



Review Article

The potential of natural adsorbents for ammoniacal nitrogen removal in Malaysia's industrial wastewater: A mini review

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Abstract

Ammonia pollution in wastewater poses a significant environmental challenge thus adversely affecting both water quality and public health especially in developing countries like Malaysia. This review seeks to highlight the potential of various natural adsorbents for effective ammonia removal, focusing specifically on their suitability within the Malaysian context. The findings reveal that locally available materials such as coconut husk, palm oil biomass, biochar and algae possess substantial ammonia adsorption capacities which employ mechanisms like ion exchange and other physicochemical interactions. Notably, modified coconut husk and activated carbon produced from agricultural waste have shown impressive ammonia removal efficiencies reaching up to 88.6%. The implications of this research are considerable. Incorporating these natural adsorbents into wastewater treatment strategies not only improves ammonia removal but also supports sustainability by utilizing agricultural and industrial by-products, thus addressing both pollution and waste management issues. Additionally, this review underscores the importance of aligning these practices with environmental regulations of Malaysia. Future research should concentrate on optimizing adsorption conditions, investigating the regeneration capabilities of these natural materials, and developing hybrid treatment systems to enhance nitrogen removal efficiency in various wastewater contexts. By leveraging local resources and promoting environmentally friendly solutions, Malaysia can advance towards a more sustainable approach to managing industrial wastewater and mitigating the effects of ammonia pollution on sensitive ecosystems.

Keywords: ammonia removal, wastewater treatment, natural adsorbents, coconut husk, palm oil biomass

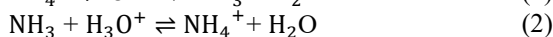
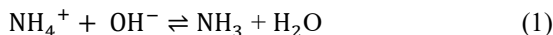
Introduction

Over the past century, inadequate disposal practices within industrial and agricultural sectors have inflicted severe social, economic and environmental repercussions [1,2,3]. Rampant discharge of domestic and industrial waste into natural ecosystems has significantly heightened the contamination of surface and groundwater reservoirs. Notably, a prevalent constituent within most waste materials is nitrogen [1]. The industrial sectors in Malaysia significantly contribute to nitrogen pollution in wastewater impacting both environmental and human health. Various industries including agriculture, manufacturing and aquaculture release effluents rich

in nitrogen compounds leading to serious ecological consequences [4]. For instance, rubber industrial wastewater containing high ammonia levels can cause eutrophication in water bodies [5]. Besides, the rapid expansion of aquaculture has led to significant wastewater generation containing high levels of nutrients and organic materials further stressing water quality [6]. This excess of nitrogenous nutrients has the potential to induce eutrophication, a process that engenders toxic algal blooms, fish mortality, and a myriad of associated issues [7,8].

The primary nitrogenous component commonly encountered in wastewater is ammonia [1], which

typically exists in the form of ammonia (NH_3) and ammonium (NH_4^+). The chemical equilibrium between ammonium and ammonia is represented by Equations (1) and (2).



Ammonia, characterized as a colorless, poisonous, reactive, and corrosive gas, emits a pungent odor. According to the Malaysian Department of Environment, ammonia presents a substantial health hazard, given its corrosive impact on the skin, eyes, and respiratory system [9]. Inhaling air with high concentrations of ammonia immediately causes burning sensations in the nasal passages, throat, and lungs [10]. The Permissible Exposure Limit (PEL) for ammonia is set at a mere 25 parts per million (ppm) to ensure the safety of workers. In contrast, exposure to 300 ppm is deemed immediately dangerous to life and health [9].

Elevated levels of total ammonium nitrogen (TAN) serve as a catalyst for increased algae proliferation and worsening eutrophication by furnishing algae and other aquatic plants with abundant nutrients [11]. The

ensuing surge in algal growth engenders diminished water clarity, oxygen depletion and disturbances in the delicate ecological equilibrium of aquatic ecosystems [11] illustrated in **Figure 1**. In Malaysia, lakes and reservoirs constitute significant water resources vital for the nation's socioeconomic development. These water bodies not only provide freshwater habitats and natural flood mitigation but also serve as ecotourism and recreational destinations [12,13,14]. However, nitrogen pollution primarily arises from several key sources, notably palm oil mills, agricultural practices and industrial manufacturing. The expansion of oil palm plantations has been linked to increased emissions of nitrogen oxides and volatile organic compounds, which contribute to ground-level ozone formation, posing risks to health and the environment [15]. Additionally, palm oil mill effluent (POME) is a significant source of nitrogen, as it contains high levels of organic nutrients and contributes to nitrogen loading in surrounding ecosystems [16,17]. Agricultural practices, particularly the use of fertilizers, further exacerbate nitrogen pollution, disrupting the nitrogen cycle and leading to increased emissions of nitrous oxide, a potent greenhouse gas [18]. Moreover, industrial activities contribute to nitrogen dioxide emissions, which are part of Malaysia's broader greenhouse gas profile [19].

