



Tandem plasma electrocatalysis: An emerging pathway for sustainable ammonia production

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The Haber-Bosch process, the dominant process for industrial ammonia production, is highly energy-intensive and a major source of carbon emissions. Plasma and electrocatalysis offer viable and promising alternatives for nitrogen reduction reactions, especially when integrated with intermittent renewable electricity. However, relying solely on plasma or electrocatalysis for direct N₂ reduction presents significant challenges. Plasma technology suffers from low conversion efficiency and high energy consumption. Similarly, electrocatalysis encounters challenges with low yield and Faradaic efficiency, primarily due to the low solubility of nitrogen gas and interference of the competing hydrogen evolution reaction. A tandem process combining plasma synthesis of NO_x (a mixture of NO and NO₂) with the electrochemical NO_x reduction reaction (eNO_xRR) can effectively use NO_x as an intermediate, thereby significantly reducing the difficulty of N₂ activation in plasma and enhancing the Faradaic efficiency of the subsequent electrocatalytic process. This promising solution has great potential to dramatically enhance the ammonia synthesis rate, making the tandem process a compelling technology for sustainable and decentralized ammonia synthesis under mild conditions. This review provides an insightful overview of the tandem plasma-electrocatalytic process, illustrating the reported methods for plasma-driven nitrogen activation to nitrogen oxides and discussing the recent advances and challenges in eNO_xRR, with a particularly focus on developing efficient electrocatalysts. Additionally, we discuss the systemic challenges of integrating these two processes, highlighting the importance of process optimization and the potential for ammonia production. The techno-economic and environmental impacts of the tandem process are also evaluated and compared to the Haber-Bosch process, providing insights into future development pathways for this innovative approach.

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Keywords

Plasma, Electrocatalysis, NO_x synthesis, Ammonia synthesis, Nitrate reduction.

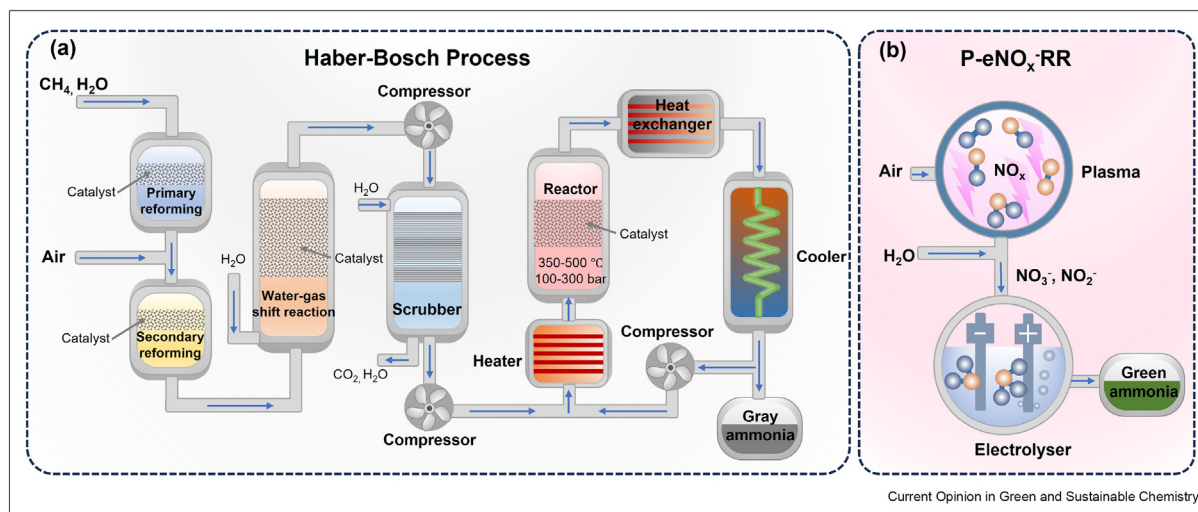
Introduction

Ammonia (NH₃) is a fundamental chemical with extensive applications in agriculture, industry, and emerging energy technologies. The Haber-Bosch (H–B) process, developed in the early 20th century, revolutionized ammonia production, but it operates under harsh conditions, demands large scale and complex infrastructure, and is associated with significant carbon emissions, particularly when the energy is sourced from fossil fuels (Figure 1a). In recent years, there has been growing interest in developing emerging technologies, such as plasma and electrocatalysis, for more sustainable ammonia synthesis under mild conditions [1,2].

