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Review

Recovered Ammonia as a Sustainable Energy Carrier: Innovations in Recovery, Combustion, and Fuel Cells

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Abstract: Recovered ammonia, extracted from waste streams such as industrial leachates and organic waste, represents a unique opportunity to harness a sustainable, carbon-free energy resource. This paper focuses on the energy potential of ammonia recovered from waste, emphasizing its role as a critical element in the transition to a low-carbon economy. Integrating recovered ammonia into energy systems enables industries to reduce dependence on conventional ammonia production, lower greenhouse gas emissions, and advance circular economy practices. The study reviews advanced technologies for recovering ammonia from waste, as well as its application in combustion processes and fuel cells. Particular emphasis is placed on optimizing ammonia combustion to minimize nitrogen oxide (NOx) emissions and on utilizing recovered ammonia in direct ammonia fuel cells and hydrogen generation for fuel cells. Challenges associated with scaling waste recovery technologies and integrating recovered ammonia into existing energy infrastructures are critically examined. By providing an in-depth assessment of the environmental and economic benefits of using recovered ammonia as an energy source, this paper highlights its potential to decarbonize sectors such as transportation, industry, and power generation.

Keywords: ammonia recovery; ammonia to energy; waste to energy; wastewater plants; ammonia fuel cells; ammonia combustion; ammonia fuel

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1. Introduction

Ammonia (NH_3) is a fundamental chemical compound extensively used in agriculture, industry, and energy applications. Globally, over 180 million tons of ammonia are produced annually, primarily for fertilizers that support food production for an expanding population [1]. Despite its widespread utility, ammonia emissions pose significant challenges to both the environment and human health. Unregulated emissions from sources such as intensive livestock farming, landfills, and industrial processes contribute to widespread environmental degradation and health risks, necessitating a critical balance between its industrial value and environmental impact [2,3].

From an environmental perspective, ammonia is a major contributor to the formation of fine particulate matter (PM2.5), a pollutant linked to poor air quality and numerous adverse effects on ecosystems. Ammonia deposition, occurring through both dry and wet processes [3], introduces excessive nitrogen into terrestrial and aquatic environments. In water bodies, this triggers eutrophication, leading to harmful algal blooms that deplete oxygen levels, disrupt aquatic ecosystems, and reduce biodiversity. On land, nutrient overload caused by ammonia deposition impairs soil quality, hinders plant growth, and decreases agricultural productivity. These imbalances persist over time, threatening the long-term sustainability of ecosystems [3–5].

The health risks associated with ammonia emissions are equally concerning. Ammonia acts as an irritant, particularly affecting the respiratory system, eyes, and skin. Acute exposure can cause immediate symptoms such as coughing, shortness of breath, and eye irritation, while chronic exposure is linked to bronchitis, asthma exacerbation, and lung tissue damage [6,7]. Populations living near high-emission sources, including livestock farms and landfills, face increased risks of respiratory illnesses. Vulnerable groups, such as children, the elderly, and individuals with pre-existing conditions, are disproportionately affected [6].

In landfills, ammonia is generated during the anaerobic decomposition of organic waste and is particularly concentrated in leachate. Similarly, in intensive livestock farming, ammonia is released during the decomposition of manure and urine, contributing to local pollution and odor issues that affect surrounding communities. Ammonia concentrations in waste streams, such as landfill leachate and agricultural digestates, often exceed 1000 mg/L, necessitating advanced treatment technologies for safe removal and resource recovery [5,8–10].

Efforts to mitigate these impacts have led to the implementation of regulatory frameworks in many countries. In Europe, the National Emission Ceilings (NEC, 2016/2284/EU) Directive aims to reduce ammonia emissions from the agriculture and industry, targeting gaseous pollutants to improve air quality and protect human health [11]. The Industrial Emissions Directive (IED) complements this by regulating ammonia emissions in liquid effluents from industrial plants to safeguard water quality and environmental integrity [12]. These measures underscore the importance of addressing ammonia emissions comprehensively, recognizing their multifaceted implications for ecosystems and public health [4,5].

While the regulatory focus has primarily been on mitigating the adverse effects of ammonia emissions, an expanding field of research highlights its potential as a resource rather than merely a pollutant [13,14].

The concept of Power-to-Ammonia (P2A) represents a key strategy for integrating renewable energy into sustainable energy systems by leveraging ammonia as a multifunctional energy carrier. Recovered ammonia, in particular, offers significant advantages over conventionally produced ammonia via the Haber–Bosch process, including reduced CO_2 emissions and the reuse of waste resources. Recent studies have demonstrated that P2A systems can balance the intermittency of renewable energy sources, provide long-term energy storage, and contribute to the decarbonization of the industrial and transportation sectors [15].

A relevant practical example is the integration of recovered ammonia into solid oxide fuel cells (SOFCs) at wastewater treatment plants, where ammonia is converted into electricity with an efficiency of 48%, reducing greenhouse gas emissions by 3.5 kg of CO_2 equivalent per person per year [16,17]. Moreover, recovered ammonia has the potential to be used as a direct fuel or as a hydrogen source for energy and industrial applications, further enhancing its role in the circular economy [18].

However, the large-scale implementation of P2A technologies requires addressing certain challenges, such as achieving high ammonia purity to avoid catalytic poisoning and the high initial costs of infrastructure [17,19]. Despite these challenges, P2A represents a promising solution to maximize the use of renewable resources and accelerate the transition towards a low-carbon economy.

Ammonia also represents a transformative opportunity as a renewable energy carrier and fuel. The concept of Power-to-Ammonia (P2A) has emerged as a promising solution for converting surplus renewable electricity into ammonia, providing a stable and easily transportable medium for energy storage. Ammonia's high energy density (22.5 MJ/kg) and established global distribution infrastructure make it an attractive alternative to traditional energy carriers. Unlike hydrogen, which requires complex cryogenic or high-pressure storage systems, ammonia can be stored and transported efficiently, reducing logistical challenges [15,20].

Ammonia's role as an energy carrier extends to its ability to facilitate the transition to a hydrogen-based economy. Through catalytic processes, ammonia can be decomposed into hydrogen and nitrogen, offering a cost-effective and scalable pathway for hydrogen production. This has significant implications for energy systems, as ammonia-powered technologies—such as fuel cells, gas turbines, and internal combustion engines—are being developed to support decarbonization efforts [20].

Ammonia is also gaining recognition as a direct fuel for various applications. Its use in gas turbines and internal combustion engines demonstrates the potential to generate power with minimal carbon emissions, provided that nitrogen oxide (NO_x) emissions are adequately managed through advanced combustion and catalytic control technologies [21]. Although these emission control technologies may increase operating costs, ongoing research—particularly focusing on pre-combustion and combustion techniques—is already showing promising results in terms of technical and economic viability. Studies have indeed demonstrated that blending ammonia with small amounts of hydrogen (up to 10% wt) can result in NO_x emissions in conventional internal combustion engines comparable to those of diesel engines [22]. In Japan, ammonia is being co-combusted with methane in gas turbines under the SIP program to reduce greenhouse gas emissions while maintaining high efficiency [23]. Similarly, in the United States, the REFUEL program is advancing ammonia-based technologies for energy storage and transport, with applications spanning stationary power and transportation sectors [24]. Ammonia-powered maritime engines are also being explored to meet the International Maritime Organization's emission reduction targets, leveraging its high energy density and existing infrastructure for production, storage, and transport [5,20]. Although hydrocarbon fuels exhibit higher energy densities, ammonia-powered fuel cells offer superior efficiency, enabling direct ammonia fuel cells to achieve performance levels comparable to liquid propane gas (LPG)-powered internal combustion engines (ICEs) [25]. In contrast, alternative low-carbon energy vectors, such as lithium-ion batteries and liquid-to-gas expansion systems, exhibit significantly lower energy densities compared to all chemical storage solutions. Their applicability is more contingent upon the specific energy requirements of the application or journey [15]. Ammonia is particularly advantageous for transportation systems that demand substantial energy over prolonged durations, where battery-based solutions or direct electrical connectivity may be impractical or economically unfeasible. This includes applications in heavy-goods vehicles, rail transport, aviation, and shipping. For example, MAN Energy Solutions' demonstration program, which involves retrofitting existing liquefied natural gas (LNG) marine engines to operate on ammonia, provides a cost-effective pathway to decarbonize large-scale maritime transportation [26]. Furthermore, ammonia-based direct solid-oxide fuel cells (SOFCs) offer a high-efficiency solution for both transportation and future power generation. Progress in solid-oxide fuel cell technology, such as developments from the NASA Glenn Research Center, has resulted in high specific and volumetric power densities of up to 2.5 kW/kg and 7.5 kW/L, respectively [27]. These advancements enable the powering of unmanned aerial vehicles (UAVs) and present significant potential to reduce carbon emissions in the aviation sector.

In renewable energy grids, ammonia plays a pivotal role in grid stabilization by serving as a storage medium for excess energy during periods of surplus production, which can be released during peak demand periods. Projects in Northern Europe have demonstrated the successful integration of Power-to-Ammonia (P2A) systems, highlighting ammonia's dual function as both a renewable feedstock for fertilizers and an energy balancing agent within renewable-dominant grids. This capacity to address the intermittency of renewable energy sources, such as solar and wind, makes ammonia an indispensable component in the transition to sustainable energy systems [20].

Given ammonia's dual nature as both an environmental concern and a renewable energy solution, its management necessitates the development of innovative technologies. Catalytic cracking and advanced separation methods for ammonia removal and valorization are essential for mitigating its environmental impact while maximizing its potential as a critical element in sustainable energy infrastructure. This paper delves into these aspects, examining ammonia's environmental and health implications, state-of-the-art removal technologies, its application as a fuel, and its transformative role in facilitating the transition to renewable energy (Figure 1). By addressing both the risks and opportunities associated with ammonia, we aim to provide a comprehensive understanding of its potential to contribute to global sustainability.

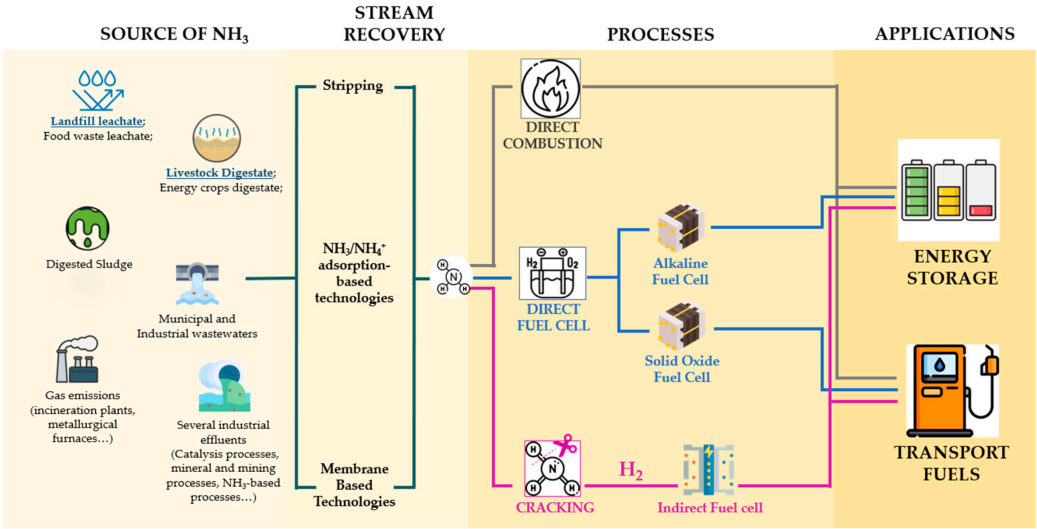


Figure 1. Graphical representation of ammonia sources (**left**), recovery processes such as stripping, NH_3/NH_4^+ adsorption and membrane-based technologies, and potential applications for recovered ammonia (**right**). The applications are categorized based on their end-use objectives, including energy storage and the production of transport fuels.

2. Main Sources of Ammonia Recovery from Waste

A wide range of industrial and agricultural processes yield nitrogen-rich byproducts that present valuable opportunities for ammonia recovery (Figure 1). These byproducts, spanning both liquid and gaseous forms, arise in substantial quantities across multiple sectors, each characterized by distinct feedstocks, operational conditions, and process outputs (Table 1). Their nitrogen composition and ammonia content vary significantly, reflecting differences in raw materials, production techniques, and end-product requirements. For instance, the nitrogen load in industrial wastewater effluents from fertilizer manufacturing may differ markedly from that in landfill leachate or from ammonia-laden gases released in metallurgical refining [28].

Table 1. Summary table of key characteristics of ammonia-containing waste streams.

Sector (Industrial Activity)	Waste Material (Flow) Containing Ammonia	N-NH_3 (mg/L)	pH	COD (mg/L)	Ref.
Landfills (waste management)	Leachate	<30–>4000	<6.5–9	<1000–70,000	[3,4,13,29–41]
Biogas production	Livestock digestate liquid phase	834–3795	5.9–9.1	3814–46,300	[42–56]
Wastewater treatment	Municipal sewage wastewater	10–68	6.5–7.5	95–506	[57]
Wastewater treatment	Digested sludge	1510–1679	7.4–7.8	1684–2398	[58]
Agriculture	Energy crops digestate	1300–2400	7.5–8.4		[51]
Animal byproduct processing	Slaughterhouse wastewater	100–140		1100–1400	[59]
Food waste	Leachate from food waste	30–140	4.3	1100–199,500	[60–62]
Chemical industry	Spent catalysts, process effluents	177	11.6	188	[63]
Fertilizer production	Process effluents	50–200	6.0–9.0	500–1500	[28]
Mining and mineral processing	Ammonium-laden effluents from ore leaching	20–80	6.0–8.5	-	[64,65]
Plastic and rubber production	Wastewater containing ammonia from polymerization	5–300	3.7–9.4	2834–26,914	[66]
Textile industry	Dye bath effluents, process wastewater	34–49	6.3–13.1	635–4459	[67,68]
Tanning and leather industry	Effluents from ammonia-based deliming processes	123–150	3.3–12.4	1670–11,413	[69–71]
Pharmaceuticals	Fermentation by-products, process effluents	5000–6000	8.0–12.0	500–3000	[72,73]
Pulp and paper	Black liquor, bleaching effluents	42–200	6.3–9.0	1195–20,000	[74–76]
Petroleum refining	Ammonia-rich sour water from refining processes	20–335	4.3–10	1200–3134	[77–79]
Energy production (coal-fired power plants)	Flue gas desulfurization (FGD) wastewater	1–10	6.0–9.0	50–500	[80]
Steel and metallurgical sector (rare)	Ammonia-containing wastewater	50–200	8.5–9.5	200–6500	[81–83]
Gas emission:	Off-gas from incineration plants, metallurgical furnaces, etc.	Gas emissions 6–8 g/Nm^3 (raw coke oven gas); 10–40 mg/Nm^3 (BF off-gas) Note: For each tonne of coke produced, approximately 3 kg of ammonia is generated.			[83]

Table 1 provides a comprehensive overview of ammonia-containing wastes from diverse sectors, documenting key parameters such as ammoniacal nitrogen (N-NH_3) concentration, pH, and Chemical Oxygen Demand (COD). By cataloging a broad spectrum of ammonia sources, Table 1 assists in pinpointing the most promising targets for ammonia reclamation and informs decision-making on technology selection, process design, and regulatory compliance.