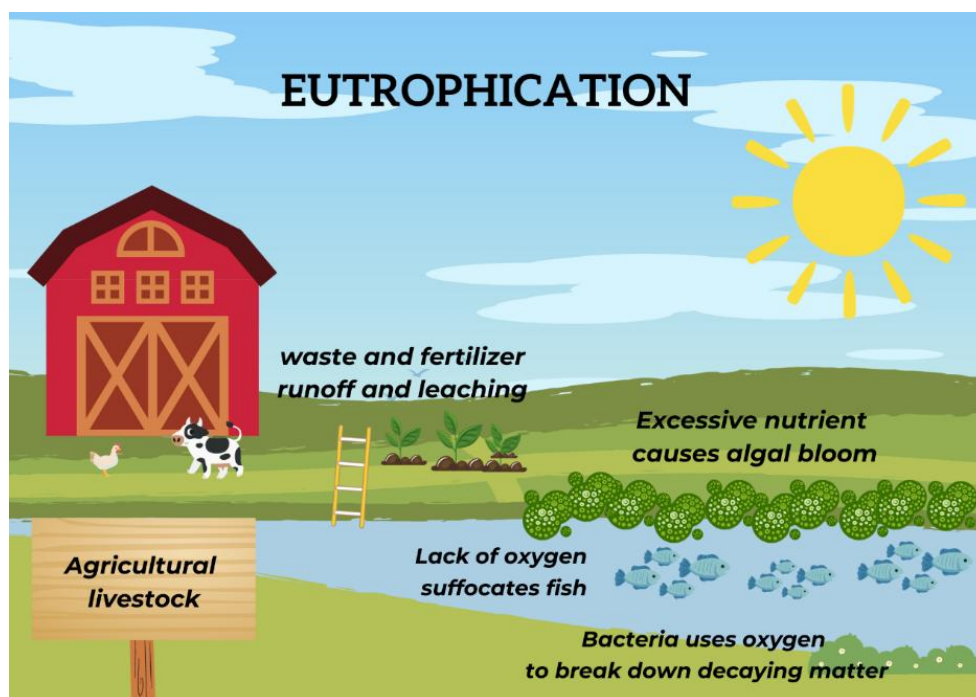


Figure 1. Eutrophication process

In Malaysia, conventional methods for ammonia removal from industrial wastewater such as extended aeration systems and sequencing batch reactors, face significant limitations, including high operational costs and inefficiencies in treating ammonia-nitrogen. While sewage treatment plants (STPs) have shown effectiveness in removing organic pollutants, their nitrogen removal capabilities are often inadequate, leading to non-compliance with the stringent Environmental Quality Act standards [20]. Additionally, the increasing nitrogen deposition from agricultural activities, particularly from palm oil plantations, exacerbates the challenge of managing nitrogen levels in wastewater [21]. The reliance on traditional methods may not sufficiently address the indirect emissions of nitrogen compounds, such as N_2O from drainage systems in oil palm plantations, which further complicates mitigation efforts [22]. Therefore, there is a pressing need for innovative and cost-effective treatment technologies to enhance nitrogen removal efficiency in Malaysia's industrial wastewater management [23]. Collectively, these sources highlight the urgent need for effective nitrogen management strategies to mitigate pollution in Malaysia.

Despite the pressing need for effective ammonia removal strategies, there remains a notable research gap in the applicability of various natural adsorbents for this purpose within the Malaysian context. Current studies largely lack comprehensive assessments of the effectiveness of locally available materials such as coconut husk, palm oil biomass and biochar in treating ammonia-rich wastewater. Moreover, there is insufficient exploration into optimizing the operational parameters that govern adsorption efficiency which could further enhance the capabilities of these natural adsorbents. Additionally, there is a crucial need for research that evaluates the long-term performance and regeneration potential of these materials as sustainable application is essential for practical wastewater management solutions. The exploration of innovative treatment methods and the integration of hybrid systems combining natural adsorbents with existing technologies could significantly improve nitrogen removal efficiencies. Natural adsorbents offer significant benefits for ammonia mitigation in Malaysia particularly in wastewater treatment. Research indicates that materials such as luffa, coconut husk and banana trunk fiber when combined with chitosan, can effectively reduce ammonia levels by up to 90% in kitchen wastewater [24,25]. Additionally, natural zeolites, characterized by their porous structure, have shown high affinity for ammonium ions, achieving substantial adsorption rates and making them a cost-effective solution for treating ammonia-contaminated

water [26,27]. The use of biological processes such as employing a mixed bacterial consortium in sand filters, has also demonstrated a remarkable 96-98% reduction in ammonia levels, showcasing the potential of biological treatment methods [28]. Finally, a detailed economic analysis is necessary to assess the cost-effectiveness of utilizing natural adsorbents compared to conventional methods. This will inform practical implementations in wastewater treatment facilities equipped to handle Malaysia's unique industrial challenges.

This review underscores the critical need for integrating natural adsorbents into wastewater management strategies to mitigate ammonia pollution and safeguard aquatic ecosystems in Malaysia while addressing these research gaps.

Nitrogen species in industrial wastewater

Nitrogen compounds are essential components in various biological and environmental processes, with key types including ammonia (NH_4^+), nitrite (NO_2^-), and nitrate (NO_3^-). Ammonia is a crucial raw material for synthesizing nitrogen-containing organic compounds, such as amino acids and nucleic acids, which are vital for life [29,30]. In aquatic ecosystems, the concentration of these nitrogen species can vary significantly; for instance, studies have shown that ammonium nitrogen often exceeds permissible levels in various water bodies due to agricultural runoff and sewage [31]. Nitrites, while typically present in lower concentrations can indicate pollution and are subject to strict regulatory limits, as excessive levels can lead to health issues like methemoglobinemia [32]. Nitrates, on the other hand, are generally found within acceptable limits but can contribute to environmental problems like eutrophication when present in high concentrations [33]. Overall, the balance and concentration of these nitrogen compounds are critical for both ecological health and human safety.