However, the strong N≡N triple bond in inert nitrogen molecules results in high reaction energy barriers during nitrogen activation and direct hydrogenation, leading to low synthesis rates in ammonia production. In the plasma ammonia synthesis from N₂ and H₂, enhancing the reaction rate is challenging due to limitations from both N₂ dissociation and the equilibrium constraints of the reverse reaction (NH₃ → N₂ + H₂) [2]. Although the electrochemical nitrogen reduction reaction (eNRR) is promising, it suffers from slow kinetics and low selectivity due to the low solubility of N₂ and competition with the hydrogen evolution reaction (HER) [1,3]. Furthermore, a plasma-activated proton conducting solid oxide electrolyzer system has been proposed for ammonia production from N₂ and water, achieving a Faradaic efficiency surpassing that of eNRR, though with a relatively high energy consumption (605 MJ mol⁻¹) [4].

A tandem process that combines non-thermal plasma (NTP) and electrocatalysis (P-eNO_xRR) has recently emerged as a promising solution for ammonia synthesis (Figure 1b). This innovative process uses NO_x as an intermediate, thereby avoiding the direct reduction of N₂ and achieving significant improvements in both N₂ conversion rates and ammonia production efficiency.

Figure 1



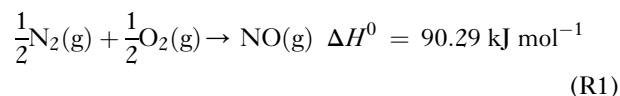
Schematic illustration of ammonia production from (a) the Haber-Bosch (H-B) process, and (b) the Plasma-coupled eNO_x⁻RR tandem process (P-eNO_x⁻RR).

Specifically, in the initial stage, plasma technology is employed to directly activate N₂ and O₂, producing a high concentration of NO_x (a mixture of NO and NO₂). In the subsequent stage, ammonia is directly synthesized through the electrochemical reduction of nitrogen oxides (eNO_x⁻RR). Since NO_x species have lower bond energies (204 kJ mol⁻¹) compared to N₂ (945 kJ mol⁻¹) [5], this enables more efficient conversion to ammonia. Currently, eNO_x⁻RR studies primarily use NO_x sourced from industrial wastewater. However, the concentration of NO_x⁻ in wastewater is typically quite low (<4 mM) [6], and the impurities present in the wastewater can significantly disrupt the eNO_x⁻RR process. To achieve stable and efficient ammonia synthesis via eNO_x⁻RR, a reliable and clean source of NO_x is essential. Plasma technology offers a solution by converting atmospheric nitrogen into NO_x, providing a clean and controllable NO_x⁻ source for subsequent electrochemical reduction. This tandem approach leverages the strengths of both plasma and electrocatalysis technologies, potentially resulting in higher ammonia yields compared to methods that rely on a single technology.

Plasma-driven NO_x production

NO_x formation from N₂ and O₂ is an endothermic process (R1) with a low standard enthalpy of 0.09 MJ mol⁻¹ [7]. However, thermochemical NO_x production, which requires high temperatures, is limited to thermal equilibrium, resulting in high energy consumption [8]. Theoretical calculations have shown that the minimum energy consumption of thermochemical NO_x production is higher than 2 MJ mol⁻¹, more than ten times higher than that for plasma-enabled NO_x production [8,9]. Plasma is a partially ionized gas composed of electrons,