Among the eighteen categories of ammonia-bearing wastes detailed in Table 1, seventeen focus on liquid streams. Landfill leachate exemplifies a particularly challenging yet significant source due to its elevated ammonia levels, variable composition, and substantial environmental impact if left untreated [29]. Generated by the breakdown of organic matter under anaerobic conditions in landfills, leachate accumulates ammonia and other contaminants, making it a priority candidate for recovery efforts. Similarly, biogas digestates—resulting from the anaerobic digestion of agricultural residues, animal byproducts, and organic waste—are increasingly abundant as biogas production scales up. These digestates also possess considerable ammonia concentrations and nutrient loads, rendering them prime targets for recovery processes that not only reduce pollution but also recycle valuable nitrogen resources [42].

In contrast, only one category—ammonia removal from gaseous emissions in the metallurgical sector—pertains primarily to gas-phase sources. Here, ammonia may emerge as a byproduct of ore processing and metal refinement, often under high-temperature conditions. Air scrubbing and related gas-cleaning technologies can capture and concentrate this ammonia, preventing its atmospheric release [83]. Though these gaseous streams are less common, they demonstrate the versatility of ammonia recovery solutions, extending beyond liquid effluents to encompass a broader range of industrial scenarios.

The relevance and potential of each ammonia source depend on multiple factors, notably the ammonia concentration, treatment feasibility, and the associated environmental implications. Higher ammonia concentrations generally improve economic feasibility; when the nitrogen load is substantial, less energy and fewer materials are required per unit of ammonia recovered [84]. For example, streams with extremely high ammonia content, such as landfill leachate or concentrated digestates, allow for more cost-effective reclamation due to a more favorable energy and resource balance. Conversely, lower-ammonia streams (e.g., certain mining and textile industry effluents) may necessitate more complex, multi-stage treatment systems or novel technologies to achieve economically viable recovery, potentially involving advanced adsorbents, specialized membranes, or energy-intensive stripping processes [52,84].

Treatment feasibility is also influenced by the availability and maturity of suitable technologies. Established methods, like ammonia stripping or conventional adsorption, may prove adequate for moderately concentrated streams with relatively stable compositions. However, more challenging effluents—those with either exceptionally high ammonia concentrations or complex matrices containing organic contaminants, suspended solids, and variable pH—may require cutting-edge approaches, including advanced nanofiltration membranes, novel adsorbent materials (e.g., functionalized biochar or metal–organic frameworks), or integrated treatment systems that combine physical, chemical, and biological steps [84–87]. The scale of the waste stream further shapes technological choices: large, continuous flows (such as those from major food-processing plants or centralized municipal wastewater treatment facilities) may justify substantial capital investments in robust, highly efficient processes, whereas smaller or more intermittent streams may favor modular or simpler solutions that minimize operational complexity.

Environmental considerations and regulatory frameworks play a pivotal role as well. Stricter discharge limits for nitrogen compounds push industries toward more rigorous ammonia recovery measures to comply with water quality standards and air emission regulations. Likewise, the drive toward sustainability and the circular economy prompts the extraction of ammonia from waste not merely as a pollution control tactic, but also as a resource recovery strategy, transforming what would otherwise be an environmental liability into a valuable input for fertilizer production or other nitrogen-dependent processes. In this context, ammonia recovery supports nutrient recycling, reduces dependence on synthetic fertilizers manufactured from fossil-based feedstocks, and curtails the environmental footprint of waste management practices.

As industries and governments increasingly prioritize sustainability, understanding these interrelated factors—ammonia concentration, treatment feasibility, technological readiness, economic considerations, regulatory constraints, and environmental objectives—is crucial. Such a comprehensive perspective ensures the selection of the most efficient, cost-effective, and environmentally responsible ammonia recovery methods, thereby advancing both ecological integrity and resource efficiency.

To further refine the classification of liquid ammonia-rich wastes, the streams in Table 1 can be categorized by their N-NH_3 concentrations:

(A) Concentration of ammoniacal nitrogen (N-NH_3) < 100 mg/L

Waste streams in this category contain relatively low ammonia concentrations. Although such dilute conditions may initially seem less economically attractive for ammonia recovery, the presence of large volumes or stringent regulatory requirements can still justify reclamation efforts. Typical examples include effluents from mining and mineral processing (where ore leaching may release small amounts of ammoniacal nitrogen), textile industry dye baths, and flue gas desulfurization wastewater from coal-fired power plants [68,82]. Additionally, certain gas emissions from energy production and steelmaking, such as those originating from coke ovens and blast furnaces, also fit into this group. While the ammonia load is relatively low, the cumulative effect of large quantities and the push for stricter environmental compliance can make ammonia recovery both feasible and beneficial.

(B) Concentrations of N-NH_3 between 100 and 500 mg/L

Wastewaters in this range have moderate ammonia concentrations and often arise from more chemically intensive processes. Examples include process effluents in fertilizer and plastics production, where ammonia is generated as a polymerization byproduct, as well as waste streams from tanning and leather operations, pulp and paper bleaching, petroleum refining, and certain steelmaking processes [28,66,74]. Compared to the lower-ammonia category, these intermediate-level streams present enhanced recovery opportunities due to their increased ammonia content.

(C) Concentrations of N-NH_3 between 500 and 2000 mg/L

At this concentration range, waste streams are considered significantly ammonia-rich and become prime candidates for ammonia recovery. One prominent example is sludge produced during wastewater treatment, where nitrogen compounds, including ammonia, accumulate as part of the purification process. Recovering ammonia from such high-strength sources proves economically viable and can provide substantial environmental benefits. By lowering the nitrogen load in effluent discharged into waterways, the risk of eutrophication is reduced. Moreover, ammonia recovery as a valuable nutrient potentially minimizes the dependence on synthetic fertilizers and contributes to more sustainable resource management [87].

(D) Concentrations of N-NH_3 > 2000 mg/L

This group encompasses the most ammonia-rich waste streams, offering both significant challenges and substantial opportunities for ammonia reclamation. Notable examples include landfill leachates, which can reach extremely high ammonia levels as organic waste decomposes over time, as well as biogas digestates derived from agricultural residues and animal byproducts, and effluents from pharmaceutical fermentation processes. The complexity and variability of these streams, combined with increasingly strict regulatory oversight, may necessitate advanced treatment technologies and multifaceted approaches [29,42]. However, the potential rewards are considerable: effective ammonia recovery from highly concentrated sources can greatly reduce pollution, ensure compliance with rigorous environmental standards, and transform hazardous wastes into valuable nitrogen resources suitable for fertilizer production or other industrial applications. In doing so, ammonia recovery at this level aligns closely with the principles of the circular economy and sustainable development.

Within Group D, landfill leachates and biogas digestates are considered especially promising for large-scale ammonia recovery. Although pharmaceutical effluents may reach even higher concentrations (5000–6000 mg/L), their overall volumes are typically lower, diminishing their relative impact on large-scale nitrogen recycling efforts. By contrast, biogas digestates derive from abundant feedstocks and align closely with emerging renewable energy and nutrient recycling initiatives, making them critical focal points for both innovation and investment. Furthermore, stringent waste management regulations [10,11] and the recognized environmental hazards posed by uncontrolled disposal reinforce the strategic importance of tackling these high-ammonia wastes.

2.1. Landfill Leachate

Landfill leachate represents a particularly challenging and environmentally significant ammonia-rich waste stream. Formed by the percolation of water through landfilled waste and the subsequent leaching of dissolved organic and inorganic constituents, leachate composition is governed by a complex interplay of biological and physicochemical processes [5]. A defining attribute of landfill leachate is its elevated ammonia content, which arises primarily from the microbial breakdown (ammonification) of nitrogen-rich organic substrates, such as food residues and other biodegradable materials [41].

The concentration of ammonia in leachate varies considerably, reflecting differences in waste composition, landfill age, and site-specific operational conditions. Younger landfills often exhibit higher ammonia concentrations due to the rapid decomposition of fresh organic waste, whereas older landfills, having undergone more extensive degradation and stabilization, tend to display comparatively lower ammonia levels [31].

Among the flows in group D, which represent the richest sources of ammonia, residues from waste treatment activities (including landfill leachates and biogas digestates), are considered more promising than industrial wastes from other sectors such as pharmaceutical process effluents and agricultural and animal byproducts. On the other hand, animal and agricultural residues are often processed in biogas plants, which makes biogas digestates a more substantial source of ammonia, also encompassing agricultural and animal byproducts [54].

Moreover, biogas digestate and landfill leachate are classified as waste which need to be treated under strict rules, and their improper management can pose highly concerning environmental risks [35,88]. Therefore, landfill leachate and biogas digestate represent the most relevant sources to be considered for ammonia recovery from waste flows.

Moreover, the nature of the disposed waste exerts a significant influence on ammonia production: municipal solid waste (MSW) landfills, especially those lacking pretreatment steps such as separate collection or organic fraction removal, typically yield higher ammonia concentrations than landfills designed for inert materials. Consequently, the combined effects of landfill age and waste type are pivotal in shaping the potential for ammonia recovery [39].

Recent policy interventions in regions like the European Union have contributed to a reduction in ammonia loads within landfill leachates. Regulatory measures—encompassing separate collection, pretreatment, and the restricted disposal of untreated organic municipal waste—have led to a decline in ammonia concentrations observed in leachates over time [78]. Beyond ammonia content, the total volume of leachate produced also influences resource recovery potential. Factors such as the landfill's surface area, capping systems, and local rainfall patterns determine leachate generation rates, with precipitation often playing a dominant role [29].

From an environmental protection standpoint, untreated landfill leachate poses considerable risks. Leachate contaminants, including ammonia, can migrate into groundwater and surface water, resulting in ecological damage and public health concerns. Legislative frameworks like the Landfill Directive (1999/31/EC) [89] mandate the collection and treatment of leachate to prevent pollution, while the Water Framework Directive (2000/60/EC) [90] imposes strict discharge limits on pollutants, including ammonia. These regulations encourage the deployment of advanced treatment technologies that not only meet compliance targets but also support the broader goals of sustainability and resource efficiency.

Leachate management typically begins with collection via drainage systems, followed by pretreatment steps such as sedimentation or filtration to remove suspended solids and oils [34]. However, conventional treatment methods (e.g., biological processes and chemical neutralization) often struggle to handle the high ammonia loads found in landfill leachates, resulting in reduced treatment efficiency and potential regulatory noncompliance [41,78,91]. In response, ammonia stripping has emerged as a leading technique for effective ammonia recovery from landfill leachate. By elevating the pH to convert dissolved ammonium ions into gaseous ammonia, stripping processes facilitate the subsequent removal of ammonia via air or steam [32].

Beyond stripping, struvite (magnesium ammonium phosphate) precipitation offers an additional route for ammonia recovery. Struvite crystallization not only reduces ammonia concentrations but also yields a valuable fertilizer product. This approach can be employed independently or integrated with other technologies like stripping or membrane separation to enhance overall recovery and resource utilization [5].

Such ammonia recovery strategies align with the principles of the Circular Economy Action Plan (2020) [92], by transforming a problematic pollutant into a useful resource. In doing so, they advance EU objectives related to water quality, waste management, and environmental protection. By adopting and refining ammonia recovery methods for landfill leachate, it is possible to foster more sustainable landfill operations, mitigate environmental impacts, comply with stringent regulations, and promote the efficient recycling of nutrients within the broader resource economy.

Landfill leachate is a complex byproduct resulting from a combination of biological and physicochemical transformations occurring in landfilled waste, coupled with the infiltration of water that dissolves a wide range of organic and inorganic substances [5]. As illustrated before, one of its defining features is the high concentrations of ammonia, primarily due to the microbial decomposition of nitrogen-rich organic waste, such as food residues and other biodegradable materials, a process known as ammonification [41].

The characteristics of landfill leachates are notoriously highly variable, influenced by factors such as the type of disposed waste, the landfill's age, and operational conditions, such as capping, although operational factors have been less extensively studied. For example, younger landfills often exhibit elevated ammonia levels due to the rapid decomposition of fresh organic material, whereas older landfills may show lower levels as organic content stabilizes over time [31]. The type of disposed waste is equally significant in determining ammonia concentrations. For example, landfills containing urban waste typically have higher ammonia levels compared to those designed for inert materials, as the decomposition of the organic fraction in urban waste generates more ammoniacal nitrogen.

In this context, the combined effects of landfill age and waste type play a pivotal role in shaping the concentrations of recoverable ammonia in leachates [39].

Municipal solid waste (MSW) landfills, particularly those that do not pretreat organic fractions, tend to contribute the highest levels of ammonia during the early phases of operation [33]. However, regulatory measures in regions like the EU, including mandates for separate collection, pretreatment, and disposal, have significantly reduced the landfilling of untreated organic municipal waste. As a result, ammonia concentrations in landfill leachates have generally declined in recent years compared to earlier observations.

In addition to ammonia concentrations, the potential for resource recovery also depends on the quantity of leachate generated. This volume is largely determined by factors such as surface area, covering systems, and rainfall patterns; with rainfall often being the primary one [29].

2.2. Biogas Digestate Liquid Fraction

Biogas digestate, a byproduct of anaerobic digestion (AD), is removed from the reactor after the digestion process has been completed [93]. Its composition and characteristics are largely influenced by the nature of the feedstock and the operating parameters of the AD process, which together shape both its physical and chemical properties [44]. In particular, the ammonia content of the digestate depends on the total nitrogen content of the feedstock.

The liquid fraction of digestate derived from livestock manure frequently contains significant ammonia concentrations, which can vary widely from as low as 7.8 mg/L to as high as 12,567 mg/L, depending on the type of livestock—e.g., pig, chicken, or cow slurries—and the operational conditions under which the AD process operates [50]. To address the logistical challenges posed by the large volumes of digestate produced, it is typically separated into solid and liquid fractions. This separation is commonly achieved using centrifugation, screw presses, or belt filters [51]. The solid fraction, which has a high organic nitrogen content, can be spread on agricultural soils directly or after undergoing a composting process [44].

On the other hand, the liquid fraction contains high levels of ammoniacal nitrogen, phosphorus, and other macro- and micronutrients, making it both a valuable resource and a significant challenge for recovery and disposal. Conventionally, the liquid fraction is treated through processes such as lagooning [42], nitrification–denitrification [39], or constructed wetlands. However, these approaches are increasingly being complemented—or replaced—by resource recovery techniques better aligned with circular economy principles, such as ammonia stripping [56], membrane filtration [48], or fertigation [94]. These methods enable nutrient recycling and minimize environmental impacts, supporting sustainable agriculture and waste management. Nevertheless, the improper management of liquid digestate can have severe environmental consequences, including ammonia emissions, soil contamination, and water eutrophication [42,43]. The importance of proper digestate management is underscored by several key regulatory frameworks. The Nitrates Directive (91/676/EEC) aims to limit water pollution from nitrogen compounds, emphasizing the importance of careful liquid digestate management. Additionally, the Renewable Energy Directive (2018/2001/EU) promotes sustainable biogas production and the valorization of byproducts like digestate, while the Fertilizing Products Regulation (EU 2019/1009) supports the use of recycled nutrients, including ammonia, in fertilizers.