Removing nitrogen pollutants from industrial wastewater presents several challenges, primarily due to the complexity of the wastewater composition and the inefficiencies of existing treatment processes. One significant issue is the accumulation of nitrate nitrogen (NO_3-N) byproducts, which adversely affects nitrogen removal efficiency (NRE) in systems like anammox, particularly when nitrite-oxidizing bacteria (NOB) are not effectively inhibited [34]. Additionally, the slow growth rate of anaerobic ammonium oxidizing bacteria (AnAOB) and their sensitivity to operational conditions complicate the application of the anammox process [35]. The integration of partial nitrification and denitrification processes can enhance nitrogen removal, but achieving stable performance remains a challenge [36,

37]. Furthermore, the addition of exogenous compounds such as glycine betaine has shown promise in improving reaction rates and microbial stability yet the requirement for carbon sources still poses limitations [37].

In Malaysia, the regulatory standards for nitrogen levels in wastewater are primarily governed by the Environmental Quality Act (EQA) 1974, which sets forth Standard B for effluent discharges. This standard mandate that wastewater treatment facilities must effectively reduce nitrogen compounds, particularly ammonia-nitrogen, to protect water quality and public health [38]. Recent studies have shown that various sewage treatment plants, such as those in Klang Valley, have been successful in meeting these standards by employing advanced treatment methods like extended aeration systems and sequencing batch reactors, which facilitate significant nitrogen removal [39,40]. However, some facilities still struggle to comply, particularly those treating effluents from industrial sources like chemical and petrochemical plants, which often exceed permissible nitrogen levels [38].

Natural adsorbents for ammonia removal

Natural adsorbents are materials sourced directly from nature that possess innate adsorption properties. Examples include zeolite, agricultural waste materials, clays, biochar, algae and microbial biomass, and geopolymer materials. These materials have inherent adsorption capabilities due to their structure or composition and are effective at trapping pollutants from water or air. In the context of the ion exchange interaction between adsorbent and ammonia (NH_3), a stoichiometry equation can be formulated to represent the adsorption process. Equation (3) illustrates a simplified stoichiometry for the interaction between the adsorbent and NH_3 .



The equation encapsulates the essence of the ion exchange interaction between a general adsorbent and ammonia, wherein the ammonia molecules interact with the adsorbent surface through an exchange process, leading to the formation of an adsorbed complex. However, adsorbents adsorption capacities might be limited or less specific to certain pollutants. Modified adsorbents, on the other hand, are engineered to have enhanced adsorption properties making them more effective and efficient in capturing specific contaminants especially ammonia. While modified adsorbents might be more tailored and efficient, the processes involved in modifying them

could add to the production cost. The choice between the two depends on the specific needs of the application, considering factors such as pollutant type, concentration, and treatment objectives.

Criteria for an effective natural adsorbent

An effective natural adsorbent should possess several key criteria, particularly in terms of adsorption capacity (**Figure 2**). Firstly, the surface area of the adsorbent is crucial; for instance, natural zeolite modified with HCl exhibited an increase in surface area from $19.118 \text{ m}^2/\text{g}$ to $57.838 \text{ m}^2/\text{g}$, enhancing its capacity to adsorb contaminants like methylene blue and lead ions [41]. Additionally, adsorption kinetics are important. Studies have shown that pseudo-second-order models often best describe the adsorption processes, indicating a high efficiency in contaminant removal [42]. Furthermore, the choice of adsorbent material significantly impacts performance. For example, animal bones demonstrated a superior oil removal capacity compared to anise residues, achieving a 94% removal rate [43].

Low-cost materials such as animal bones, anise residues, fish scales, and various plant materials (e.g., neem, rice husk, and citron peel) have been shown to effectively remove contaminants from water, including oil, dyes, and heavy metals [44,45,46]. The use of these materials not only reduces the financial burden associated with wastewater treatment but also promotes environmental sustainability by utilizing waste products. For instance, fish scales, often discarded, can be treated to enhance their adsorption properties, making them a viable and economical option for treating tannery effluents [46]. Optimal conditions such as pH and contact time are vital, with specific studies indicating that a pH of around 4.0 maximizes adsorption efficiency for certain pollutants [47].

The availability of natural adsorbents is also a crucial criterion for their effectiveness in pollutant removal. Natural adsorbents, such as chitin and chitosan derived from crustacean shells, are not only abundant but also renewable, making them a sustainable choice for environmental remediation [48]. Additionally, materials like animal bones and anise residues have been identified as effective adsorbents for oil removal, showcasing the potential of readily available organic waste products. The use of biomass materials, such as peanut shells, further emphasizes the accessibility of natural adsorbents, as they can be sourced from agricultural byproducts [49].