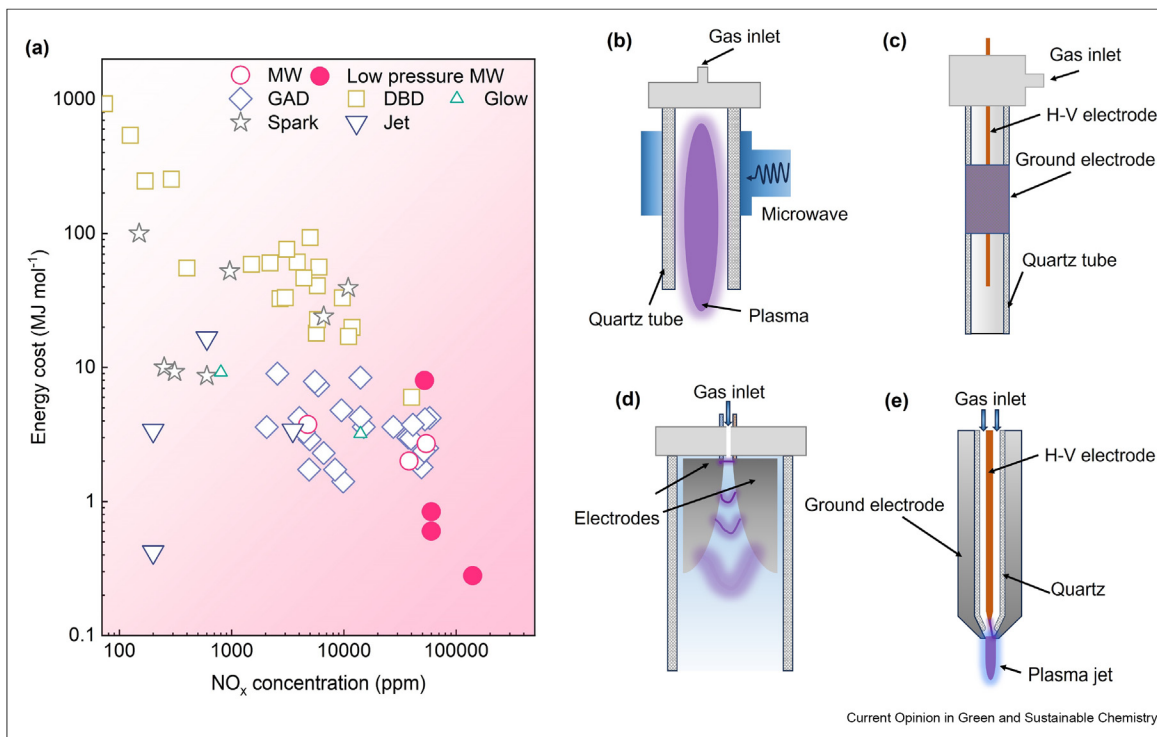
ions, and neutral particles. In NTP, the formation of NO_x involves a complex interplay of reactions. High-energy electrons in the plasma collide with N₂ molecules, generating reactive nitrogen species, such as N atoms and excited N₂. These reactive species can subsequently react with O₂ to produce NO_x at ambient temperatures.



Various types of NTP have been studied for NO_x generation. Figure 2a compares the performance of NO_x synthesis across different plasma reactors, primarily under atmospheric pressure unless otherwise specified. The lowest energy consumption of 0.28 MJ mol⁻¹ and the highest NO_x concentration of 140,000 ppm were achieved using a low-pressure (10–100 Torr) microwave (MW) discharge (Figure 2b) [10]. However, the additional energy required for vacuum pumps at low pressures often negates these efficiency gains. As a result, recent studies on MW discharge for NO_x production have shifted towards (near) atmospheric pressure discharges, with the lowest reported energy cost at atmospheric pressure being 2 MJ mol⁻¹, yielding a total NO_x production of 3.8 % [11].

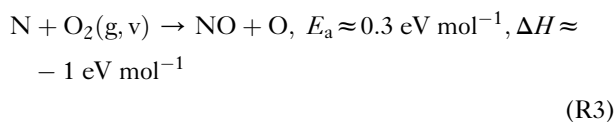
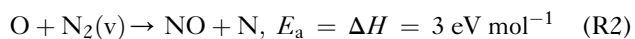
The energy efficiency of NO_x formation is primarily determined by the method used to break the strong N₂ bond (~10 eV) [8,12]. In dielectric barrier discharge (DBD) plasma (Figure 2c), the reduced electric field typically falls within the 100–200 Td range. In this range, the reaction system is mostly governed by direct

Figure 2



(a) Energy cost and concentration of nitric oxides from plasma synthesis reported in the literature; the schematics of (b) MW, (c) DBD, (d) GAD, and (e) Jet plasmas.

electron impact dissociation, which requires large electron energies, resulting in low energy efficiency. In contrast, other plasma discharges, such as gliding arc discharge (GAD), operate with a reduced electric field in the 10–100 Td range, where N₂ vibrational excitation is the predominant route for NO_x production [8,13]. This pathway follows the Zeldovich mechanism ((R2) and (R3)) for N–N bond breaking, which is recognized as the most energy-efficient method for producing NO_x.



GADs benefiting from abundant N₂ vibrational excitation (Figure 2d), have shown promising results compared to other plasma types, particularly in novel reactor configurations. Wang *et al.* [8] demonstrated

that a pulsed GAD outperforms thermal NO_x synthesis in both yield and energy efficiency, achieving an energy cost of 4.8 MJ mol⁻¹. Van Alphen *et al.* [14] further improved gas–discharge interaction by using a rotating gliding arc reactor with an effusion nozzle, achieving a 5.9 % NO_x yield at an energy cost of 2.1 MJ mol⁻¹.