3. Ammonia Recovery Technologies and Circular Solutions

Building upon the characterization of the diverse ammonia-rich waste streams described in the previous section, this section focuses on the methods and technologies available for ammonia recovery. The analysis in Section 2 highlights that the characteristics of ammonia-bearing streams, ranging from relatively dilute industrial effluents to highly concentrated landfill leachate and biogas digestates, necessitate tailored recovery strategies. In the following sections, we examine state-of-the-art solutions targeting both liquid-phase and gas-phase ammonia removal, as well as the pretreatment steps required to optimize these processes. By considering both established and emerging approaches—such as adsorption-based methods, membrane separation, and stripping technologies—this section aims to identify pathways that can maximize ammonia recovery efficiency while minimizing environmental impact and operational costs. Moreover, the integration of ammonia recovery within a circular economy framework underscores the importance of transforming residual nitrogen into valuable resources, thereby advancing sustainable waste management and resource utilization practices.

3.1. Liquid-Phase Ammonia Recovery

A substantial portion of ammonia removal efforts targets the liquid phase, specifically addressing ammonium ions (NH_4^+). Unlike gas-phase processes, which primarily involve directly capturing NH_3, liquid-phase ammonium removal relies on materials and methods that exhibit a strong affinity for NH_4^+. Various adsorbents, including natural and modified bentonites, zeolites, biochars, and activated carbons, have demonstrated significant NH_4^+ uptake capacities and high removal efficiencies, depending on factors such as initial NH_4^+ concentration, solution pH, and contact time. For instance, natural bentonite can achieve an adsorption capacity of 19.01 mg g^{-1} at neutral pH, with removal efficiencies around 81.2% in the 10–1000 mg L^{-1} range, while chitosan-composite bentonite reaches 100% removal at lower concentrations, making it suitable for applications requiring complete NH_4^+ capture [95,96]. Zeolites, such as Chinese natural zeolite (13.18 mg g^{-1} capacity), leverage ion-exchange capabilities for selective NH_4^+ adsorption [85]. Similarly, biochars with tailored surface chemistries can achieve substantial capacities (up to 114.84 mg g^{-1} at neutral pH), enabling efficient nutrient recovery and enhancing the sustainability of wastewater treatment processes [97].

Membrane technologies also play a pivotal role in liquid-phase ammonia removal, often relying on semipermeable barriers modified with chemical agents to selectively allow NH_4^+ passage. Hydrophobic membranes, for example, can be modified for improved selectivity and fouling resistance. Nanofiltration (NF) systems, enhanced with polyelectrolytes, achieve NH_4^+ recoveries of up to 92% at low pressures (0.5–1 bar) with initial ammonium concentrations near 1000 mg/L [98]. In another NF approach, membranes grafted with polyamidoamine dendrimers exhibit high selectivity and antifouling characteristics, reaching recovery efficiencies close to 90% in domestic wastewater with elevated ammonium and suspended solids [97,99].

Pervaporation membranes offer another solution for industrial wastewaters rich in ammonium and volatile fatty acids (VFAs). Operating under conditions such as 70 °C and negative pressure, these membranes have achieved 95% ammonium removal at initial TAN concentrations of 4.5 g/L [97]. However, the high organic load in such streams necessitates frequent cleaning and pretreatment. Hollow fiber contactor membranes, configured for direct contact between the ammonium-bearing solution and the membrane, excel in environments with elevated pH and high organic content. Achieving up to 90% recovery at pH 10–11 and TAN concentrations as high as 6 g/L, these systems, when paired with acidified water as a recovery agent, minimize dispersion [86]. The incorporation of materials like graphene oxide (GO), zinc oxide (ZnO), and polyetheretherketone (PEEK) further enhances fouling resistance and suitability for challenging industrial effluents [79].

These advancements clearly highlight the superior performance of advanced membranes over traditional ones, particularly in terms of efficiency, fouling resistance, durability, and cost-effectiveness. Traditional polymeric membranes, such as cellulose acetate and polysulfone, typically achieve recovery efficiencies in the range of 60–75%, but their performance can decline significantly in high-COD streams due to fouling and competition for adsorption sites by organic matter [100–102]. Advanced membranes, such as graphene oxide (GO)-enhanced nanofiltration membranes, address these limitations by achieving recovery rates of up to 89% while maintaining resistance to fouling, even in challenging conditions [79,103,104]. Gas-permeable membranes (GPMs), particularly those made of polytetrafluoroethylene (PTFE), further enhance performance, with recovery rates exceeding 85% in various applications, from laboratory-scale setups to industrial wastewater treatment systems [8,104]. These innovations provide a significant leap forward, particularly in terms of adaptability to diverse wastewater compositions.

Another critical improvement offered by advanced membranes is their durability. Traditional membranes often require replacement within 2–3 years due to chemical degradation and mechanical wear, especially when exposed to harsh conditions [98,105]. In contrast, advanced materials such as PAMAM-grafted forward osmosis membranes and PTFE-based GPMs have demonstrated lifespans exceeding five years, with enhanced chemical resistance and structural stability reducing the frequency of replacements and associated maintenance efforts [8,79,106]. This increased durability translates into lower operational interruptions and contributes to the overall sustainability of ammonia recovery processes.

Although advanced membranes generally entail higher initial costs, approximately 30–40% more than traditional polymeric options, their cost-effectiveness becomes evident over the long term. The reduced energy consumption, lower maintenance requirements, and extended operational lifespans more than compensate for the upfront investment. Hybrid systems that integrate advanced membranes with forward osmosis or membrane distillation technologies further optimize operational costs while maintaining high recovery efficiencies [98,104,107]. These economic advantages make advanced membranes particularly suitable for large-scale or long-term applications, such as high-COD streams or systems with variable feed compositions.

Table 2 summarizes the comparison between traditional and advanced membranes across key parameters, highlighting their respective strengths and weaknesses:

Table 2. Comparison of traditional and advanced membranes for ammonia recovery.

Parameter	Traditional Membranes	Advanced Membranes	Ref.
Efficiency	60–75%	Up to 89%	[100–106]
Fouling resistance	Moderate	High	
Operational lifespan	2–3 years	>5 years	
Cost	Lower initial investment	Higher initial, lower long-term costs	
Applications	Low-COD effluents	High-COD or variable composition streams	

Each liquid-phase technology presents a distinct set of strengths and limitations in terms of selectivity, efficiency, and fouling susceptibility. Chemically modified membranes provide excellent selectivity but require advanced surface treatments, while pervaporation membranes demand extensive pretreatment in high-organic-load scenarios. Hollow fiber membranes perform well in complex conditions but remain sensitive to pH and effluent composition [97,99].

To navigate these strengths and limitations, the selection of adsorbents and membranes for liquid-phase ammonia recovery must align with specific criteria that address operational challenges and stream characteristics. These criteria provide a framework for optimizing performance and guiding material choices in diverse contexts.

A careful evaluation of specific stream characteristics and operational conditions is crucial for optimizing the efficiency and sustainability of these materials. High-COD streams, for instance, present significant challenges due to fouling and the competition of organic matter for active adsorption sites. Advanced nanofiltration membranes, particularly those enhanced with graphene oxide, have demonstrated superior resistance to fouling while maintaining ammonia retention rates as high as 89.3%, even in the presence of elevated COD concentrations [79,98,103]. Similarly, hydrophobic zeolites coated with microporous organic polymers have proven effective, retaining 88% of their ammonia adsorption capacity under relative humidity levels as high as 43% [106]. These properties make such materials particularly suitable for environments with high organic load and moisture levels.

Economic feasibility is another critical factor, particularly for streams with low ammonia concentrations. Materials such as biochar and modified activated carbons, with adsorption capacities ranging from 1.6 mg/g to 6.4 mg/g under low-concentration conditions, provide cost-effective solutions, especially in decentralized recovery systems [104,108]. These materials balance affordability and performance, enabling broader applicability in scenarios where ammonia is present in minimal quantities.

Scalability and flexibility are equally important for technologies targeting diverse industrial or agricultural applications. Gas-permeable membrane systems, achieving ammonia recovery efficiencies exceeding 80%, offer modular designs adaptable to varying flow rates and compositions, making them ideal for large-scale operations [8,109,110]. Additionally, adsorbents like ZSM-5 zeolites with hydrophobic coatings minimize secondary environmental impacts, such as chemical leaching, while ensuring compliance with regulatory standards [95,106,108]. These materials also support circular recovery processes by facilitating the recycling of ammonia into valuable products, such as fertilizers.

By aligning the choice of adsorbents and membranes with these criteria, ammonia recovery systems can effectively address the specific challenges posed by diverse waste streams, delivering both sustainable and economically viable solutions.

Ammonia stripping stands out as a robust and highly efficient approach for liquid-phase ammonia recovery across diverse agricultural and industrial contexts. By manipulating temperature, airflow, and pH, it is possible to optimize ammonia volatilization. Temperatures above 60 °C and pH values near 10 enhance ammonia release, and increased airflow promotes effective separation [42]. The recovered ammonia can be readily converted into valuable products such as ammonium sulfate fertilizer [111]. Stripping proves effective even at very high ammonia concentrations—up to 10,600 mg/L in certain industrial effluents—although pretreatment may be necessary to address sulfates and other ions that can influence efficiency and promote fouling [85]. In agricultural applications, integrating anaerobic digestion (AD) with stripping methods yields recovery rates up to 81% without external energy input, leveraging biogas heat to maintain temperatures near 68 °C and naturally elevating pH to around 10. Direct aeration methods enable ammonia recovery without chemical pH adjustments, relying on inherent carbonate buffering and moderate heating to achieve recovery rates of 70–90% [108].

Technological advancements have introduced more sophisticated ammonia stripping devices, such as rotating packed beds, which maximize gas–liquid interactions and enhance mass transfer to achieve efficiencies above 90%. Integrating electrokinetic processes can further refine in situ pH control without chemical additives, maintaining removal rates near 90% at relatively low energy demands (5–20 kWh/kg-NH_4^+) [85]. Although stripping offers high efficiency and adaptability, it does incur energy costs and requires regular maintenance to mitigate fouling.

Optimal ammonia recovery processes often begin with carefully designed pretreatment strategies, as summarized in Table 3.

Table 3. Pretreatment processes and key benefits across ammonia removal technologies.

Technology	Pretreatment Processes	Key Benefits	References
Stripping technologies	1. pH adjustment: use lime or NaOH to raise pH above 9, converting NH_4^+ to NH_3. 2. Coagulation: Lime helps precipitate $CaCO_3$, reducing organic and suspended solids. 3. Thermal pretreatment: heating the influent to enhance ammonia volatilization and mass transfer.	Enhances NH_3 volatilization, reduces clogging, and improves overall stripping efficiency	[42,108]
Membrane-based technologies	1. Filtration and sedimentation: removal of large suspended solids to reduce fouling. 2. Coagulation and flocculation: alum or ferric chloride help reduce organic fouling. 3. pH control: maintains membrane efficiency by optimizing ammonia selectivity.	Minimizes fouling, maintains selectivity, and prolongs membrane lifespan	[99,108]
Adsorption-based technologies	1. Coagulation or flocculation: alum or ferric chloride reduce competing organic/inorganic contaminants. 2. pH adjustment: optimizes ammonia capture within a neutral to slightly alkaline range. 3. Surface modifications: use iron-loaded activated carbon or nanocomposites.	Improves adsorption capacity, reduces organic fouling, and enhances ammonia capture	[96]

These measures serve to stabilize operational conditions and enhance the performance and lifespan of downstream technologies—ranging from stripping to membrane separation and adsorption—by effectively managing variables such as organic load, pH, and suspended solids.

Adjusting pH with lime or sodium hydroxide enhances ammonia volatilization in stripping systems, while coagulation and flocculation reduce organic and inorganic contaminants that would otherwise clog membranes or adsorption sites [108]. Techniques such as filtration, sedimentation, and chemical treatments improve the interaction of advanced adsorbents (e.g., modified biochars, zeolites) with target species, ultimately increasing adsorption capacity and reliability [96].

For stripping technologies, lime addition not only raises pH but can also precipitate calcium carbonate, thereby aggregating particulates and alleviating column fouling. Thermal pretreatment further enhances mass transfer and lowers energy consumption during the stripping phase [52,85,112]. Membrane-based approaches benefit from preliminary filtration and sedimentation steps to remove large suspended solids, while coagulation and flocculation combat organic fouling [79,113]. pH control is crucial for maintaining membrane selectivity and minimizing scaling. Adsorption-based methods also rely on coagulation or flocculation to reduce competing organic loads, preventing blockage of adsorption sites. Surface modifications to adsorbents, such as iron-loaded activated carbons or nanocomposites, increase resistance to fouling and maintain stable capacities under challenging conditions.

Each pretreatment step is tailored to the chosen ammonia recovery technology, collectively improving separation efficiency, reducing fouling, and enhancing cost-effectiveness. By ensuring stable operational conditions and lowering resource consumption, these measures foster a sustainable approach to ammonia recovery and support consistent, high-performance operation across industrial and agricultural applications.

3.2. Gas-Phase Ammonia Recovery

Managing ammonia in gaseous waste streams often involves the use of selective adsorbents or membrane technologies. Adsorbent materials can be broadly categorized into conventional classes—such as zeolites, activated carbons, and bentonites—and advanced, nanostructured options like metal–organic frameworks (MOFs), ionic liquids, and nanogels. Each category offers distinct advantages and limitations, influencing both operational efficiency and practical feasibility. Among conventional adsorbents, activated carbons are widely utilized due to their high specific surface area and chemical stability. Under 500 ppm of dry NH_3, certain activated carbons achieve adsorption capacities around 50 mg/g, increasing to approximately 73 mg/g at 1000 ppm [84,96]. However, exposure to humid environments can reduce this capacity to about 22 mg/g. Furthermore, the energy-intensive regeneration required for activated carbons progressively diminishes their adsorption performance over multiple cycles, thereby elevating operational costs in large-scale applications [84,96]. Zeolites, featuring microporous frameworks and exchangeable cations, demonstrate high selectivity and stability under dry conditions. Modified variants can adsorb 6.4 mg/g in dry NH_3 and 4.1 mg/g under humid conditions at 4500 ppm NH_3 [95]. Moisture nevertheless remains a challenge, as water molecules occupy active sites and reduce ammonia uptake. To counteract this effect, chemical modifications, including hydrophobic coatings or basic activation, have been developed to enhance ion-exchange capabilities and maintain efficiency in humid atmospheres. Bentonites, primarily composed of montmorillonite clay, utilize their negatively charged surface and cation exchange capacity to capture NH_3. Under dry conditions, adsorption can reach 38.0 mg/g, decreasing to about 12.4 mg/g in the presence of moisture. Integrating bentonites with organic resins or metal oxides increases the number of active sites and enhances their ammonia capture potential [96]. For instance, a composite of bentonite and phenolic-formaldehyde resin has demonstrated improved ammonia removal, making modified bentonites versatile for various environmental conditions [96,114]. Unconventional, nanostructured adsorbents—such as MOFs—offer promising alternatives. Their well-defined porous structures and extensive internal surface areas foster selective ammonia adsorption via coordinative bonds and electrostatic interactions. The MOF HKUST-1, for example, achieves an adsorption capacity of 73.1 mg/g at 1500 ppm NH_3 under humid conditions [115]. Despite these advantages, MOFs face constraints related to humid stability, production costs, and the need for precise regeneration protocols [114,116,117]. Other advanced materials, including boron-doped graphene oxide and multi-walled carbon nanotubes, exhibit exceptionally high capacities—up to 293.4 mg/g for graphene oxide at 2 bar and 22.69–90.05 mg/g for nanotubes at 0.368–0.744 MPa—exploiting large surface areas and favorable chemical interactions, particularly under high-pressure conditions [118]. Fundamentally, gas-phase ammonia adsorption relies on interactions with Lewis and Brønsted acid sites. In zeolites, NH_3 can form NH_4^+ ions at Brønsted sites and stabilize within pores via hydrogen bonding. MOFs often coordinate NH_3 to metal cations (e.g., Cu^{2+}), forming complexes after engaging the nitrogen lone pair. Iron-based metallacycles can physisorb and chemisorb ammonia, increasing overall capacity. Acidic functional groups, such as COOH or HSO_3, can provide additional binding sites, though bulky substituents may partially obstruct pores and lower efficiency [119].