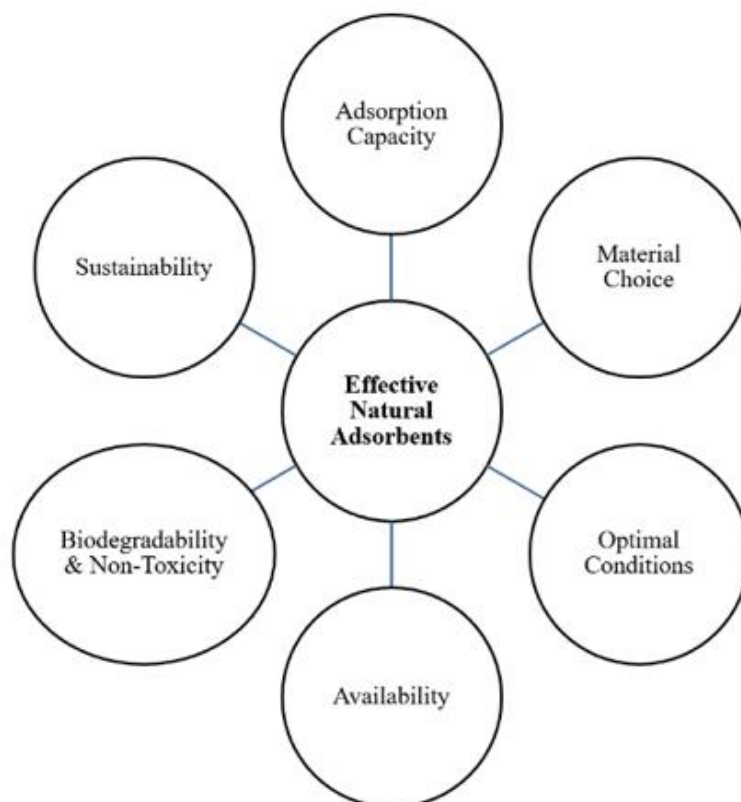


Figure 2. Criteria for an effective natural adsorbent

An effective natural adsorbent should also possess excellent regeneration potential. The ability to undergo multiple adsorption-desorption cycles without significant loss of efficiency is vital; for instance, natural pumice maintained its performance over three cycles with a 54.79% removal efficiency for nitrate [50]. Additionally, the regeneration method should be efficient and environmentally friendly; photochemical regeneration of iron oxide achieved a remarkable 96% efficiency after three cycles [51]. Furthermore, the choice of regeneration technique, whether thermal or solvent-assisted, impacts the adsorbent's structural integrity and performance, highlighting the need for a balance between effective adsorption and desorption capabilities [52].

The adsorbent should also be biodegradable and non-toxic, ensuring that it does not contribute to further pollution or harm ecosystems [53]. Additionally, the use of low-cost materials, such as natural fibers, zeolites, and agricultural by-products, can reduce the economic burden associated with water purification [54,55]. The adsorbent should facilitate easy regeneration or disposal to prevent accumulation of waste and promote sustainability in water treatment processes.

Common natural adsorbents studied for nitrogen removal

Zeolite

Zeolite is another category of natural adsorbents that are microporous, aluminosilicate minerals with a highly regular structure of pores and chambers [56, 57,58]. Based on their origin, zeolites are divided into two main categories: synthetic zeolites, which may be produced in a lab, and natural zeolites, which are extracted from the ground. Since natural zeolites were created by the chemical interactions of volcanic ash and alkaline water, they are mainly found in sedimentary rocks that were generated by volcanic eruptions. There are at least 60 species of natural zeolites that are known to exist and can be found naturally in rocks, sediments, and soils, with a concentration of these species found mostly in volcanic-derived rocks and soils [59] analcime, chabazite, clinoptilolite, erionite, mordenite, and phillipsite are the most prevalent naturally occurring zeolites [60, 61].

The morphology of zeolite plays a crucial role in its ability to adsorb NH_4^+ ions from aqueous solutions. Finer particles of zeolite were found to have better removal efficiency for NH_4^+ ions compared to coarser particles, especially at lower

initial concentrations. Smaller and finer particles allow for easier exchange of ammonium cations in the aqueous solution due to larger contact area provided by finer particles. This optimized contact area ensures a more effective exchange of ammonium cations, enhancing the overall removal efficiency of NH_4^+ ions by the zeolite. The morphology can also be altered through chemical modification method such as acid treatment [60,62,63], alkaline treatment [60], hydrothermal treatment [64], or ion exchange [65] which can enhance the sorption capacity of zeolite by improving its surface properties and ion exchange capabilities.

Zeolite has a high cation-exchange capacity and porous structure, which provides a large surface area for adsorption [66]. In recent years, industrial companies in Malaysia have increasingly recognized the need to address ammonia pollution, particularly in sectors such as palm oil processing, aquaculture and wastewater treatment. These industries are significant contributors to ammonia emissions which pose environmental and health risks. As a response, various initiatives have been undertaken to mitigate ammonia pollution and there is a growing interest in the use of natural adsorbent. The conventional ponding system for treating palm oil mill effluent (POME) involves a series of ponds where the effluent undergoes natural biological processes, including anaerobic, aerobic, and facultative treatments [67]. This method, while widely used, often results in high concentrations of chemical oxygen demand (COD) and biological oxygen demand (BOD) in the effluent, which can lead to significant environmental pollution if not adequately treated before discharge. In contrast, the COD concentration in various ponds decreased when treated with zeolite, with Pond 5 showing a removal percentage of 54.6% compared to lower reductions in the conventional method [67]. Additionally, the BOD concentration was notably high in untreated POME, but the application of zeolite helped in reducing these levels effectively [67]. For instance, several palm oil mills have begun to explore the use of natural zeolite in their effluent treatment systems [66, 68].