The lowest energy cost for atmospheric pressure plasma was achieved using a pulsed arc-formed jet reactor (Figure 2e) [15]. In this system, a plasma arc is generated between a pin electrode and a metal nozzle, with gas flow driving the formation of the jet. This setup achieved an energy consumption of 0.42 MJ mol⁻¹, approaching the theoretical minimum for non-thermal plasma, which is 0.2 MJ mol⁻¹. However, it is important to note that the energy cost calculation considered only the power used during the pulse, excluding inter-pulse power consumption. Additionally, scaling up pulse power supplies presents a significant challenge.

Coupling catalysts with plasma systems is an effective strategy to boost NO_x yield and reduce energy costs. For instance, Lei *et al.* [16] achieved a 40 % reduction in

energy costs, reaching 1.73 MJ mol^{-1} , by integrating a three-level coupled rotating electrodes air plasma with nano-sized TiO_2 . However, the mechanisms underlying plasma–catalyst interactions in NO_x synthesis remain poorly understood. Key questions include identifying the dominant plasma-activated N_2 species in plasma catalysis and determining the active catalytic sites that enhance NO_x synthesis.

Despite recent advances, the energy cost of plasma-based NO_x synthesis remains significantly above the theoretical minimum. Optimizing reactor configuration to promote effective collisions between gas molecules and high-energy electrons in the discharge space could significantly increase the NO_x synthesis rate. Theoretically, energy efficiency could also be improved by suppressing reverse reactions and minimizing the vibrational–vibrational (VV) relaxation channel. Strategies such as fast quenching or in situ removal of NO (e.g., rapid oxidation to NO_2 by O_3) may effectively suppress the reverse reactions. However, the absence of pilot-scale NO_x synthesis processes limits progress toward practical applications.

In addition to integrating plasma-enabled NO_x production with electrocatalysis (P-e NO_x -RR) for NH_3 synthesis, the direct conversion of NO_x to nitric acid also

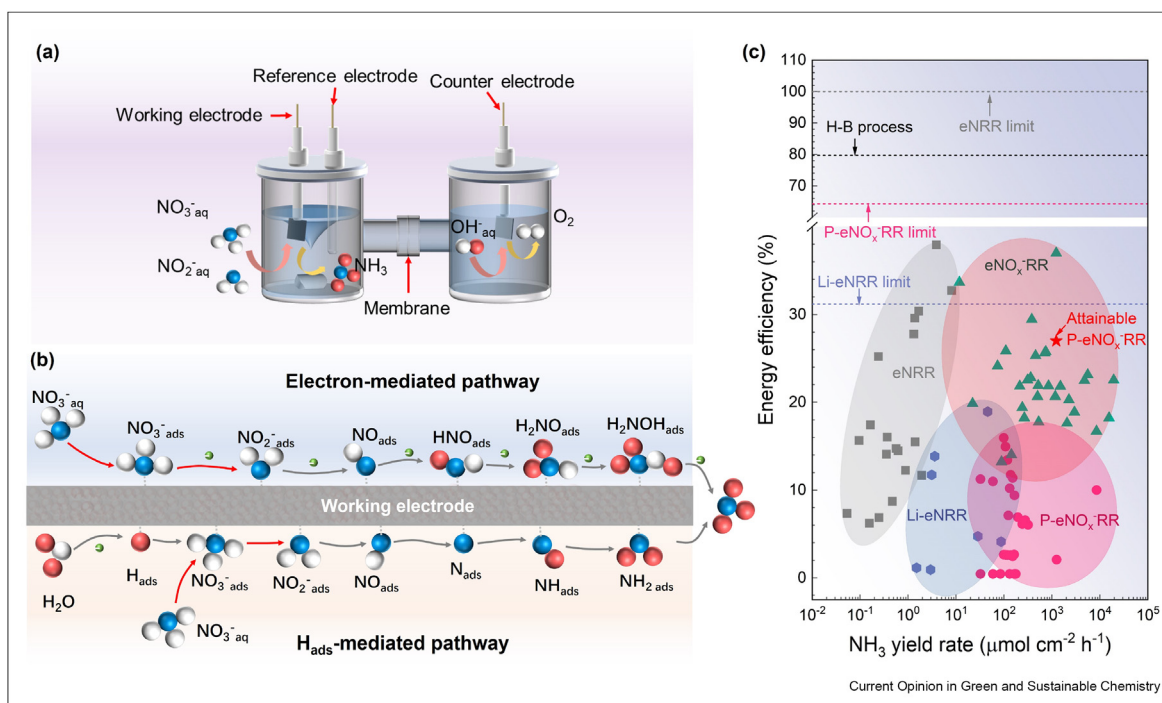
shows considerable potential. Techno-economic analysis suggests that plasma-based NO_x production could compete more effectively with electrolysis-driven Haber-Bosch and Ostwald processes for nitric acid production if energy consumption can be reduced to 1.0 – 1.5 MJ mol^{-1} [17].