Membrane technologies offer another highly selective, efficient pathway for ammonia separation from gas streams, especially in agricultural and industrial applications. Gas-permeable membranes, often constructed from hydrophobic polymers like polypropylene or polytetrafluoroethylene (PTFE), leverage ammonia's high solubility in water or acidic media to achieve selective separation [8]. Notably, these membranes maintain high performance in humid conditions where conventional adsorbents are less effective, allowing gaseous NH_3 to permeate while preventing liquid infiltration.

Such membranes have been successfully applied in livestock operations, where ammonia levels may reach 3000–12,000 mg NH_3-N/L. Utilizing membranes with areas up to 163.4 cm², ammonia recovery rates of 70–88% have been reported, reducing emissions and enabling the reuse of recovered nitrogen as fertilizer [8]. In industrial contexts, these membranes effectively handle alkaline waste streams from fermentation or food processing, which may contain up to 4.9 g N/L TAN and 41.1 g COD/L VFA. Operating at around pH 11 enhances ammonia volatilization and reduces pH adjustment costs, as higher TAN concentrations improve the partial pressure gradient and thereby separation efficiency. Dense inorganic and advanced polymeric membranes, such as ZIF-21-based systems, have been successfully employed to selectively separate NH_3 from hydrogen, demonstrating permeances of up to 1727 GPU and achieving NH_3/N_2 and NH_3/H_2 selectivities of 35 and 12, respectively [105]. This approach ensures high product purity while minimizing ammonia dispersion. A persistent operational issue with gas-permeable membranes is fouling—caused by the accumulation of organic and inorganic materials—which decreases permeability over time. This challenge is particularly evident in agricultural waste streams. To mitigate fouling, pretreatment steps, such as mechanical filtration and chemical conditioning, are commonly employed. Hydrophilic surface modifications can further reduce particle deposition and maintain overall permeability. Transmembrane chemical absorption (TMCS) complements these efforts by channeling gaseous NH_3 into acidic stripping solutions (commonly sulfuric acid), producing ammonium sulphate, an easily recoverable nitrogen-based fertilizer [120]. However, fouling and wetting remain concerns, necessitating frequent cleaning or pretreatment to sustain long-term efficiency [2,120].

A notable advancement is the introduction of vacuum-assisted gas-permeable membrane (V-GPM) systems, which enable ammonia recovery without pH adjustments. Pilot-scale studies have reported ammonia recovery efficiencies up to 92% at 70–80 °C and 30 kPa, performing effectively with biogas slurry as feedstock [106]. This approach lowers energy and chemical usage and can reduce greenhouse gas emissions by up to -1.6 kg CO_2/kg-N when recovered CO_2 is utilized in greenhouses or other carbon sinks. The modularity of these membrane units supports implementation at both small and large scales, offering a sustainable and adaptable solution for resource-intensive settings [32,94].

3.3. Technical Comparison of Liquid-Phase and Gas-Phase Ammonia Recovery Technologies

Ammonia recovery technologies are broadly categorized into liquid-phase and gas-phase systems, each offering distinct operational mechanisms and advantages. Liquid-phase technologies focus on ammonium ions (NH_4^+) in aqueous streams, while gas-phase approaches target gaseous ammonia (NH_3) in emissions. The choice of technology depends on the characteristics of the waste stream and specific operational requirements.

Liquid-phase recovery methods, including adsorption, membrane separation, and ammonia stripping, exhibit high recovery efficiencies, often exceeding 90% under optimized conditions. Adsorption-based technologies, leveraging materials such as biochars and zeolites, demonstrate significant ammonium uptake capacities (e.g., up to 114.84 mg NH_4^+/g for biochars) [13]. Membrane systems, particularly those using advanced materials like polyamidoamine (PAMAM) dendrimers, achieve selective recovery efficiencies of up to 95% in industrial effluents with high total ammonia nitrogen (TAN) concentrations [98].

Ammonia stripping is widely used for agricultural and industrial effluents, leveraging the volatilization of ammonia by increasing the pH above 10 and operating at temperatures exceeding 60 °C. This process is particularly effective for streams with high ammonia concentrations, such as landfill leachates or anaerobic digestates. Advanced configurations, such as rotating packed beds, achieve recovery efficiencies above 90% by maximizing gas–liquid interactions and reducing energy requirements [14]. In anaerobic digestion systems, biogas heat can sustain the necessary temperatures, reducing external energy demands while achieving ammonia recovery rates of up to 81% [121]. Moreover, integrating electrokinetic processes into ammonia stripping systems can enhance pH control while minimizing chemical inputs, maintaining high recovery rates at relatively low energy consumption [122].

Gas-phase technologies excel in managing ammonia-rich emissions from agricultural and industrial sources. Conventional adsorbents, such as activated carbons and zeolites, provide moderate adsorption capacities but are limited in humid environments. Advanced adsorbents, including metal–organic frameworks (MOFs) and boron-doped graphene oxide, offer enhanced capacities (e.g., up to 293.4 mg NH_3/g under specific conditions) [116]. Gas-permeable membranes (GPMs) provide an effective alternative for separating ammonia from gaseous streams, achieving recovery efficiencies of 70–92%. These membranes operate efficiently under humid conditions, making them suitable for challenging industrial and agricultural applications [8].

The Table 4 highlights the complementary strengths of liquid-phase and gas-phase ammonia recovery technologies. Liquid-phase methods excel in treating aqueous streams with high ammonia concentrations, leveraging established techniques like stripping and membrane separation to achieve high recovery efficiencies. Gas-phase technologies provide robust solutions for gaseous emissions, particularly in agricultural and industrial environments with variable ammonia concentrations and humid conditions. While both approaches offer valuable solutions, the majority of practical applications involve liquid-phase streams, making these technologies the cornerstone of ammonia recovery efforts. Their alignment with existing wastewater treatment infrastructure ensures scalability and efficiency, advancing sustainable nutrient recovery and environmental management.

Table 4. Comparison between liquid-phase and gas-phase ammonia recovery technologies.

Parameter	Liquid-Phase Technologies	Gas-Phase Technologies	Ref.
Efficiency	90–95% (e.g., membranes, adsorption, stripping)	70–92% (e.g., GPMs, MOFs)	[8,13,14,116]
Operational requirements	High pretreatment demand; temperature > 60 °C, pH > 10	Moderate pretreatment; adaptability to humid conditions	
Environmental outcomes	Enables nutrient recovery (e.g., ammonium sulfate)	Reduces NH_3 emissions; produces fertilizers (e.g., NH_4HSO_4)	
Challenges	Fouling, organic load variability	Humidity sensitivity, fouling in agricultural streams	

Despite their effectiveness, specific operational conditions can inhibit the performance of these technologies. Understanding these limitations is crucial for optimizing ammonia recovery systems.

1. High COD levels can significantly impact the performance of membrane technologies. Traditional polymer membranes, such as cellulose acetate, often experience fouling under elevated COD conditions, leading to recovery efficiencies below 60%. Advanced membranes, such as graphene oxide-enhanced nanofiltration systems, demonstrate superior fouling resistance and maintain ammonia retention rates of up to 89% even in wastewaters with COD concentrations exceeding 1000 mg/L [103].
2. Adsorbent technologies are highly sensitive to pH variability. For example, natural zeolites experience a 40% reduction in ammonium uptake capacity when the pH shifts from neutral to acidic (<6), as their ion-exchange capabilities are diminished. Similarly, biochars optimized for neutral conditions show reduced adsorption capacities in alkaline environments where ammonium is converted to ammonia gas, decreasing their efficiency [13,96].

3. Ammonia stripping remains a robust technology for liquid-phase recovery, with its performance less influenced by COD levels. By elevating pH above 10 and operating at temperatures exceeding 60 °C, stripping can achieve recovery efficiencies above 90%, even in waste streams with high organic loads. Advanced configurations, such as rotating packed beds, further enhance gas–liquid interactions and mass transfer. However, streams with low ammonia concentrations (<200 mg/L TAN) can exhibit inefficiencies, requiring additional energy input to maintain effective volatilization [85].
4. Gas-phase technologies, while effective, are hindered by high humidity levels. For instance, the ammonia adsorption capacity of activated carbons drops from 73 mg NH_3/g in dry conditions to 22 mg NH_3/g under relative humidity exceeding 70%. Even advanced materials such as metal–organic frameworks (MOFs) face capacity reductions of 15–20% under similar conditions [116].

By addressing these limitations through tailored pretreatment strategies, the selection of robust materials, and careful operational optimization, ammonia recovery systems can maintain high performance across a range of challenging waste stream conditions. Technologies like advanced membranes and ammonia stripping demonstrate particular resilience, making them suitable for diverse applications. This highlights the importance of adapting recovery approaches to the specific characteristics of the waste stream, ensuring both efficiency and sustainability in ammonia management.

3.4. Advancing Ammonia Recovery Within a Circular Economy Framework

The selection of ammonia recovery technologies depends on a number of factors that are either technology-specific, or are related to the characteristics of the source, such as the pH, the content of suspended solids or other impurities, and the organic load. Moreover, other relevant factors are the capital and operational costs, the technical feasibility, and the level of maturity of the technology. As an example, it was reported that ammonia stripping became economically interesting for concentrations >1500 mg/L [122]. The primarily applied technology for ammonia recovery from leachates is stripping by air or steam, while the use of membranes or struvite precipitation face criticalities due to the high presence of impurities, while adsorption shows low recovery efficiencies [34,42]. Ammonia stripping is also suitable for recovery from industrial wastewater, such as from coke oven wastewater in the metallurgical sector, and increasingly applied to the livestock liquid fraction of digestate [44,56,83]. Adsorption and membrane application can be suitable for specific streams, in particular industrial ones, from which it can be advantageous to recover or separate other substances in addition to ammonia [64–66,72].

The integration of ammonia recovery processes into broader industrial and agricultural systems represents a key opportunity to enhance resource efficiency and reduce environmental footprints. By capturing ammonia from liquid and gaseous waste streams, industries can repurpose this valuable nutrient for energy production, fertilizer manufacturing, or other chemical processes, thereby closing material loops and advancing circular economy objectives. Such circular approaches have already demonstrated success in various wastewater treatment plants and industrial settings, confirming their potential feasibility and impact [42]. The upgrade of a small conventional wastewater system with technologies including a Modified Ludzack–Ettinger biological treatment system, showed a reduction of 40% in the eutrophication potential of the plant [123]. The comparison among three treatment processes for wastewater, a traditional nitro–denitro treatment, anammox, and ion exchange, showed a relevant reduction (97% decrease) for the impact related to eutrophication for ion exchange, when including phosphorous capture and removal and using a high-performance zeolite adsorbent, compared to other processes [124]. Struvite precipitation was indicated as an effective technology for reducing the eutrophication potential at wastewater treatment plants [125].

A Nijhuis Ammonia Recovery system removing ammonia to recover ammonium sulphate demonstrated a negative contribution to the particulate formation when compared to a SHARON anammox plant [126]. A reduction in $PM_{2.5}$ due to NH_3 emissions of about 9–11% was estimated by Zhang et al. in 2022 [127] in Central China by modelling emissions from agricultural and non-agricultural sources under a scenario foreseeing the application of NH_3 emission mitigation policies in agricultural management, industry. and waste disposal. Zaho et al. 2017 [128] modelled a reduction in $PM_{2.5}$ formation in the Hai River Basin in China in 2030 (with respect to 2012) of 9–24% by controlling ammonia emissions with improved technologies and optimizing management and human diet.

Despite these promising outcomes, the economic viability of ammonia recovery technologies remains a critical challenge. Stripping-based systems, for instance, often incur high energy expenditures, while membrane-based solutions can suffer from fouling and the associated costs of frequent cleaning and component replacement. The overall expenses depend on specific operating conditions, waste stream characteristics, and the chosen technology. Nevertheless, life-cycle assessments (LCA) indicate that integrating ammonia recovery with existing wastewater treatment processes—and supplementing them with renewable energy sources—can offset some energy costs and improve overall sustainability [126]. In sectors such as agriculture and waste management, where margins are narrow and regulatory pressures intensify, thorough cost–benefit analyses are essential to determine the most suitable recovery strategies. Under favorable conditions, recovered ammonia from agricultural digestates or industrial effluents can be transformed into valuable fertilizers, reducing the reliance on synthetic nitrogen inputs and minimizing environmental burdens.

Innovations aimed at reducing operational energy demands and improving system efficiency show considerable promise. For example, employing solar heat to power stripping or transmembrane chemical absorption (TMCS) processes diminishes dependence on fossil fuels. Similarly, hybrid configurations that couple ammonia recovery with solid-oxide fuel cells (SOFCs) enable electricity generation from the recovered ammonia, improving energy efficiency and cutting greenhouse gas emissions [17,129,130]. The direct conversion of recovered ammonia into ammonium sulfate fertilizer further highlights the potential for sustainable nutrient recycling, supporting soil fertility and lowering the carbon footprint of food production [43,121]. However, challenges persist. High operational costs related to temperature and pH control, as well as the complexity of maintaining stable efficiency across variable waste streams, hinder widespread adoption. The need for robust, low-maintenance membrane materials and advanced control systems remains pressing. Research on alternative energy integration—particularly the use of renewables to meet heating requirements—could help mitigate these limitations and make ammonia recovery more accessible and economically attractive in diverse settings [42,131,132]. The long-term potential of these technologies is substantial. In agriculture, the strategic use of recovered ammonia reduces dependence on synthetic fertilizers, enhances soil health, and decreases emissions associated with nitrogen production. In industrial contexts, reusing ammonia for energy generation or chemical synthesis can increase overall system efficiency and resilience. By incorporating ammonia recovery into circular economy models, industries and policymakers can attain both environmental and economic benefits over the long term [126]. Rigorous LCAs have confirmed that recovering nitrogen from wastewater treatment plants and agricultural digestates curbs reactive nitrogen emissions, fostering sustainable agricultural practices and mitigating ecological impacts. The transformation of ammonia into ammonium sulfate and other valuable fertilizers exemplifies how circular economy principles can turn what was once a pollutant into a strategic resource [133].

Moving forward, breakthroughs in material science, system integration, and renewable energy utilization have the potential to make ammonia recovery technologies even more effective and cost-competitive. Continued research, innovation, and policy support will be instrumental in establishing ammonia recovery as a cornerstone of circular-economy strategies. By doing so, stakeholders can capitalize on the synergies between environmental stewardship and industrial productivity, paving the way for more sustainable, resilient, and resource-efficient production systems.