Another study conducted in Malaysia at the United Oil Palm (UOP) facility focused on mitigating ammonia nitrogen (AN) in palm oil mill effluent (POME) using zeolite and domestic wastewater (DWW) in a sequencing batch reactor (SBR) [68]. The research demonstrated that the combination of zeolite and DWW significantly enhanced the removal efficiency of ammonia, achieving removal rates between 96.19% and 98.30% [68]. The optimal conditions for ammonia removal were identified as a contact time of 18.32 hours, an aeration rate of 4.81 liters/min, and a DWW to POME ratio of 76.97% [68]. The study highlighted

the effectiveness of using zeolite, which facilitates microbial biofilm formation, thereby improving the biological degradation of ammonia. The study on heat-treated zeolite as an adsorbent for POME also revealed significant findings regarding its effectiveness in removing ammonia from the wastewater [69]. The research indicated that heat-treated natural zeolite, subjected to thermal treatment at 150°C for 2 hours, demonstrated superior adsorption capabilities compared to untreated zeolite [69]. Although specific removal efficiencies for ammonia were not detailed in the provided excerpts, the overall results suggested that the heat-treated zeolite could effectively reduce various pollutants, including ammonia, contributing to improved water quality. The optimal conditions for the adsorption process were identified as an adsorbent dosage of 4 g/L and a pH of 4, which likely facilitated enhanced ammonia removal [69].

Different types of zeolitic waste have been studied at an initial concentration of 1 mg/L and provided insights into the effectiveness of these materials as adsorbents for ammonium ions [70]. Modified zeolitic waste with hydrogen peroxide (H_2O_2) solution exhibited higher removal efficiency (47%) compared to unmodified zeolitic waste (33%) [70]. **Figure 3** illustrates the surface morphology of synthetic zeolite revealing a rough and highly structured texture compared to fly ash [71]. This distinctive surface characteristic shows more intricate and organized crystalline structure which plays a crucial role in enhancing the adsorption capabilities of the material. The synthetic zeolite grains are neatly arranged and exhibit well-defined outlines indicating a highly ordered crystal lattice [71]. This structured arrangement not only contributes to the stability of the material but also enhances its ion exchange efficiency. A particularly significant feature observed in the SEM images of synthetic zeolite is the presence of numerous dense holes and interconnected channels across the surface [71]. These porous structures are critical in increasing the overall surface area of the zeolite, which in turn provides more active sites for NH_4^+ ions adsorption. The higher the surface area, the greater the number of interaction sites available for ion exchange thereby improving the efficiency of ammonium removal from aqueous solutions. Additionally, these channels facilitate the diffusion of NH_4^+ ions into the inner layers of the zeolite allowing for deeper penetration and a more extensive adsorption process compared to materials with limited porosity such as fly ash.

The adsorption mechanism of ammonium ions onto synthetic zeolite is primarily governed by ion exchange, as illustrated in **Figure 4**. Initially, the

zeolite structure contains exchangeable cations (denoted as M^+), which are loosely bound within its porous framework. When the zeolite comes into contact with an aqueous solution containing NH_4^+ ions, a cation-exchange process occurs whereby the NH_4^+ ions in solution gradually replace the pre-existing M^+ cations in the zeolite lattice. This exchange leads to the effective capture and retention of NH_4^+ ions within the zeolite structure while the displaced cations (M^+) are released into the surrounding solution.

Due to zeolite ion exchange properties, zeolite can also be regenerated allowing for the efficient recovery of adsorbed ions such as ammonium and enabling the reuse of zeolite beds in wastewater treatment systems. Studies have shown that zeolite can be regenerated multiple times without compromising its performance, with some estimates suggesting regeneration cycles as high as 10-20 before needing replacement [72]. The regeneration efficiency can be improved by optimizing parameters such as regenerant contact time, which was shown to increase zeolite regeneration efficiency from 76% to 96% [72]

Overall, zeolites offer promising potential as NH_4^+ ion adsorbents.

In Malaysia, several industries are utilizing advanced water and wastewater treatment systems, and some incorporate zeolite in their processes. Techkem Water Technologies, for instance, is known for offering innovative wastewater solutions to industries like the rubber glove sector. Although their focus is not solely on zeolite, their treatment systems, such as Sequential Batch Reactors (SBR) and submerged fixed bed bioreactors, could incorporate various adsorbents, including zeolite, which is commonly used for ammonia and heavy metal removal in tertiary treatment [73]. Zeolite is particularly valued for its high cation exchange capacity, making it effective in removing ammonia, heavy metals, and other contaminants. It has been widely used for both drinking water and wastewater treatment due to its adsorption and ion-exchange properties. However, the specific companies exclusively relying on zeolite in Malaysia are not always explicitly mentioned in industry news.