Electrochemical NO_x reduction reaction (e NO_x -RR)

Electrocatalytic NO/NO_x -to- NH_3 conversion (eNORR and e NO_x -RR) are promising approaches for ammonia production. However, the yields reported for the eNORR process ($1239 \mu\text{mol cm}^{-2} \text{ h}^{-1}$ [18]) are approximately an order of magnitude lower than those for e NO_x -RR ($19411 \mu\text{mol cm}^{-2} \text{ h}^{-1}$ [19]), attributed to the low solubility of NO in water. Compared to eNORR, the reaction pathways of e NO_x -RR in aqueous solution are more versatile [20], involving proton-coupled electron transfer (Figure 3a). The nitrogen valence state can range from -3 to $+5$ (in e NO_3 -RR) or up to $+3$ (in e NO_2 -RR), enabling the production of multiple compounds, including NO_2^- , NO , N_2O , N_2 , NH_2OH , and NH_4^+ [21].

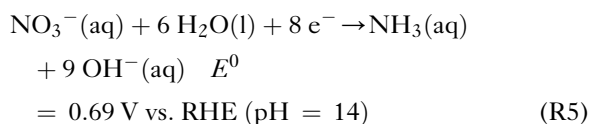
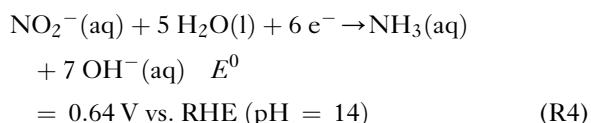
For ammonia production, e NO_x -RR requires the transfer of either eight or six electrons, depending

Figure 3

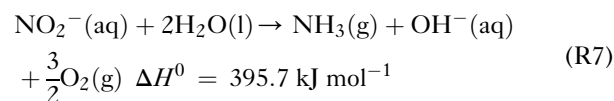
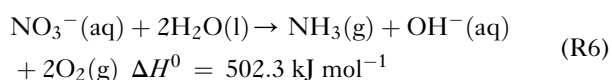


Schematic illustration of (a) e NO_x -RR, and (b) electron-mediated and H_{ads} -mediated pathways of e NO_x -RR. (c) Comparison of eNRR, Li-eNRR, e NO_x -RR, and P-e NO_x -RR in terms of NH_3 yield rate and energy efficiency (Attainable P-e NO_x -RR is estimated as the sum of the best performances of both the plasma-driven NO_x production and the e NO_x -RR).

on the specific NO_x^- species involved ((R4) and (R5)).



Most eNO_x^- RR processes are conducted in alkaline electrolytes, where the overall reactions are described by (R6) and (R7), both of which are endothermic. The theoretical energy consumption for ammonia production via eNO_3^- RR is $0.502 \text{ MJ mol}^{-1}$, while for eNO_2^- RR, it is $0.396 \text{ MJ mol}^{-1}$ [22].



Understanding the mechanism of eNO_x^- RR is crucial for effective catalyst design and optimization. The mechanisms of eNO_x^- RR-enabled NH_3 synthesis involve two primary electrocatalytic pathways: the electron-mediated pathway [23,24] and the H_{ads} -mediated pathway [25]. These mechanisms have been elucidated using in situ electrochemical characterizations and density functional theory (DFT) calculations [26,27]. In the electron-mediated process, NO_3^- adsorbs onto the catalyst surface and is reduced to $\text{NO}_2^-_{\text{ads}}$, followed by the formation of NO_{ads} , a key intermediate that governs the reaction's selectivity. Alternatively, the H_{ads} -mediated pathway involves the reduction of NO_3^- by H_{ads} , which is formed during water electrolysis (Volmer process). This pathway proceeds through intermediates including $\text{NO}_2^-_{\text{ads}}$, NO_{ads} , N_{ads} , NH_{ads} , and NH_2_{ads} , favoring the formation of NH_3 over N_2 (Figure 3b). The H_{ads} -mediated pathway typically operates at low overpotentials, minimizing side

Table 1

Summary of the state-of-the-art eNO_x^- RR performance.