In industrial applications, recovered ammonia can be used to generate energy or in production processes, improving resource efficiency and reducing environmental footprints. Such circular approaches have already been successfully implemented in various wastewater treatment plants and industrial settings [42]. The integration of ammonia capture technologies with nutrient recovery systems represents a sustainable solution within the framework of a circular economy. Parallel advancements, such as the molten steel slag gas-quenching process, highlight how resource recovery technologies can address broader environmental objectives. By recovering waste heat and capturing CO2, this method aligns industrial efficiency with decarbonization efforts, reinforcing the circular economy framework [134].

The economic sustainability of ammonia recovery technologies remains a significant challenge for large-scale implementation. Stripping systems, for instance, entail high energy costs, while membrane technologies require frequent cleaning and replacement due to fouling. The overall costs for each technology vary depending on the operating conditions and the source of ammonia. Life cycle assessments (LCAs) have shown that integrating ammonia recovery with wastewater treatment processes can offset some of the energy costs, especially when coupled with renewable energy sources [126]. In sectors such as agriculture and waste management, where operational margins are narrow, careful cost–benefit analysis is essential to determine the feasibility of different ammonia recovery systems. In this context, recovered ammonia from agricultural digestates or industrial wastewater is transformed into valuable resources, reducing reliance on synthetic fertilizers and decreasing environmental impacts.

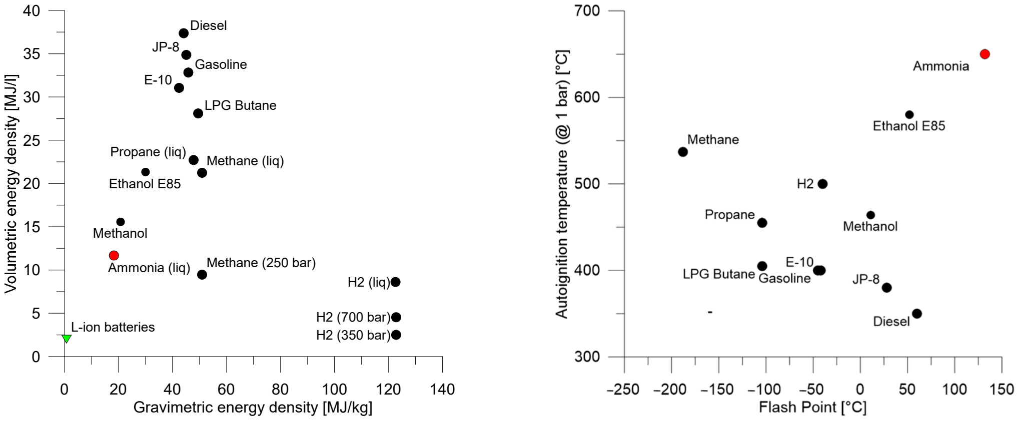
To reduce the energy costs associated with ammonia recovery processes, some studies have proposed integrating renewable energy sources. For instance, solar heat can be used to power stripping or TMCS processes, reducing reliance on fossil fuels. Additionally, hybrid systems combining ammonia recovery with solid-oxide fuel cells (SOFCs) can further optimize energy usage by generating electricity from recovered ammonia. Studies have demonstrated that such integrated systems can significantly improve energy efficiency and reduce greenhouse gas emissions [16,129,130]. One prominent example is the use of ammonium sulphate derived from recovered ammonia as a nitrogen-based fertilizer, contributing to sustainable agricultural practices [43,121]. Despite the clear advantages, current liquid-phase ammonia recovery technologies still face significant limitations. High operational costs, primarily associated with energy consumption and the need to maintain stringent parameters like temperature and pH, are major barriers to widespread adoption. Further advancements in membrane technology and alternative energy integration, such as the use of renewable sources for heating, could help mitigate these limitations, making ammonia recovery more accessible and sustainable. Additionally, the operational complexity of maintaining high efficiency over time in variable conditions makes these technologies challenging to implement in settings with limited resources [42,131,132].

4. Recovered Ammonia Uses

4.1. Direct Combustion

The recovered ammonia from waste treatment processes can be used directly as fuel given its favorable Lower Heating Value (LHV) of 18.6 MJ/kg. Unlike carbon-based fuels, ammonia combustion is entirely free of CO_2 emissions, offering a significant advantage for decarbonization efforts. Additionally, ammonia requires a lower oxygen fraction to achieve complete oxidation, making it an efficient candidate for clean energy applications.

While ammonia's gravimetric energy density is lower than that of hydrogen and many organic fuels (as illustrated in Figure 2a), it compensates with comparable energy output when used in stoichiometric mixtures with air.



(**a**) (**b**)

Figure 2. Comparison of NH_3 with other fuels and energy carriers: (**a**) gravimetric and volumetric energy density (adapted from [22]); (**b**) autoignition temperature and flash point.

For instance, the amount of energy available per unit mass of a stoichiometric NH_3/air mixture is only 7% and 8.5% lower than a comparable diesel/air and methane/air mix, respectively [22]. This implies that adapting existing infrastructure for ammonia combustion—such as adjusting combustor sizing—may require minimal modifications. Furthermore, ammonia's high volumetric energy density and its ability to exist as a liquid under moderate pressures (~20 bar) and ambient temperatures make it cost-effective to store and transport [135]. However, because ammonia is classified as a flammable and highly toxic substance, its storage as a fuel necessitates the implementation of appropriate health and safety measures. Table 5 shows three Acute Exposure Guideline Levels (AEGLs) defined by the Environmental Protection Association (EPA) in the United States:

- AEGL-1 is the airborne concentration causing notable discomfort to the general population, set at 30 ppm regardless of exposure duration. All effects are fully reversible after exposure ceases.
- AEGL-2 represents concentrations at varying exposure times that may cause irreversible or serious, long-lasting adverse health effects, or impair the ability to escape.
- AEGL-3 represents the airborne concentrations above which the general population, including susceptible individuals, could experience life-threatening health effects or death.

Table 5. Ammonia Exposure Guideline Levels (AEGLs) classification according to the Environmental Protection Agency (EPA) [20].

Ammonia Concentrations Expressed in ppm					
Exposure Time	**10 min**	**30 min**	**1 h**	**4 h**	**8 h**
AEGL-1	30	30	30	30	30
AEGL-2	220	220	160	110	110
AEGL-3	2700	1600	1100	550	390

Mitigating factors of the possible hazards due to an ammonia leakage are being lighter than air, which favors its dispersion, and its perceptible odor even at safe concentrations. Liquid flashing leakage, however, can cause greater risks due to the formation of a dense aerosol [136].

From a combustion point of view, ammonia exhibits poor ignition quality and low laminar flame velocity, factors that hinder efficient ignition and stable flame propagation. As shown in Figure 2b, ammonia has a particularly high flash point (132 °C) and autoignition temperature (650 °C) compared to other fuels and energy vectors. These thermochemical characteristics are often associated with lower combustion efficiency, necessitating advanced ignition technologies and optimized combustion strategies to overcome these limitations. Furthermore, ammonia's flammability limits, as detailed in Table 6, indicate additional constraints when compared to other energy carriers.

Table 6. Flammability limits for ammonia and other fuels.

Compound	**Lower Flammability Limit (%)**	**Upper Flammability Limit (%)**
Ammonia	15	28
Methanol	6.7	36
Ethanol (E85)	1.4	19
Methane	4.4	17
Propane	4.4	17
LPG butane	2.1	9.5
Gasoline (E10)	1.9	8.5
Jet fuel (JP8)	0.6	8
Diesel	0.6	6.5
Hydrogen	4	75
Marine Diesel Oil (MDO)	0.6	6.5
Dimethyl ether (DME)	3.4	28

In light of these factors, ammonia's direct combustion stands out as a promising yet technically demanding pathway for sustainable energy production. By addressing these challenges through innovations in combustion technologies, ammonia's potential as a carbon-free energy source can be further realized, supporting global efforts to transition away from fossil fuels. Despite its potential, ammonia's inherent combustion challenges mentioned before necessitate innovative strategies to enhance its performance in practical applications. By leveraging targeted approaches to improve combustion efficiency, the usability of ammonia as a sustainable fuel can be significantly expanded.

One effective strategy involves the use of catalysts or alternative fuels as combustion promoters. This approach not only enhances flame stability and ignition but also takes advantage of readily available compounds in ammonia recovery streams. Table 7 shows a comparison in terms of costs and well-to-tank emissions for fuels mostly used in the shipping industry. Prices refer to the Amsterdam–Rotterdam–Antwerp (ARA) market.

Table 7. Cost and well-to-tank pollutant emissions comparison for different fuels used in the shipping industry; for certain types of energy carriers, a distinction is considered between conventional production from fossil fuels or from renewable energy sources—numbers between brackets—(Table adapted from [137]).

Compound	**Fuel Cost**	**Emissions (Well-to-Tank)**			
		CO_2	**CO**	**SO_x**	**NO_x**
	USD/kg_{eq} *	**(g/kg_{fuel})**			
Marine Fuels (Source: International Maritime Organization IMO)					
Marine Gas Oil (MGO)	0.68	575	N/A	<0.005	0.01
Marine bio-diesel	1.53	19 + 67 **	N/A	0.0400	0.06
Ammonia	1.32	64.9–84.4	0.004	0.0004	0.04
	(2.87)	(18.6–29.8)	(-)	(-)	(0.04)
Methanol	0.98	20.0	0.006	0.0020	0.05
	(2.30)	(17.0 + 120.0 **)	(0.025)	(0.0480)	(0.06)
Hydrogen	1.50	78.0–84.2	<0.074	<0.0700	<0.10
	(2.93)	(7.9–9.7)	(<0.007)	(<0.0640)	(<0.04)
Liquified Natural Gas (LNG)	0.66	8.3–26.7	0.003	<0.0230	<0.09
Liquified biogas (LBG)	2.29	27.0	0.010	0.0730	0.05

* prices expressed per equivalent kg of Marine Gas Oil (MGO) conventionally used in the shipping industry; ** biogenic carbon uptake.

Figures 3 and 4 show the potential improvements achievable on the laminar flame burning velocity (Figure 3) and the ignition delay time (Figure 4) of pure ammonia by adding variable concentration of methane (CH_4), carbon monoxide (CO), and hydrogen (H_2), for variable equivalence ratios and a fixed combustion pressure of 20 bar. These ignition promoters have been selected for their high reactivity with oxygen to improve the poor ignition quality of ammonia and increase its reactivity. Such data relate to experimental measurements available in the literature [138–140].

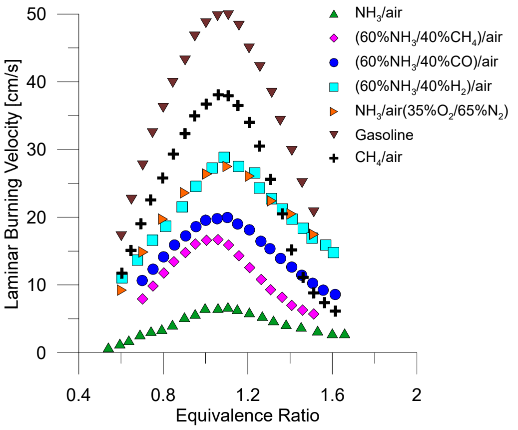
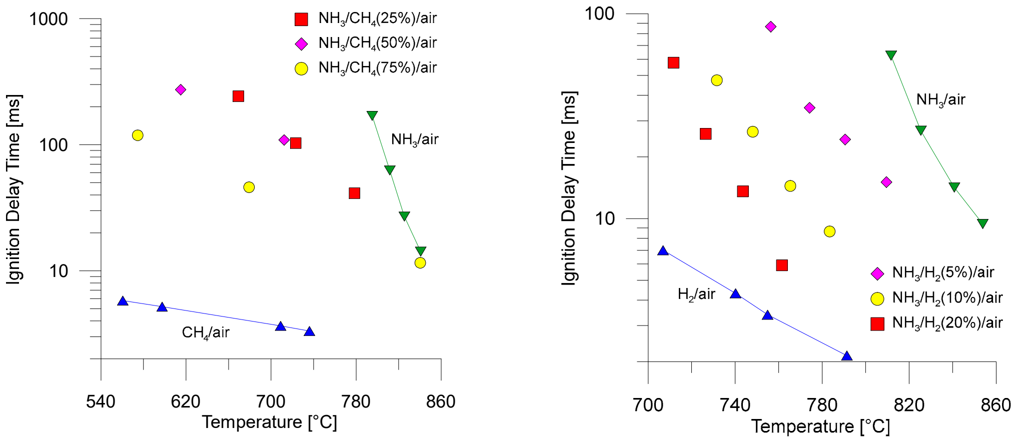


Figure 3. Comparison of experimental measurements found in the literature of laminar flame burning velocities of ammonia and ammonia mixtures with one of the other fuels such as methane and gasoline (adapted from [135]).

Higher laminar burning velocities than those of pure ammonia are required for efficient combustion in conventional engines [138], which would allow one to minimize capital investments thanks to the use of standard components. Indeed, Figure 3 shows how a mixture of ammonia and hydrogen (40% wt of H_2) allows one to achieve a laminar velocity of nearly 30 cm/s for a stochiometric mixture (equivalence ratio equal to 1). This value is comparable to the laminar burning velocities of methane/air and gasoline/air mixtures. It is also interesting to notice that similar results can be achieved by mixing ammonia and oxygen-enriched air (NH_3/air (35% O_2/65% N_2), represented by red triangles in Figure 3);

this approach leverages the availability of pure oxygen often required in wastewater treatment processes. On the other hand, adding methane to ammonia is less effective, since a stochiometric mixture of ammonia and methane (60% $NH_3/40\% CH_4$) with air allows one to achieve a laminar burning velocity of 16 cm/s, almost half of the NH_3/H_2/air and NH_3/O_2/air mixtures.



(**a**) (**b**)

Figure 4. Comparison of ignition delay time of (**a**) ammonia/methane mixtures and (**b**) ammonia/hydrogen mixtures for an equivalence ratio and a combustion pressure of 1 and 20 bar, respectively (data retrieved from [138–140]).

The addition of methane also has a smaller effect on the ignition delay time, an important parameter for combustion efficiency in engines. Figure 4a shows that a methane concentration in the mixture of 50% only moderately shortens the ignition delay time, equal to 100 ms at a temperature of 730 °C. Longer ignition delay times imply a higher amount of premixed combustible mixture in the chamber, leading to more vigorous types of combustion and a potential risk of damage to the engine. In this sense hydrogen is more effective, since smaller additions in mass lead to consistently lower ignition delay times even at lower adiabatic flame temperatures (Figure 4b).

While the addition of hydrogen to ammonia significantly improves combustion performance, it also introduces challenges, particularly regarding the formation of nitrogen oxides (NOx). These emissions pose environmental concerns that must be addressed for ammonia/hydrogen mixtures to become viable in practical applications.

A possible disadvantage of using ammonia/hydrogen mixtures is the propensity to form nitrogen oxide (NOx) emissions, especially the NO compound [141], which have high ozone depletion and global warming potential as well as being important contributors to photochemical smog [142,143]. To mitigate this limitation, several strategies are being investigated, including adjusting combustion temperature and pressure and incorporating methane into the NH_3/H_2 mixture [144].

This approach was investigated by Hewlett S. et al. in [145] who simulated in CHEMKIN PRO a typical waste stream derived from the cleansing of coke oven gas (COG), a by-product of steel-making factories typically rich in ammonia, hydrogen, and methane. The authors tested several ratios of ammonia vapor, and alternatively anhydrous ammonia recovered from the vapor, mixed with COG or methane at air-to-fuel equivalence ratios between 1.0 and 1.4 under elevated temperatures of 280 °C.

