonia combustion with two injection conditions (premixed and non-premixed) [103]	• In the non-premixed injection, by increasing the water content NOx is reduced at all ϕ values. • Increasing water content resulted in Box reduction from 440 ppm to 280 ppm. • NOx emissions were decreased by 21%, 29%, and 36% for water content of 25%, 50%, and 75%, respectively. • Maximum NOx reduction was 20% at fuel-rich and stoichiometric conditions. • The obtained results indicated that NOx emissions range was 200–800 ppm for lean condition for both modes and they reduced to 130 ppm for stoichiometric condition • NOx emissions were lower than 10 ppm at rich condition for premixed mode and lies between 10 ppm and 100 ppm for non-premixed.
NOx emissions numerical evaluation study on micro combustor powered with ammonia with three different ring-shaped rips in a fuel-rich condition [104]	• NOx emissions are reduced by 47% when x (dimensionless distance) is 3/9, and condition is compared of ϕ being 1.1 and 1.0.
NOx reduction from ammonia powered micro power system using a perforated plate [35]	• Results illustrated a reduction in NOx by up to 73.3% as compared with the absence of perforated plate due to the recirculation zone formation with a low flame temperature. • NO emissions were minimized by increasing the perforated plate hole dimensionless width.
Reduction of NOx using various injection strategies of ammonia in diesel-hydrogen engines [105]	• It was found that the shape of the flow rate of ammonia injection triangular, parabolic, or rectangular and the duration of injection had a significant effect on NOx reduction. • Results indicated that for parabolic shape of injection and 1° injection duration, NOx reduction improved by 11.4%, corresponding to 78.2% as an overall reduction.

conditions through NH_2 and NH fuel radicals, where it improved the production of OH and NH_3 reforming. Also, higher pressures play a significant role in NOx-air bonds reduction during the pulses.

Several technologies were used to remove NOx emissions such as SCR, SCNR, simultaneous removal, pre-combustion, control combustion and post combustion methods. Despite the advantages of various commercial NOx removal methods, several disadvantages have been mentioned by the researchers. The methods of low-NOx combustion can achieve the maximum removal efficiency of 30%–50%, where post-combustion achieves higher reduction efficiency. SNCR reduction is achieved using ammonia or urea at 850–1100 °C, while the maximum reduction is between 30% and 70% with NH_3 to NOx molar ratio of 1.5–2.5. SCR reduce NOx by 80%–95% at ratio of 0.9–1. However, ammonia tends to form NOx at an unsuitable temperature for both technologies. SCR could reduce NOx emissions from 1000 ppm to lower than 10 ppm using a prototype biofuel combustor. Porous medium (PM) reduced NO by 81.6% at high inlet velocity, where it reduced by 99.98% by increasing the porous material thermal conductivity and decreased N_2O emissions by 71.5% through changing the porosity from 0.5 to 0.9. Adding water to reactants on a MILD ammonia will decrease NOx emissions at all φ values through De-NOx route.

In summary, more efforts should be made to gain a thorough understanding of the impacts of plasma on ammonia combustion and NOx reduction chemical kinetics in order to overcome the limitations of ammonia use in practical applications.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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