Electrocatalyst	Ammonia production rate ($\mu\text{mol cm}^{-2} \text{h}^{-1}$)	Faradaic efficiency (%)	Electrolyte	Potential (V) vs. RHE	Ref.
MWCNTs	264.57	90	0.1 M PB + 0.4 M KNO_3	-0.85	[33]
CoP/TiO ₂ nanoarray	499.8	87.5	0.1 M NaOH + 0.1 M NO_3^-	-0.5	[34]
BiFeO ₃ flakes	212.82	96.85	0.1 M KOH + 0.1 M KNO_3	-0.6	[35]
Ru@Cu NWs	707.33	93	0.1 M Na ₂ SO ₄ + 0.1 M NO_3^-	-0.2	[36]
vCo-Co ₃ O ₄ /CC	517.5	97.2	0.1 M NaOH + 0.1 M NaNO ₃	-0.6	[37]
Cobalt-doped Fe@Fe ₂ O ₃	88.58	68	0.1 M Na ₂ SO ₄ + 500 ppm NO_3^-	-0.95	[38]
FeOOH/CP	142.29	68	0.1 M (PBS) + 0.1 M NaNO ₃	-0.8	[39]
MoO ₂ -C NBF	109.28	99.05	1 M KOH + 0.1 M KNO_3	-0.3	[40]
Cu/Cu ₂ O	240	95.8	0.5 M Na ₂ SO ₄ + 200 ppm NO_3^-	-0.85	[41]
Strained Ru Nanoclusters	1170	~100	1 M KOH + 1 M KNO_3	-0.8	[42]
Fe-N/P-C	528.82	86	0.1 M KOH + 0.1 M KNO_3	-0.8	[43]
Ru ₁₅ Co ₈₅ HNDs	1230	97	0.1 M KOH + 0.1 M KNO_3	0.3	[32]
0.6 W-O-CoP@NF	4760	95.2	1 M KOH + 0.1 M NO_3^-	-0.5	[44]
Pd-Co ₃ O ₄ /TM	745.6	98.7	0.1 M KOH + 0.1 M NO_3^-	-0.3	[45]
CuNi NPs/CF	5563	97.03	1 M NaOH + 0.715 M NO_3^-	-0.48	[46]
Ni nanoparticles	15490	93	1 M NaOH + 1 M NaNO ₃	-0.93	[3]
Ru ₁ Cu ₁₀ /rGO	380	98	0.1 M KNO_3 + 1 M KOH	-0.05	[47]
Mn-incorporated Co ₃ O ₄ nanotubes	2058.8	99.5	0.5 M K ₂ SO ₄ + 0.1 M KNO_3	-1.2	[48]
D-Ni(OH) ₂ /Ni@CF	2941.2	99	1 M KOH + 0.1 M KNO_3	-1	[49]
FeB ₂	1500	96.8	1 M KOH + 0.1 M NO_3^-	-0.6	[50]
self-supported carbon nanosheet array	8470	88.6	1 M OH ⁻ + 1 M NO_3^-	-1.03	[51]
Co-CNF/ZIF-CoP	2260	90	1 M KOH + 0.1 M KNO_3	-0.6	[52]
Cu/Cu ₂ O/Pi NWs	360	96.6	1 M KOH + 0.1 M KNO_3	-0.5	[53]
Pd-Cl/Cu ₂ O	19411	~100	1 M KOH and 1 M NO_3^-	-0.6	[19]
CoP@TiO ₂ /TP	849.57	97.01	0.1 M NaOH + 0.1 M NO_2^-	-0.6	[5]

reactions. In both pathways, nitrate adsorption and nitrite generation are considered the rate-determining steps.

Strategies for improving NO_x^- RR performance

Developing efficient electrocatalysts is essential for enhancing NO_x^- RR performance in NH_3 production. Ideal catalysts should offer high activity, selectivity, stability, and operate at low overpotential. Various materials and catalyst engineering strategies have been employed to improve eNO_x^- RR performance [20]. Beyond catalyst innovation, optimizing membranes and electrolytes (e.g., pH, composition, concentration) can further enhance performance [28,29]. Although an acidic condition provides a high availability of protons for eNO_x^- RR, most of the state-of-the-art results (summarized in Table 1) are achieved in a strong alkaline electrolyte with high NO_x^- concentrations (>100 mM). Conducting eNO_x^- RR in acidic conditions presents significant challenges, including catalyst corrosion, competition with the HER, and mass transport limitations.