The energy efficiency of the overall process was modelled in Aspen considering a Joule–Brayton cycle. The numerical analyses showed that adding 15% of COG to ammonia vapor or anhydrous ammonia substantially improved the combustion characteristics of the mixture and reduced the pollutants emitted. While the use of ammonia vapor reduced the air-to-fuel ratio, it increased NOx emissions. Conversely, using a mixture of ammonia vapor and anhydrous ammonia in a recuperated Brayton cycle engine achieved system efficiencies of 47.3% and 50.1%, respectively [146].

The same mixtures considered in the numerical analyses were tested by the authors in [142] in a swirl burner. The results confirmed that it was possible to combust at 280 °C and atmospheric pressure a mixture of 15% COG and anhydrous ammonia as well as a mixture of 15% COG, anhydrous ammonia, and water vapor (70%/30% mixture). The chemical analysis of the combustion emission products showed that the emissions of NO and NH3 were limited to concentrations lower than 200 ppm and 300 ppm, respectively, by adjusting the air-to-fuel equivalent ratio. According to the authors, further optimization of this ratio could reduce emissions even further. However, limited research is available on emissions resulting from the combustion of ammonia mixtures with oxygen-enriched air, a configuration that could offer a promising application for wastewater treatment plants.

4.2. Ammonia Cracking

Ammonia has emerged as a highly promising solution to overcome the logistical and technical challenges associated with hydrogen as an energy carrier. Boasting a hydrogen content of approximately 17.8% by weight and the capability for liquefaction under moderate pressures (8.6 bar at 20 °C), ammonia provides an efficient means of storing and transporting significant quantities of chemical energy. Its widespread application in the chemical industry has established a robust and extensive global infrastructure, making ammonia a practical and scalable candidate for facilitating hydrogen-based energy systems [147].

Hydrogen extraction from ammonia, commonly referred to as ammonia cracking, involves its thermal decomposition into hydrogen and nitrogen. The reaction proceeds as follows:

$$2NH_3 \rightarrow N_2 + 3H_2 \tag{1}$$

This reaction is endothermic, with a standard enthalpy change of 46.22 kJ/mol, necessitating external energy input to drive the process. Achieving complete conversion typically requires operational temperatures between 400 and 950 °C, depending on the design and configuration of the reactor as well as the catalytic system employed. Catalysts are critical in lowering the activation energy of the reaction, enabling efficient hydrogen generation at reduced temperatures. Among these, ruthenium- and nickel-based catalysts have shown significant potential, demonstrating high catalytic activity and reduced thermal energy requirements in laboratory and industrial applications.

Catalysts used for ammonia cracking are generally classified into three primary categories based on their composition: ruthenium-based catalysts, nickel-based or alternative metal catalysts, and advanced composite catalysts. Each category presents unique advantages tailored to specific applications and operating environments. Ruthenium-based catalysts are characterized by their superior activity at moderate temperatures [148], whereas nickel-based systems offer a cost-effective and scalable alternative, albeit at higher operational temperatures [148]. Advanced composite catalysts, encompassing mixed oxides, perovskites, and carbon-supported systems, represent the forefront of innovation, combining catalytic efficiency, thermal stability, and economic feasibility [148]. Together, these catalytic systems form the foundation for optimizing ammonia-cracking technologies, which are essential for realizing the full potential of ammonia as a hydrogen carrier in future energy systems.

4.2.1. Ruthenium-Based Catalysts

Ruthenium-based catalysts are widely regarded as the benchmark for ammonia cracking due to their exceptional performance at moderate operating temperatures, typically between 400 and 600 °C. A classic example is the Ru/Al_2O_3 catalyst, which achieves conversion efficiencies exceeding 99% when processing high-purity ammonia (>99.9%) [147]. The catalytic activity of these systems can be further enhanced by the addition of alkaline promoters, such as potassium hydroxide (KOH), which lower the activation energy and boost hydrogen production rates. For instance, a Ru-K/Al_2O_3 catalyst tested at 500 °C demonstrated a reaction rate approximately 20% higher than unpromoted Ru/Al_2O_3 under identical conditions [148].

To improve the stability and activity of ruthenium-based systems, researchers have explored alternative supports. For example, Ru/MgO catalysts have shown enhanced resistance to deactivation during extended operations at 450 °C, maintaining conversion rates above 95% even after 100 h of continuous use [149]. Similarly, Ru/CeO_2 catalysts exhibit high activity at temperatures as low as 400 °C, benefiting from the ability of cerium oxide to activate ammonia molecules and mitigate sintering effects [150]. These advancements demonstrate the potential for optimizing catalytic systems to balance performance with durability under varying operating conditions.

Economic considerations remain a significant challenge for ruthenium-based catalysts, as the high cost of the metal limits its application in large-scale industrial processes [151]. To address this issue, innovative dispersion techniques have been developed. For instance, nano-sized Ru particles supported on carbon nanotubes (CNTs) achieved comparable activity to Ru/Al_2O_3 while using 30% less ruthenium. Operating at 450 °C, this system demonstrated a conversion efficiency of 98%, making it a cost-effective alternative for ammonia cracking [148]. Additionally, bimetallic catalysts, such as Ru-Ni systems, have been proposed to further reduce costs while maintaining catalytic efficiency. These systems have achieved conversion efficiencies above 90% at 500 °C, offering a promising pathway for economic scalability [147].

Ruthenium-based catalysts are particularly advantageous in advanced systems, such as membrane reactors, where precise control over operating conditions is essential. In these configurations, they enable simultaneous ammonia decomposition and hydrogen separation, achieving conversion rates exceeding 99% and hydrogen purity levels of 99.998% at temperatures as low as 425 °C [152]. The integration of vacuum conditions on the permeate side of the membrane further enhances performance by facilitating selective hydrogen removal and reducing byproduct accumulation.

4.2.2. Nickel-Based and Alternative Metal Catalysts

Nickel-based catalysts are a cost-effective and scalable alternative to ruthenium for ammonia cracking, particularly suited for large-scale and decentralized hydrogen production. Ni/Al_2O_3 catalysts, in particular, have demonstrated impressive ammonia conversion rates exceeding 90% at operating temperatures between 700 and 900 °C, even when processing industrial-grade ammonia with purities of approximately 99% [153]. While the required operating temperatures are higher than those for ruthenium-based systems, the significantly lower cost of nickel and its robust catalytic performance, especially in fixed-bed reactors, make it a practical choice for industrial-scale applications. These characteristics have positioned nickel-based catalysts as an essential component in ammonia-to-hydrogen conversion systems targeting large volumes and cost-sensitive operations.

To overcome the thermal limitations of conventional nickel-based catalysts, advanced formulations such as Ni-Mg-Al composites have been developed. These composites exhibit enhanced resistance to sintering, a common issue under elevated temperatures, and are less prone to deactivation during extended operation. For instance, a Ni-Mg-Al catalyst maintained stable conversion rates above 85% over 150 h of continuous operation at 800 °C, demonstrating excellent durability and suitability for rigorous industrial conditions [150]. Moreover, the incorporation of promoters like potassium oxide (K_2O) has been shown to lower the activation energy required for ammonia dissociation, further enhancing catalytic activity and enabling higher efficiencies at reduced energy inputs.

In addition to nickel, cobalt-based catalysts have emerged as a promising alternative, offering comparable catalytic performance in ammonia cracking. Co/Al_2O_3 systems, for example, have achieved ammonia conversion efficiencies of up to 85% at 850 °C, underscoring their potential for high-temperature applications. However, the relatively higher cost of cobalt compared to nickel has limited its widespread adoption to niche applications where superior activity and specific operational requirements justify the additional expense [148]. These applications include scenarios demanding enhanced performance or where the catalyst's thermal and chemical stability under harsh conditions provides a significant operational advantage.

Despite these advancements, challenges remain in optimizing nickel- and cobalt-based catalysts to reduce their operational temperature requirements while maintaining or improving conversion efficiency. Innovations in catalyst composition, support materials, and promoter strategies continue to push the boundaries of performance and economic feasibility. For instance, doping nickel-based systems with other transition metals or exploring alloy compositions could unlock new pathways to enhance activity and stability.

In summary, nickel-based catalysts represent a robust and economically viable solution for ammonia cracking at an industrial scale, while cobalt-based alternatives provide additional options for high-performance applications. Continued research and development efforts focused on improving thermal resistance, reducing energy requirements, and enhancing catalytic efficiency are essential to fully harness the potential of these systems in facilitating the transition to hydrogen-based energy technologies.

4.2.3. Advanced Composite Catalysts

Advanced composite catalysts represent a transformative solution for ammonia cracking, offering a unique combination of high catalytic activity, thermal stability, and economic feasibility. This category encompasses perovskites, mixed metal oxides, carbon-based composites, and metal-alloy hybrids, each designed to overcome the limitations of traditional catalysts while optimizing performance under varied operating conditions. Perovskite materials, such as lanthanum nickel oxide ($LaNiO_3$), stand out for their structural adaptability and thermal stability, achieving ammonia conversion rates exceeding 85% at temperatures as low as 600 °C [148]. Their ability to incorporate dopants enhances oxygen mobility and catalytic activity, providing versatility for diverse reactor systems.

Similarly, mixed metal oxides like ZrO_2-CeO_2 ensure long-term stability and resistance to sintering under high-temperature conditions, with Ru-supported versions maintaining over 95% conversion efficiency during 200 h of continuous operation at 450 °C [148,154]. Carbon-based composites, including carbon nanotubes (CNTs) and graphene oxides, improve catalyst dispersion and thermal conductivity, mitigating hot spots and extending catalyst lifespans. For instance, Ru/CNT catalysts have demonstrated 98% efficiency at 450 °C while reducing ruthenium usage by 30%, significantly lowering material costs [149]. Metal-alloy hybrids such as Ni-Fe and Ni-Co systems further enhance catalytic activity and resistance to poisoning through synergistic interactions between metals, achieving conversion rates of up to 87% at 700 °C with industrial-grade ammonia [147]. However, challenges persist in scaling the production of these advanced materials, as the synthesis of perovskites and carbon-based composites often requires precise temperature control and complex chemical processes, increasing costs. Recent innovations in metal–organic frameworks (MOFs) and hybrid nanostructures [154] are addressing these limitations by streamlining production and reducing expenses. Furthermore, the performance of advanced composite catalysts is closely tied to the reactor technologies in which they are deployed. Membrane and fixed-bed reactors provide environments that optimize catalyst efficiency while enabling operational parameters, such as temperature and hydrogen purity, to be tailored to specific applications.

4.2.4. Reactor Technology

Ammonia decomposition for hydrogen production leverages various reactor technologies, with membrane reactors and fixed-bed reactors being the most widely employed due to their distinct advantages. Membrane reactors combine ammonia decomposition and hydrogen separation into a single unit, surpassing thermodynamic limits and achieving high efficiency. For example, a Ru/Al_2O_3 catalyst paired with a palladium-based membrane has demonstrated conversion rates exceeding 99% at 425 °C, producing hydrogen with a purity of over 99.998% [104,147,152]. The use of vacuum on the permeate side of the membrane enhances hydrogen removal, prevents byproduct accumulation, and optimizes reaction kinetics, making these reactors particularly effective for applications requiring ultra-high-purity hydrogen. However, their performance is highly dependent on ammonia feedstock quality, as impurities such as oxygen, sulfur, or chlorine can poison catalysts, significantly reducing their activity. For instance, H_2S concentrations above 1 ppm have been shown to decrease ruthenium catalytic efficiency by 50% within 100 operational hours [147]. Pretreatment systems, including zeolite-based adsorbents or specialized chemical filters, are therefore critical for removing impurities and maintaining reactor performance. Fixed-bed reactors, on the other hand, are a well-established technology for large-scale industrial applications, typically operating with nickel- or ruthenium-based catalysts on Al_2O_3 supports at temperatures between 850 and 950 °C under near-atmospheric pressure. A Ni/Al_2O_3 reactor, for instance, achieves a 95% conversion rate at 900 °C using industrial-grade ammonia (~99% purity), producing hydrogen with purities ranging from 98% to 99.5%. Achieving higher purity requires additional downstream purification steps, such as Pressure Swing Adsorption (PSA) systems or selective hydrogen membranes. The effectiveness of both reactor types is closely tied to the ammonia feedstock quality, as impurities can reduce catalytic activity, operational efficiency, and reactor longevity. An integrated approach encompassing advanced feedstock pretreatment, optimized catalyst selection, and tailored reactor design is essential for maximizing the efficiency and scalability of ammonia decomposition technologies, enabling their broader adoption in hydrogen production applications.

4.2.5. Purity and Operational Considerations in Ammonia Cracking

The efficiency, durability, and hydrogen quality in ammonia cracking are profoundly influenced by the interdependent factors of feedstock purity and operational conditions. High-purity ammonia, typically exceeding 99.5%, is critical to prevent catalyst poisoning by impurities such as sulfur, chlorine, and oxygen. Trace levels of H_2S can decrease the activity of ruthenium-based catalysts by 50% within 100 h, while chlorine compounds cause irreversible deactivation of nickel-based systems. To address these challenges, rigorous pretreatment methods, including zeolite-based adsorbents and chemical scrubbers, are essential for ensuring feedstock suitability for catalytic cracking [147,149]. Operational conditions such as temperature, pressure, and flow rates further play a pivotal role in optimizing hydrogen production efficiency and maintaining catalyst longevity. Membrane reactors, which integrate ammonia decomposition and hydrogen separation in a single system, operate effectively at temperatures between 400 and 600 °C. By continuously removing hydrogen from the reaction system, these reactors prevent product accumulation, surpass thermodynamic limits, and achieve hydrogen purity levels exceeding 99.998%. However, membrane reactors are highly sensitive to ammonia purity, requiring feedstock purities above 99.9% to avoid fouling of separation membranes and ensure long-term stability. Advanced purification steps upstream, such as chemical filtration and adsorption, are often necessary to meet these stringent requirements, particularly for applications like Proton Exchange Membrane (PEM) fuel cells, where hydrogen derived from ammonia must contain less than 0.1 ppm of residual ammonia. In contrast, fixed-bed reactors operate at higher temperatures (850–950 °C) and are preferred for large-scale applications prioritizing robustness and simplicity. While less sensitive to feedstock impurities, these systems still require upstream purification to protect catalysts from H_2S or Cl_2, which could otherwise degrade performance. For instance, Ni/Al_2O_3 catalysts in fixed-bed reactors can achieve conversion rates of up to 95% with industrial-grade ammonia (~99% purity), producing hydrogen with purities between 98% and 99.5%. Additional purification stages, such as Pressure Swing Adsorption (PSA) or selective membranes, are typically needed to meet the stringent purity demands of downstream applications [153]. The interplay between ammonia purity and operational conditions underscores their combined impact on the economic and environmental viability of ammonia cracking technologies. High-purity feedstock reduces maintenance costs by extending catalyst lifespan while ensuring consistent hydrogen production. Furthermore, optimizing reactor conditions—such as lowering activation energy in membrane reactors or enhancing robustness in fixed-bed systems—supports sustainability goals by reducing energy consumption and emissions while maximizing resource efficiency. Together, these considerations highlight the necessity of an integrated approach to ammonia cracking, where feedstock quality, catalyst choice, and reactor design are meticulously tailored to specific operational and application requirements. By aligning these factors with broader environmental objectives, ammonia cracking technologies can significantly reduce greenhouse gas emissions, improve resource utilization, and enhance system performance, contributing to their long-term economic and environmental sustainability.