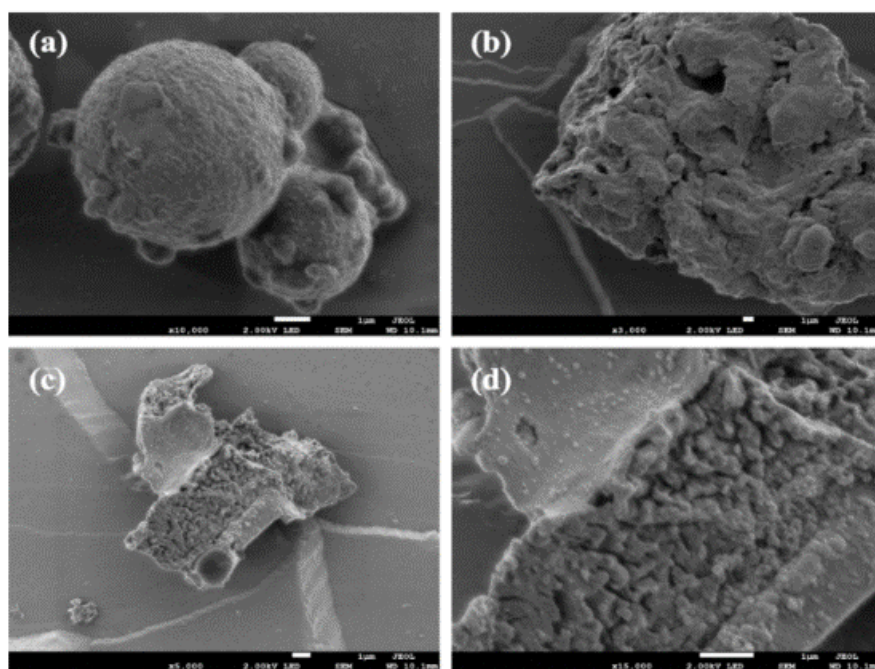


Figure 3. Scanning electron microscopy (SEM) of coal fly ash and synthetic zeolite: (a) coal fly ash; (b–d) synthetic zeolite [71]

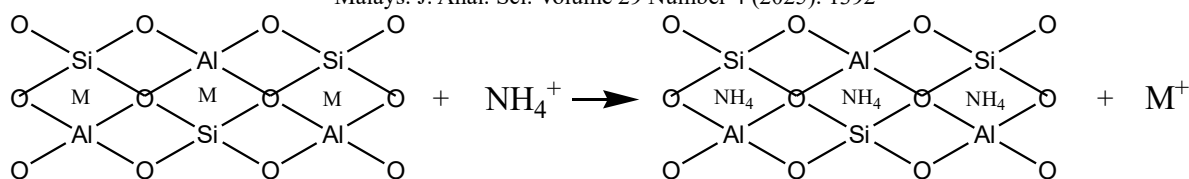


Figure 4. Ion exchange process in zeolite

Agricultural waste materials

Agricultural waste materials in Malaysia such as corncob, rice husk, and oil palm empty fruit bunch (OPEFB), represent a significant resource, with millions of tons produced annually [74]. These lignocellulosic materials possess various functional groups that can effectively bind heavy metals and other contaminants, making them suitable for biosorption applications [75].

Coconut husk, a natural adsorbent derived from agricultural waste materials has shown promise in the removal of nitrogen from wastewater. When the coconut husk is modified or used in its native form, it exhibits high adsorption capacities for various contaminants, including nitrogenous compounds. Research indicates that the chemical modifications of coconut husk enhance its surface area and porosity, leading to improved adsorption efficiency. For instance, stearic acid grafted coconut husk demonstrated significant oil sorption capabilities, which can be extrapolated to nitrogen removal due to its structural properties [76]. Additionally, the valorization of coconut waste has been highlighted as a sustainable approach for treating contaminated water, emphasizing the potential of coconut by-products in environmental remediation [77]. Furthermore, studies on the adsorption kinetics and isotherms suggest that coconut-based adsorbents can effectively remove nitrogen, aligning with the need for eco-friendly solutions in wastewater treatment [78].

Other local agricultural waste including palm oil biomass have shown potential as low-cost adsorbents for ammonia nitrogen, with adsorption capacities varying significantly; for instance, some materials achieved capacities up to 3.58 mg/g [79]. The effectiveness of these biosorbents can be influenced by factors such as initial ammonia concentration, pH, and stirring rate, with optimal conditions typically around pH 7 [79]. Additionally, the use of composite materials, such as clay/biochar, has demonstrated high removal efficiency, reaching up to 88.6% under specific conditions [80]. Furthermore, the modification of adsorbents can enhance their capacity for ammonia retention, which is crucial for

maximizing removal efficiency. Overall, utilizing palm oil biomass in conjunction with other agricultural wastes could provide an effective and sustainable solution for ammonia nitrogen removal from wastewater.

Activated carbon derived from agricultural waste, such as coconut shells and cow bone, has also been explored for the removal of Ammonia Nitrogen ($\text{NH}_3\text{-N}$) from Palm Oil Mill Effluent (POME) [81]. Activated carbon (AC) is renowned for its high surface area and exceptional adsorption properties, effectively removes various pollutants, including organic compounds and heavy metals. The surface of activated carbon contains various functional groups (e.g., carboxyl, hydroxyl, carbonyl, amino) that exhibit good affinity towards heavy metal cations, enhancing adsorption capacity [82,83]. Under hydrophobic conditions, the optimum reduction of $\text{NH}_3\text{-N}$ was achieved at a volumetric dosage ratio of 15:25 cm^3 of activated cow bone powder (ACBP) to activated coconut shell carbon (ACSC), resulting in a 48.2% removal efficiency [81]. In contrast, under hydrophilic conditions, the best performance was observed at a dosage of 35 cm^3 , which led to a 65.4% removal of $\text{NH}_3\text{-N}$ [81]. When combining hydrophobic and hydrophilic conditions, the optimal ratio of 25:15 cm^3 yielded a 68.5% reduction in $\text{NH}_3\text{-N}$ [81]. The study also highlighted that the adsorption process followed the Langmuir isotherm model, indicating that the adsorption occurred on a monolayer surface, suggesting a homogeneous distribution of adsorption sites. Additionally, the kinetic data fitted better to the pseudo-second-order model, indicating that the adsorption mechanism was chemisorptive. These findings underscore the potential of using natural adsorbents for effective ammonia removal from POME, with specific conditions enhancing their performance.