Recent advancements have achieved ammonia yield rates as high as $19,411 \mu\text{mol cm}^{-2} \text{h}^{-1}$ with current densities exceeding 2 A cm^{-2} [19]. Faradaic efficiencies above 90 % have been widely reported, but optimal ammonia production in most studies requires potentials far from the theoretical eNO_x^- RR potential (0.69/0.64 V for eNO_3^- RR/ eNO_2^- RR at pH = 14 [30,31]). The energy cost of eNO_x^- RR is directly related to the cell voltage ($U_{\text{cell}} = E_{\text{cathode}} - E_{\text{anode}}$, U refers to the cell voltage; E refers to the electrode potential.), making it crucial to reduce overpotentials in both eNO_x^- RR and the oxygen evolution reaction (OER) to improve energy efficiency. Recently, Han et al. [32], reported employing $\text{Ru}_{15}\text{Co}_{85}$, achieving an onset potential of +0.4 V vs. RHE, which is close to the theoretical onset potential of eNO_3^- RR. In their study, they achieved an optimal energy efficiency of 42 %, corresponding to an energy consumption of $\sim 1.00 \text{ MJ mol}^{-1}$, along with an ammonia production rate of $1230 \mu\text{mol cm}^{-2} \text{h}^{-1}$ at a potential of 0.3 V vs. RHE.

Integration of plasma-based NO_x generation and electrocatalysis

Integrating plasma-based NO_x generation with electrochemical NO_x^- reduction could offer a sustainable approach to ammonia synthesis by utilizing low-cost feedstocks such as air and water. Moreover, this combined process has the potential to achieve higher ammonia yields compared to electrocatalysis alone due to a continuous replenishment of NO_x^- .

Maximizing ammonia yields and energy efficiency in a tandem P- eNO_x^- RR system requires careful

consideration of plasma reactor configuration, electrocatalyst selection, and optimization of operating parameters. Sun et al. [54] achieved an ammonia yield of 23.2 mg h^{-1} using a system combining an NTP bubble column reactor and a scalable electrolyzer with Cu nanowire electrocatalysts in acidic conditions. The energy consumption was $14.78 \text{ MJ mol}^{-1}$, with NO_x^- production accounting for 13.7 MJ mol^{-1} and electrolysis for 1.08 MJ mol^{-1} , resulting in an overall energy efficiency of 2.59 %.

The energy consumption of ammonia in the P- eNO_x^- RR system heavily depends on the NO_x^- production. Studies [55,56] employing P- eNO_x^- RR systems incorporating DBD reactor or spark discharge are expected to suffer from high energy consumption, considering the poor performance (generally $>10 \text{ MJ mol}^{-1} \text{NO}_x$) of these discharges for gaseous NO_x production (Figure 2). The work by Liu et al. [57] confirmed this, with their P- eNO_x^- RR system achieving a high energy consumption of $18.36 \text{ MJ mol}^{-1} \text{NH}_3$ (2.08 % in terms of energy efficiency), despite the superior performance of the $\text{Ni}(\text{OH})_2/\text{Cu}$ (e.i., high activity with ammonia yield rate of $\sim 1250 \mu\text{mol cm}^{-2} \text{h}^{-1}$ and low overpotential with $E_{\text{cathode}} = -0.25 \text{ V vs. RHE}$).

Noted that although GAD and its variants are efficient for NO_x production, P- eNO_x^- RR systems employing these discharges could also result in high energy consumption due to inefficient NO_x trapping to NO_x^- . For example, Wu et al. [58] used a rotating GAD above a liquid surface to produce aqueous NO_x^- from generated NO_x , but only 2.9 % conversion was achieved due to high NO selectivity in NO_x , resulting in a high energy cost of $83.44 \text{ MJ mol}^{-1}$ for NO_x^- production, inevitably leading to ultra-low energy efficiency (0.45 %) of their P- eNO_x^- RR system. In another study combining GAD with eNO_x^- RR, the energy consumption for NO_x^- production was reduced to 4.38 MJ mol^{-1} by bubbling the generated NO_x into a 0.1 M KOH solution [59]. Their tandem P- eNO_x^- RR system achieved an ammonia production rate of 7.3 mg h^{-1} ($429.41 \mu\text{mol cm}^{-2} \text{h}^{-1}$) at a record low energy consumption of 6.33 MJ mol^{-1} (6.05 % in terms of energy efficiency) using plasma-engraved N-doped defective MoS_2 as the electrocatalyst.