4.2.6. Environmental and Economic Impacts

The use of ammonia as a hydrogen carrier offers a greenhouse gas-neutral pathway for energy production, as its decomposition emits only nitrogen and hydrogen, avoiding direct carbon dioxide (CO_2) emissions. A fully sustainable cycle involving ammonia synthesis via electrolysis powered by renewable energy and subsequent ammonia cracking results in a process devoid of carbon by-products. Life-cycle assessment (LCA) studies have demonstrated that ammonia decomposition can reduce CO_2 emissions by over 80% compared to traditional methane reforming, particularly when ammonia is synthesized using "green" technologies [147,153]. However, maximizing the environmental benefits of ammonia-based systems requires addressing the potential formation of nitrogen oxides (NO_x) during ammonia combustion in applications such as turbines or engines. Effective NO_x mitigation strategies are essential to align these processes with stringent environmental standards. Despite the high cost of catalysts, especially those based on ruthenium, ammonia cracking remains an economically scalable solution for decentralized hydrogen production. Alternative catalysts, such as Ni/Al_2O_3, have significantly reduced operational costs, enabling broader accessibility for small-scale and modular applications. An economic analysis of pilot plants has projected hydrogen production costs via ammonia cracking to fall below USD 2 per kilogram when integrated with renewable energy systems. The modularity of these systems allows for flexible deployment, ranging from residential-scale setups to large industrial applications, thereby addressing diverse capacity requirements. Ammonia's production via renewable-powered electrolysis further enhances the sustainability of this cycle. When coupled with photovoltaic or wind technologies, ammonia serves as a stable and high-density energy carrier for storing intermittent renewable energy in chemical form, surpassing compressed hydrogen in terms of energy density and storage stability. Recent studies have shown that a closed-loop system combining solar-powered electrolysis, ammonia production, and cracking for hydrogen generation can achieve overall efficiencies of 50%, significantly reducing energy costs while leveraging ammonia's superior storage characteristics [153]. In industrial settings, ammonia cracking facilitates on-site hydrogen production for chemical and metallurgical processes, eliminating the logistical challenges of transporting and storing large volumes of hydrogen gas. At smaller scales, decentralized ammonia cracking systems provide hydrogen for micro-cogeneration plants, enhancing energy self-sufficiency and reducing local CO_2 emissions. Together, these capabilities underscore the critical role of ammonia as a versatile hydrogen carrier in advancing sustainable and scalable energy solutions.

4.2.7. Integrated Energy Use of Recovered Ammonia from Waste

The recovery and energy utilization of ammonia represent a strategic approach to valorize waste streams through a sustainable integrated cycle. Recovered ammonia can be used to produce hydrogen via cracking and integrated into other energy processes, contributing to greenhouse gas emission reduction and optimizing overall efficiency. A notable example is the NWaste2H2 technology, which combines ammonia cracking with methane reforming in a single fixed-bed reactor. The heat generated from ammonia decomposition ($2NH_3 \rightarrow N_2 + 3H_2$) drives the endothermic methane-reforming process ($CH_4 + H_2O \rightarrow CO + 3H_2$), thereby enhancing energy efficiency. Experiments conducted with nickel-based catalysts demonstrated an ammonia recovery efficiency of 89.9%, operating at temperatures between 650 and 800 °C and pressures of 1 bar [17]. Aspen Plus simulations confirmed that this system achieved hydrogen yields close to thermodynamic equilibrium while significantly reducing greenhouse gas emissions and optimizing energy use. Input ammonia purity exceeding 99% is essential to avoid catalytic poisoning from impurities such as sulfur and chlorine, which could compromise catalyst stability.

Hydrogen produced from recovered ammonia cracking has been used in various energy applications, including fuel cells and transport networks. In solid-oxide fuel cells (SOFCs), hydrogen with a purity above 99.5% was used to produce both electricity and heat, achieving a 45% system efficiency improvement. Simulations highlighted a reduction in greenhouse gas emissions of up to 4.3 kg of CO_2 per kg of biomethane treated, showcasing the potential of SOFCs for large-scale applications [153]. Hydrogen injection into energy networks, while economically less favorable compared to other options, showed advantages for decentralized applications, although further purification to exceed 99.99% purity was required [155].

The entire integrated cycle, starting with ammonia recovery from waste and culminating in its transformation into high-purity hydrogen, represents a virtuous model of a circular economy. For instance, a pilot plant recovering ammonia from digestate demonstrated a 92% recovery efficiency, producing hydrogen suitable for micro gas turbines. This approach reduced the system's total energy demand by 15% through the recovery and utilization of heat generated during the process. Simulations and real processes highlighted the scalability and applicability of these technologies in various contexts, offering both environmental and economic benefits [149,152,153].

4.3. Direct Ammonia Fuel Cells

Compared to direct combustion, where the chemical energy of the fuel is transformed into thermal, mechanical, and then in electrical energy, fuel cells allow one to perform this transformation in a single electrochemical reaction. For this reason, they are seen as an attractive energy conversion technology especially at low-to-medium-scale applications, where their limited power density is counterbalanced by superior efficiency, minimal emissions, no moving mechanical parts, and their flexibility about fuel usage type.

For ammonia recovery applications, the use of direct fuel cells is particularly attractive because of the theoretical possibility of extracting a higher amount of electrical energy compared to direct combustion. In addition, exhaust gases from certain types of direct ammonia fuel cells can still be fed to a combustor to extract the residual chemical energy contained in the compounds not oxidized in the cell. This allows them to generate both electrical and thermal energy that can be eventually reused in the upstream industrial processes.

Compared to combustors, however, feeding direct ammonia fuel cells requires one to obtain a higher level of purity for the recovered ammonia, and therefore a higher number of treatments are needed. Other challenges are represented by the low technological maturity of such devices, which are mainly demonstrated in laboratory environments.

Direct ammonia fuel cells can be divided into two macro-families, depending on the type of electrolyte used, which can be alkaline (alkaline solution, molten hydroxide or alkaline membrane) or solid.

4.3.1. Alkaline Electrolyte Fuel Cells

Ammonia alkaline fuel cells operate using the exchange of anions. Air is fed into the cathode side, where oxygen reacts with water molecules to form hydroxide anions. These hydroxide anions are transported through an anion exchange membrane or an alkaline electrolyte, depending on the cell design. Ammonia is fed as fuel on the anode side, where it reacts with hydroxide ions to produce nitrogen and water. The reaction between oxygen and water at the cathode consumes electrons, while the reaction between ammonia and hydroxide ions at the anode produces electrons, resulting in an electric current. Alkaline-type cells typically operate at low to medium temperatures (below 450 °C). Since ammonia is not decomposed but directly oxidized, the reaction kinetics are slow, necessitating the use of catalysts. While noble and transition metals are commonly used as catalysts, current research focuses on less expensive materials, such as nickel, to reduce development costs.

Among the main technological challenges hindering the development of alkaline direct ammonia fuel cells are ammonia crossover, which reduces the open-circuit voltage (OCV) and, consequently, the cell's efficiency; low power density; catalyst deactivation caused by the adsorption of intermediate chemicals during ammonia oxidation at the anode; and membrane degradation. Another significant drawback is the requirement to use CO_2-free air or pure oxygen at the cathode to prevent the reaction between CO_2 and hydroxide ions, which leads to the formation of carbonate precipitates, thereby degrading cell performance. This requirement introduces an efficiency penalty due to the energy needed to remove CO_2 from the air or produce pure oxygen. To address this, ongoing research is focused on developing CO_2-tolerant OH^- ion conductors for use as electrolytes in alkaline ammonia cells.

In general, the alkaline electrolyte is the most critical component since its chemical stability is crucial for achieving the practical longevity of alkaline fuel cells. Recent advancements have improved the power density and longevity of membranes for use in ammonia alkaline fuel cells through the development of anion exchange membranes with high conductivity and enhanced chemical and mechanical stability. These membranes are typically designed with two distinctive features: a polymer backbone and an ion-conductive moiety.

Poly(aryl piperidinium) (PAP) is a commonly used material for fabricating polymeric membranes since it allows one to overcome the temperature-dependent limitations of conventional AEMs.

The thickness of the electrolyte in alkaline electrolyte fuel cells is a critical factor affecting fuel-cell performance. Thinner membranes can reduce ohmic losses by shortening the diffusion and mass transfer distances for OH^- ions, enabling faster delivery of reactants to the anode. However, thinner membranes must maintain good mechanical properties and robustness. Excessively thin membranes can lead to fuel permeation across the membrane, resulting in fuel crossover and potential cathode flooding. Such issues have been observed, leading to a reduced OCV and overall fuel-cell performance [156].

4.3.2. Solid Electrolyte Fuel Cells

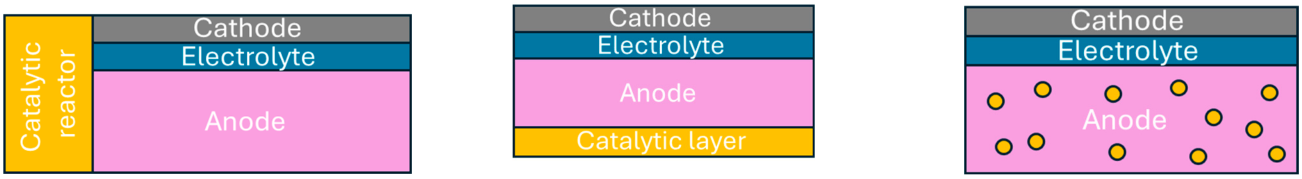
Solid electrolyte fuel cells presents higher performance in terms of power density and efficiency because of the greater operating temperature, which can vary between 500 °C and 1000 °C. At elevated temperature, the ammonia decomposes to form nitrogen and hydrogen, which allows the oxidation of hydrogen instead of ammonia, with much faster kinetics. Increasing the operating temperature of solid electrolyte fuel cells leads to higher ammonia decomposition rates, with consequent higher performance of the cell. Besides the operating temperature, the concentration of hydrogen and ammonia in the anodic chamber also affects the decomposition rate, and therefore the fuel flow in the cell is an important control parameter to maximize the cell performance. The decomposition rate of the ammonia can in fact be expressed as the ratio between the partial pressure of ammonia and hydrogen in the anodic chamber, following the Temkin−Pyzhev relation [157].

In direct ammonia solid electrolyte fuel cells, the decomposition of ammonia occurs internally within the cell. The heat generated by the electrochemical exothermic reaction drives the endothermic ammonia decomposition reaction. This heat transfer enhances the efficiency of the cell, as it eliminates the need for external thermal energy when occurring at the optimal temperature.

The decomposition, or cracking, of ammonia can occur via indirect or direct methods, altering the cell layout. In indirect methods, the cracking reactor is internal to the cell and in close proximity to the electrodes and electrolyte to facilitate effective heat transfer. In direct methods, the cracking reaction takes place within the anode chamber itself, utilizing the heat generated from the exothermic electrochemical reaction. This arrangement requires a multifunctional electrode capable of providing high catalytic activity for both ammonia decomposition and the electrochemical reaction.

Direct internal decomposition layouts can be realized either by incorporating a porous catalytic layer at the entrance of the anode chamber or by diffusing a nanocatalytic phase within the anode material. While the direct ammonia decomposition layout offers advantages such as increased efficiency and a more compact cell design, it also exposes the solid electrolyte fuel cells to degradation risks. Specifically, the anode may suffer from the formation of nickel nitrides, which occupy a larger volume than the nickel-based particles that act as catalysts in the anode. This expansion can induce mechanical stress within the electrode, potentially leading to cell failure.

Figure 5 illustrates a schematic representation of the different types of layouts explored in the literature for ammonia decomposition in direct ammonia fuel cells.



(**a**) (**b**) (**c**)

Figure 5. Comparison of different ammonia decomposition fuel cell arrangements: (**a**) indirect decomposition, (**b**) direct decomposition via porous catalytic layer, and (**c**) direct decomposition via nano-catalyst diffusion on the cell anode (adapted from [157]).

Other distinguishing features of solid electrolyte fuel cells include their manufacturing design, which can be planar, tubular, segmented-in-series, or monolithic; the location of the cell support, which can be at the anode, cathode, electrolyte, or interconnector; and the type of flow configuration, such as co-flow, counter-flow, or cross-flow. In general, the performance of the cathode and anode is governed by the sum of the polarizations associated with the rates of charge transfer at the triple-phase boundary (TPB), the effective ionic and electronic conductivity of the electrode, and the transport of gaseous species through the porous electrode. The electrolyte's performance, on the other hand, is determined by its ionic conductivity.

The greater thickness of the ceramic membrane negatively impacts its ionic conductivity, which, in turn, affects the overall cell performance. Therefore, a trade-off exists in the design of solid electrolyte fuel cells between decreasing the electrolyte thickness to enhance cell performance and increasing it to improve mechanical resistance and long-term durability under thermal stresses and gradients.

Depending on the type of ionic species conducted by the ceramic electrolyte, which can be oxygen ions or protons, two main classes of solid electrolyte fuel cells can be distinguished: solid-oxide fuel cells (SOFCs, also referred to in the literature as SOFC-O) and Protonic Ceramic Fuel Cells (PCFCs, also referred to in the literature as SOFC-H).

Solid-Oxide Fuel Cells (SOFCs)

In solid-oxide fuel cells (SOFCs), oxygen ions are reduced at the cathode and transported via the ceramic membrane to the anode, where hydrogen, formed by the decomposition of ammonia, is oxidized. However, the presence of undecomposed ammonia and nitrogen in the anode chamber can lead to competing reactions with hydrogen oxidation, resulting in a decrease in cell performance. Additionally, these reactions produce water and nitrogen oxides (NO_x), which are expelled as anode gas exhaust. NO_x are harmful pollutants that negatively impact the environment, contributing to ground-level ozone formation, photochemical smog, and acid rain. Consequently, a post-treatment system is required for these cells to remove such pollutants from the anode exhaust gases.

Due to the oxidation reactions occurring at the SOFC anode, anode materials must exhibit adequate properties, including high conductivity, porosity, fuel flexibility, chemical and mechanical stability, electrochemical activity, and resistance to catalyst poisoning. The choice of anode material depends on the fuel-cell operating temperature, with common materials being nickel-based or noble metals.

Experimental tests with Ru-based anodes have demonstrated the highest catalytic activity for ammonia decomposition, while Ni-based anodes have shown the best performance among non-noble materials. Nickel-based materials also offer higher electronic conductivity and are cost-effective. However, a disadvantage of using Ni-based anodes is their tendency to form nickel nitrides, which can negatively impact the catalytic and conductive activity of the cell.

Electrolytes in SOFCs are typically ceramic materials designed to provide optimal oxygen ion transport and chemical stability at high temperatures. It is also critical that the electrolyte has a coefficient of thermal expansion (CTE) similar to that of the electrode materials to minimize mechanical stresses during thermal transients. Low electronic conductivity is also essential. Yttria-stabilized zirconia (YSZ) is a widely studied electrolyte material due to its long-term stability in different environments and high ionic conductivity.