In the investigation on the utilization of activated carbon for mitigating ammonia emissions during composting digestate from food waste, AC emerged as a potent tool in reducing ammonia emissions through adsorption mechanisms. Notably, the addition of activated carbon led to a remarkable 34% reduction in

NH₃ emissions during the composting process. This reduction was achieved through the effective adsorption of both NH₃ and NH₄⁺ by activated carbon, thereby curbing the release of ammonia into the atmosphere. Moreover, the presence of activated carbon facilitated the accelerated degradation of protein-like organic matter, consequently resulting in higher NH₃ content in the initial stages of composting. However, this expedited degradation ultimately contributed to an overall reduction in ammonia emissions. The study revealed that the NH₃ and NH₄⁺ produced during composting were adsorbed by the activated carbon, leading to a notable decrease in NH₃ emissions.

Recent research efforts have also focused on enhancing the efficiency of activated carbon as an adsorbent for ammonia removal by modifying the adsorbent. A study found that with a gas space velocity of 900/h and a total inlet concentration of 550-650 mg/m³ at 50°C, the adsorption capacity of NH₃ for the modified activated carbon (MAC) was 24.17 mg/g [84]. The combination of high-pressure hydrothermal modification followed by metal salt solution impregnation modification led to the optimal adsorption performance for NH₃, highlighting the importance of modification techniques in enhancing the adsorption capacity of activated carbon for removing ammonia.

A study that compares the efficiency of ammonia removal between Biological Ion Exchange (BIEX) and Biological Activated Carbon (BAC) filters found that BIEX had a similar efficiency to BAC in removing ammonia [85]. However, the BIEX filter released 15% of the ammonia in warm waters due to factors like the small column diameter affecting backwash effectiveness [85]. The ammonia release in the BIEX filter mainly originated from the top 10 cm layer where most Natural Organic Matter (NOM) was removed [85]. During backwash, inadequate air injections failed to break down the solid biomass layer crust, leading to ammonia release. When compared to other filters like Granular Activated Carbon (GAC) and BAC, BIEX showed varying results in releasing ammonia emphasizing the significance of operational factors in ammonia removal efficiency [85]. Despite challenges in ammonia release, BIEX demonstrated good long-term performance in removing ammonia.

Charcoal is a carbon material derived from natural sources. It is commonly used as a raw material for producing activated carbon which is an important adsorbent in various industries and research applications [86,87,88,89]. Another study showed rice husk charcoal emerged as the most efficient boasting an impressive maximum removal rate of 96.8% [90].

However, despite inherent capabilities of charcoal in ammonia removal, it inherently possesses a lower surface area and fewer functional groups compared to activated carbon, thereby constraining its adsorption capacity for certain molecules like ammonia. Consequently, modification becomes imperative to enhance its efficacy in ammonia removal. A study employed the HNO₃/H₃PO₄-NaNO₂ system to oxidize charcoal resulting in modified charcoal (MC) with slightly lower carbon and hydrogen content yet higher nitrogen and oxygen content compared to the raw material [91]. This increase in oxygen content, attributed to the formation of oxygen-containing groups like carboxyl or carbonyl groups, facilitates the introduction of more functional groups, thereby enhancing surface area and porosity resulting in MC to exhibit a noteworthy ammonia adsorption capacity of 6.19 mg/g. Moreover, BET analysis showcased a substantial augmentation in the surface area of MC from 35.5 m²/g for charcoal to 157.1 m²/g, indicating heightened efficacy in entrapping and retaining ammonia molecules for enhanced removal [91]. The significant reduction in nonionized NH₃ concentrations in rearing water upon supplementation with wood charcoal (WC) and activated charcoal (AC) [92]. Specifically, fish fed an experimental diet containing 20 grams of WC per kilogram of feed, and fish fed with a similar dosage of AC exhibited the most effective reduction in NH₃ levels, plummeting to as low as 3.55 ppb and 3.13 ppb, respectively, from an initial concentration of 5.97 ppb [92]. This notable decline underscores the remarkable effectiveness of charcoal in ammonia removal.

Overall, AC serves as a formidable adsorbent for ammonia removal, owing to its exceptional adsorption characteristics and versatility across various environmental scenarios. Its facile modifiability and capacity for chemical compositional adjustments augment its adsorption efficiency, positioning it as a versatile and potent solution for addressing ammonia contamination challenges.

Clays

Bentonite, a clay mineral primarily composed of montmorillonite, has shown significant potential as a natural adsorbent for the removal of ammonia nitrogen from wastewater. Its high cation exchange capacity and reactivity allow for effective adsorption of ammonia, with studies indicating that bentonite can achieve ammonia removal capacities of up to 11.6 mg/g when combined with chitosan [93]. The adsorption process is influenced by factors such as pH, temperature, and contact time, with optimal conditions typically found at pH 6 [93]. Additionally, bentonite has been observed to mitigate ammonia inhibition in anaerobic digestion processes, enhancing

methane production rates and overall process resilience [94]. The findings regarding the use of bentonite in anaerobic digestion processes can also have significant implications for wastewater treatment. The ability of bentonite to mitigate ammonia inhibition can enhance the efficiency of anaerobic treatment systems commonly used for treating wastewater, particularly those with high nitrogen content. In wastewater treatment, ammonia is a common pollutant that can inhibit the activity of methanogenic bacteria, which are crucial for the breakdown of organic matter and the production of biogas.