However, in all reported P- eNO_x^- RR systems, the required energy consumption for NO_x^- generation is significantly elevated due to the low efficiency of trapping gaseous NO_x , which underscores a high demand for the development of more effective technologies to enhance the transformation of NO_x from gas phase to liquid phase. Enhancing gas-liquid interactions and increasing solubility can be achieved by optimizing reactor design and reaction conditions, such as pressurizing. Increasing the relative proportion of NO_2 can also improve solubility, as NO_2 has a higher solubility in water

(Henry's law constant = $1.2 \times 10^{-4} \text{ mol}^{-1} \text{ m}^{-3} \text{ Pa}^{-1}$) compared to NO ($1.9 \times 10^{-5} \text{ mol}^{-1} \text{ m}^{-3} \text{ Pa}^{-1}$).

To facilitate the industrial implementation of P-eNO_x⁻RR, it is crucial to increase ammonia yield rates at low overpotential. This necessitates the use of active electrocatalysts and optimized electrolyzer designs. Recently, Guo et al. [60] achieved a record ammonia yield rate of 2578 mg h⁻¹ by integrating La_{1.5}Sr_{0.5}Ni_{0.5}Fe_{0.5}O₄ perovskite oxide into a series-connected stacked membrane electrode assembly (MEA) for acidic (pH = 0) eNO_x⁻RR. This catalyst enables direct ammonium salt production and facilitates integration with plasma-generated NO_x⁻. However, operating the MEA at a high current (20 A) resulted in a significant overpotential ($U_{\text{cell}} \approx 7.7 \text{ V}$) and an energy consumption of $\sim 4.5 \text{ MJ mol}^{-1}$ from eNO_x⁻RR. Future efforts should focus on reducing this overpotential by enhancing electrolyzer design to improve ion conductivity and reduce resistance.

Figure 3c highlights that eNO_x⁻RR outperforms Li-mediated eNRR (Li-eNRR) and eNRR in terms of ammonia production rate. However, the energy efficiency of the state-of-the-art P-eNO_x⁻RR-based ammonia synthesis remains significantly lower than both the theoretical P-eNO_x⁻RR limit (64.3 %) and the H-B process (79.8 %) [22], in which the most energy consumption comes from H₂ production [7]. By combining the best NO_x production results [15] with the state-of-the-art eNO_x⁻RR performance [32], the P-eNO_x⁻RR system is expected to achieve an energy efficiency of 27 % with an ammonia production rate exceeding 1000 μmol cm⁻² h⁻¹.

Conclusion and outlook

The tandem plasma-electrocatalysis approach for ammonia synthesis offers a promising solution to the challenges associated with traditional ammonia production methods. While this technology has the potential to contribute to a more sustainable and distributed ammonia production landscape, significant research is still needed to optimize system performance and reduce costs. Currently, the majority of energy consumption in P-eNO_x⁻RR comes from NO_x⁻ production ($>10 \text{ MJ mol}^{-1}$) rather than from the electrolysis stage ($\sim 1.5 \text{ MJ mol}^{-1}$). This indicates that reducing the energy consumption associated with NO_x⁻ production is crucial for making this method more viable.

Future research should prioritize reducing the energy consumption of gaseous NO_x production. This can be achieved by optimizing reactor design to enable efficient discharge of feed gases, along with fast quenching and in situ NO removal to suppress reverse reactions. Additionally, developing high-efficiency plasma-liquid systems to capture NO_x as aqueous NO_x⁻ is essential.

Research on stable electrocatalysts for eNO_x⁻RR at high current densities remains limited, so further efforts are needed to enhance electrocatalyst activity and reduce overpotential by improving electrolyzer design, which would increase ion conductivity and reduce resistance. Scaling up experiments and performing techno-economic and life cycle assessments are necessary to evaluate the commercial viability and environmental impact of this tandem process. By integrating the strengths of plasma and electrocatalysis, this approach can pave the way for a greener and more sustainable ammonia production industry.

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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Data availability

Data will be made available on request.

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Papers of particular interest, published within the period of review, have been highlighted as:

- * of special interest
- ** of outstanding interest

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