The cathode in SOFCs must have sufficient porosity to support oxygen diffusion, high ionic conductivity for oxide ions, appreciable electronic conductivity, and high electrocatalytic activity for efficient oxygen reduction reactions (ORRs). It must also have a CTE compatible with the electrolyte to prevent thermal stresses that could compromise structural integrity. Additionally, the cathode must remain stable in oxidizing environments and chemically inert toward the electrolyte material.

The cathode is considered a bottleneck for reducing the operating temperature of SOFCs, as the ORR is a temperature-dependent process. At lower temperatures, the reaction kinetics slow down significantly, increasing polarization resistance and limiting overall cell performance. Reduced operating temperatures also lead to limited ionic and electronic conductivity in cathode materials, restricting oxygen ion availability at reaction sites, and reduced catalytic activity, which hinders oxygen dissociation and ionization. To address this challenge, advanced cathode materials such as lanthanum strontium cobalt ferrite (LSCF), perovskite-based compounds, and mixed ion–electron conductors are being developed.

These materials offer improved ORR kinetics and ionic conductivity at lower temperatures, though their long-term stability and compatibility with other cell components remain areas of active research.

Proton-Conducting Fuel Cells (PCFCs)

Compared to solid-oxide fuel cells (SOFCs), Proton-Conducting Fuel Cells (PCFCs) eliminate the disadvantage of generating NOx in the exhaust gases. At the PCFC anode, hydrogen gas produced by the decomposition of ammonia is oxidized to protons (H^+) in the presence of a catalyst. The H^+ ions are transported through a proton-conducting electrolyte to the cathode–solid electrolyte interface, where they react with oxygen to form water vapor. The resulting water vapor and unreacted oxygen exit the fuel cell through the cathode side, while the anode exhaust gases consist only of nitrogen and undecomposed ammonia.

Due to the reaction mechanisms at the PCFC anode, which avoid the dilution effect caused by the oxidation of undecomposed ammonia or nitrogen, PCFCs achieve efficiencies approximately 10% higher than SOFCs and peak power densities 20–30% greater than SOFCs at an operating temperature of 800 °C [157]. At temperatures above 800 °C, the peak power density of an SOFC can be about 50% higher than that of a PCFC, but this is primarily attributed to uncertainties in estimating the exchange current density of PCFCs compared to the rate expressions used for SOFCs [158]. Furthermore, the unexpected hydrogen dilution at the SOFC-H cathode reduces power output, highlighting areas for improvement to unlock the full theoretical potential of SOFC-H systems [157].

Temperature significantly affects PCFC performance, as protonic conductivity across the electrolyte increases with temperature, as described by the Arrhenius equation. Higher temperatures reduce cell resistance by minimizing ohmic losses at the electrolyte and polarization losses at the electrodes. Additionally, elevated temperatures promote more complete ammonia decomposition, enhancing proton availability and, consequently, cell performance.

Similar to SOFCs, materials such as Pt, Ag, Pd, and Ni, as well as perovskites, can be used for PCFC anodes. In PCFCs, the electrolyte is the key limiting material for reducing operating temperature and improving performance. The proton conductivity of PCFC electrolytes, such as barium cerate ($BaCeO_3$) and barium zirconate ($BaZrO_3$), is highly temperature-dependent. At lower temperatures, the mobility of protons decreases significantly, leading to reduced ionic conductivity and diminished cell performance. Moreover, the activation of the proton-conducting electrolyte requires a certain level of thermal energy.

The electrolyte must be dense and sufficiently thin to reduce ohmic resistance, optimize protonic conduction, and minimize electronic conduction to prevent current leakage. Recent research focuses on reducing electrolyte thickness to lower internal resistance; however, excessive thinning can compromise mechanical integrity, increasing the risk of structural issues like cracking or gas leakage. Thus, a trade-off exists between maximizing performance and ensuring the long-term durability of the fuel cell. For the cathode, noble-based materials, Ni-based materials, and perovskites are commonly employed [158].

4.3.3. Semiconductor Fuel Cells

The use of semiconducting-ionic materials for the electrodes and electrolyte of SOFCs has garnered significant interest in the research community due to their potential for achieving higher power densities at lower temperatures compared to traditional SOFCs and PCFCs. This type of fuel cell, often referred to in the literature as semiconductor fuel cells, represents an emerging technology that enhances ion and electron conductivity at lower temperatures and can be manufactured as a single layer. This single-layer design reduces contact losses between the different layers of the fuel cell, potentially leading to higher efficiency. Additionally, it offers the promise of lower manufacturing costs and reduced system complexity [158]. Figure 6 shows the maximum peak power densities achieved by the ammonia semiconductor fuel cells tested in the literature and compares them with the power densities achieved by the other types of ammonia fuel cell technologies. The figure also highlights the typical operating temperature range of each technology, which also depends on the electrodes' material used. Figure 6 shows the potential of semiconductor fuel cells to achieve elevated power density values between 200 mW/cm^2 and 800 mW/cm^2 at lower operating temperatures (around 450–500 °C) compared to SOFCs and PCFCs.

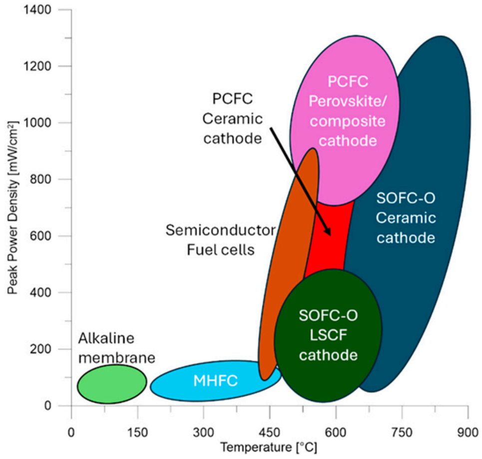


Figure 6. Comparison of the different types of direct ammonia fuel cells experimentally tested in the literature divided by their operating temperature range, peak power densities achieved, and types of anode/cathode employed (from data retrieved in [140,156–165]).

4.3.4. Microbial Fuel Cells

Microbial fuel cells (MFCs) are an alternative technology that has garnered significant attention for their ability to treat wastewater while simultaneously generating electricity. These fuel cells are particularly promising for integration with wastewater treatment processes, as they require minimal processing of ammonia-recovered waste streams. MFCs utilize microorganisms to convert chemical energy from biodegradable material into electrical energy. Electricity is generated when bacteria on the anode oxidize organic substrates present in wastewater, producing electrons. The electrons flow from the anode through an external circuit, where they combine with protons and oxygen at the cathode to form water. Electricity is harnessed by placing a load between the two electrodes. Several experiments reported in the literature have demonstrated the potential of MFCs to simultaneously remove ammonia from wastewater and generate power, achieving high ammonia removal efficiency and power density (99% and 100 mW/m^3, respectively) [165]. These cells often employ air-breathing cathodes to simplify the system and make it more scalable [165]. However, this approach can result in ammonia emissions into the atmosphere, posing environmental and health risks. Mitigation solutions are therefore required to dilute ammonia emissions to levels that are safe for human operators [166]. Another common challenge with this type of cell is biofouling, caused by biofilm formation on the cathode, which can lead to catalyst deterioration and reduced performance. One potential solution to this issue is the use of magnetic cleaning methods for cathodic components, as proposed by Rossi [167]. MFC technology has also been explored as a method for air purification, particularly for removing ammonia [168]. In this context, Sn-doped V_2O_5 nanoparticles have proven to be effective catalysts for the rapid removal of ammonia via photo-electro-catalysis (PEC) in MFCs. An optimal ammonia degradation efficiency of 96.4% was achieved. Sn-doping reduced the nanoparticle size, increased the oxidizing capacity, and enhanced the number of active sites. Oxygen vacancies played a critical role in the ammonia oxidation process [168].

4.4. Case Studies on the Experiment of Recovered Ammonia Reuses

4.4.1. Direct Combustion and Direct Reuse of Ammonia

Among the few case studies available in the literature, few come from the steelmaking industry. In [140], the authors experimentally tested the combustion of ammonia mixed with coke oven gas recovered from a steelmaking process in a swirl burner. The typical composition of the COG was reproduced in a laboratory, with a high percentage of hydrogen and methane. The results confirmed that it was possible to combust at 280 °C and atmospheric pressure a mixture of 15% COG and anhydrous ammonia as well as a mixture of 15% COG, anhydrous ammonia, and water vapor (70%/30% mixture). A wide range of the air-to-fuel equivalent ratios for the abovementioned mixtures were tested to assess the optimal value able to maximize the combustion efficiency and minimize NO_x emissions. The chemical analysis of the combustion emission products showed that the emissions of NO and unburned NH3 were limited to concentrations lower than 200 ppm and 300 ppm, respectively, for an air-to-fuel equivalent ratio of 1.4. According to the authors, further optimization of this ratio could reduce emissions even further. However, limited research is available on emissions resulting from the combustion of ammonia mixtures with oxygen-enriched air.

In [169], the authors examined the production of ammonia and, subsequently, urea for fertilization purposes via the reforming of recovered steel mill gases. This process was compared with the conventional base case, which typically involves using steel mill gases for internal plant operations or as fuel for energy generation in power plants. By redirecting a portion of the gas originally allocated for power generation to ammonia production, it was estimated that the Thyssenkrupp Steel plant in Duisburg could produce approximately 850 t/day of urea. In that scenario, hydrogen availability was the limiting factor for production. By utilizing natural gas to meet the plant's internal energy needs and using all by-product gases for power generation and urea production, the plant's urea production capacity could potentially be increased to 3500 t/day. In both cases, the authors estimated a reduction in CO_2 emissions resulting from the combined processes of steelmaking and urea production.

Another significant advancement in the potential reuse of ammonia in the steel industry is the Ammonia Direct Reduction (ADR) process, as investigated in [170]. This method, proposed as a replacement for the hydrogen-driven direct reduction process, demonstrates strong potential for reducing the carbon footprint of steel production. By eliminating the need for preliminary ammonia cracking and achieving reduction kinetics at 700 °C comparable to those of hydrogen-based methods, the ADR process produces a mixture of metallic iron and an iron nitride protective phase, which can be removed through melting. The process is autocatalytic, as the reduced iron facilitates further ammonia decomposition, enabling additional cost savings and improved efficiency.

4.4.2. Fuel Cells

Case studies on the direct use via fuel cells of recovered ammonia are also scarce in the literature and mainly focused on industrial, municipal, and agricultural wastewaters. In [110], the authors simulated the potential ammonia recoverable from a Wastewater Treatment Plant (WWTP) located in United Kingdom, showing that 82% of ammoniacal nitrogen present in digestate liquor could be used for energy applications. In [129], the authors performed a similar analysis using an electrodeionization process to produce ammonia from synthetic ammonium-rich wastewaters which was reformed together with biogas synthetically produced by a lab-scale anaerobic digestate (AD) reactor using as input local sewage sludge. The analysis showed that a 60% improvement in net energy output could have been achieved compared to the use of a conventional Combined Heat and Power (CHP) system.

However, the ammonia recovered went to a reforming process together with biomethane to generate hydrogen that was consequently fed to an SOFC. The only experimental test of recovered ammonia fed in a direct fuel cell was reported in [130], where the researchers considered a recovery process from struvite precipitation. The analysis showed that the full cracking of ammonia in the SOFC was achieved, with an efficiency equal to 99.6%. The tests also showed that the fuel cell was able to generate at 0.6 V a current density varying from 2000 A/m^2 to 3000 A/m^2 when the ammonia concentration in the ammonia/water stream varied from 14% up to 100%.

Other studies explored the viability of using low-temperature direct ammonia fuel cells to remove ammonia from waste streams and therefore produce electricity. In [171], the authors tested an alkaline fuel cell with not-noble electrode materials to remove 96.5% of ammonia from landfill leachate. The fuel cell, composed of an alkaline membrane as the electrolyte, a NiCu-C as the anode, and a-MnO as the cathode, allowed the authors to generate a power density of 0.35 mW/cm^2 for a 3 M NH_3/water stream at an operating temperature of 30 °C. A commercial direct ammonia alkaline fuel cell coupled with an ammonia stripping unit was tested in [172]. The fuel cell, operating at a temperature of 90 °C, achieved a power density of roughly 0.04 mW/cm^2 for a 5 M NH_3/water solution. In [148], the authors presented an overall review of microbial fuel cells used for ammonia removal from wastewater streams.

5. Conclusions

Recovered ammonia represents a transformative solution to the challenges of sustainable energy production, waste management, and resource efficiency. By recovering ammonia from diverse waste streams such as landfill leachates, biogas digestates, and industrial effluents, it is possible to mitigate environmental pollution while harnessing a renewable energy carrier. Advanced recovery technologies, including ammonia stripping, membrane separation, and adsorption systems, have demonstrated remarkable efficiency in capturing ammonia even from high-concentration and complex waste streams. These methods align with circular economy principles, enabling industries to reduce dependency on synthetic fertilizers, minimize environmental impacts, and transform waste into valuable resources. Hybrid recovery systems, which integrate multiple processes and utilize renewable energy sources, offer promising solutions to improve sustainability and scalability while reducing operational costs.

Ammonia's versatility as an energy carrier further enhances its appeal, with applications ranging from direct combustion to hydrogen generation and fuel cell integration. Its high energy density, carbon-free combustion characteristics, and established global distribution infrastructure position it as a viable alternative to traditional fossil fuels. This versatility aligns with the framework of integrating complete hydrogen energy chains, optimizing renewable energy utilization and decarbonization strategies in multi-energy systems [173]. Advances in ammonia combustion technologies, including co-firing strategies and NOx emission mitigation techniques, have improved its efficiency and environmental performance. Simultaneously, ammonia cracking has emerged as a scalable pathway for producing high-purity hydrogen, supported by innovations in catalytic materials and reactor designs. These advancements enable the integration of ammonia into solid-oxide fuel cells (SOFCs) and other energy systems, achieving high conversion efficiencies while reducing greenhouse gas emissions.

Despite these advantages, significant challenges remain, including the high cost of advanced catalysts, energy-intensive recovery and cracking processes, and the need for robust integration with existing infrastructures. Issues such as catalyst deactivation, membrane fouling, and NOx emissions during combustion underscore the importance of continued research and innovation. Improving catalyst efficiency, reducing energy demands through renewable energy integration, and optimizing system designs for varied applications are essential to overcoming these barriers. Additionally, life-cycle assessments and techno-economic analyses are critical for ensuring the long-term sustainability and economic viability of ammonia-based technologies.

Beyond its role as an energy carrier, ammonia offers broader implications for global sustainability. By enabling circular economy practices and turning waste into a resource, ammonia recovery reduces dependency on non-renewable feedstocks and supports decarbonization goals. Its integration into agriculture, transportation, and industrial energy systems aligns with global efforts to achieve carbon neutrality, enhance energy security, and address climate change. Scaling these technologies will require coordinated action across academia, industry, and government, with policies that incentivize ammonia recovery, promote renewable energy integration, and fund research into emerging technologies. Collaborative initiatives will accelerate the deployment of ammonia-based solutions in sectors such as heavy-duty transport, maritime shipping, and grid-scale energy storage.

In conclusion, recovered ammonia represents a unique opportunity to address the intertwined challenges of waste management and sustainable energy production. Its ability to decarbonize energy systems, enhance resource efficiency, and contribute to global sustainability underscores its transformative potential. By addressing current limitations and fostering innovation, recovered ammonia can become a cornerstone of the energy transition, reshaping industrial and energy practices while paving the way for a more resilient and sustainable future.

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