By incorporating bentonite into anaerobic reactors treating wastewater, the onset of ammonia toxicity can be delayed, allowing for higher ammonia concentrations to be processed without adversely affecting methane production. This is particularly beneficial in scenarios where wastewater streams contain elevated levels of ammonia, such as those from agricultural runoff or industrial effluents. The addition of bentonite can shorten the lag phase associated with microbial adaptation to these high ammonia levels, leading to quicker stabilization of the digestion process and enhanced methane production rates. Moreover, bentonite's properties may contribute to the overall stabilization of the anaerobic digestion process, helping to maintain operational efficiency even under fluctuating conditions. The presence of cations in bentonite could further assist in counteracting ammonia's inhibitory effects, thereby promoting a healthier microbial community that is more resilient to toxic compounds.

In practical terms, the use of bentonite in wastewater treatment systems could lead to improved biogas yields, making the process more economically viable. Additionally, the stabilization of the digestion process can enhance the overall treatment efficiency, resulting in better removal of organic pollutants and reduced environmental impact. This approach could be particularly advantageous for wastewater treatment facilities looking to optimize their operations and increase biogas production while effectively managing nitrogenous pollutants. Overall, the integration of bentonite into wastewater treatment processes presents a promising strategy for enhancing treatment efficiency and sustainability. The versatility of bentonite, including its modification into various forms, further optimizes its adsorption properties for practical applications in wastewater treatment [93, 94].

Biochar

Biochar is a carbon-rich by-product obtained from the pyrolysis or gasification of biomass. It exhibits a

significant amount of ion exchange capacity, making it effective for removing ammonia [95]. The effectiveness of biochar in removing ammonia from water is primarily attributed to its high surface area, porous structure, and surface functional groups that facilitate the adsorption of ammonium ions. The type of biochar, production conditions, and potential modifications play crucial roles in optimizing biochar for enhanced removal of ammonia in water treatment applications. Unmodified biochars often exhibit low adsorption capacities for contaminants like ammonia, especially for anionic forms due to electrostatic repulsion. The majority of unmodified biochars may only weakly adsorb nitrogen, necessitating modifications to enhance their adsorption capabilities [95].

A study had synthesized nanoporous biochar from microalgae followed by KOH-activated high-temperature pyrolysis at different temperatures (650°C, 700°C, and 800°C) and among the biochars produced, NP-MBC-700 activated at 700°C exhibited the highest performance with a specific surface area exceeding 1100 m²/g. This biochar demonstrated a maximum ammonia removal of 72% within 120 minutes and an adsorption capacity of over 69 mg/g [96]. While NP-MBC-800 had an even higher specific surface area, surpassing 1700 m²/g, it showed reduced efficiency attributed to the damage caused to its textual characteristics at the higher pyrolysis temperature of 800°C. When biochar is subjected to high temperatures during pyrolysis, it can lead to changes in its physical and chemical properties, including pore structure, surface chemistry, and overall texture. In the case of NP-MBC-800, the high pyrolysis temperature caused damage to its textual characteristics, resulting in pore agglomeration and potentially altering the distribution and accessibility of pores within the biochar structure. As a result of these changes, the biochar may have lost some of its effective surface area for adsorption and its ability to interact with ammonia molecules in wastewater.

The choice of feedstock for biochar production is critical as it influences the physicochemical properties and adsorption capacities of the resulting biochar. Banana leaves represent an ideal feedstock due to their abundance in tropical regions especially in Malaysia, where banana cultivation is prevalent [97]. Utilizing banana leaves, an agricultural waste product, for biochar production not only reduces waste but also enhances the sustainability of the biochar production process. The research on biochar derived from banana leaves reveals its significant potential for ammonium ion (NH₄⁺) removal from aqueous solutions. The maximum adsorption capacity of the biochar was determined to be 0.97 mg/g in the absence of organic

compounds, with enhanced capacities observed in the presence of organic substances such as BSA, lactose, and acetic acid, reaching up to 1.874 mg/g for acetic acid [97]. The adsorption process was notably influenced by the pH of the solution, with optimal removal rates occurring at pH 9 where a maximum removal percentage of 25.45% was achieved using a biochar dose of 200 mg [97]. Furthermore, increasing the biochar dose correspondingly increased the amount of NH_4^+ adsorbed, exemplified by the removal of 0.34 mg/L of NH_4^+ with 500 mg of biochar at pH 9 [97].

Three different preparation methods of oil palm fiber (OPF) biochar had been evaluated for appropriate preparation method to optimize the ammonia removal efficiency: (1) pyrolysis, (2) acid activation before pyrolysis, and (3) activation after pyrolysis with oxidizing agents [98]. The first method, which involved direct pyrolysis of OPF at different temperatures and holding times achieved a maximum ammonia removal efficiency of 50% at 300°C and 2 hours [63]. Acid activation of raw OPF before pyrolysis did not show improvement in ammonia removal and led to leaching of ammonia, while activation after pyrolysis with oxidizing agents did not enhance ammonia removal efficiency compared to the first method [63]. Bamboo biochar (BB) was also produced through bamboo pyrolysis at 450°C in a nitrogen environment. At the same time, ball milled bamboo biochar (BMBB) was created by subjecting BB to ball milling, resulting in the development of oxygen-containing functional groups on its surface [63]. The research revealed that BMBB had a significantly higher ammonium adsorption capacity of 22.9 mg/g compared to the capacity of BB of 7.0 mg/g. The oxygen-containing functional groups, particularly the negatively charged ones formed during ball milling, played a vital role in enhancing BMBB's adsorption performance for ammonium. The oxygen-containing functional groups on the surface of biochar can include carboxyl ($-\text{COOH}$), hydroxyl ($-\text{OH}$), and carbonyl ($-\text{C}=\text{O}$) groups. These groups provide active sites for chemical interactions with contaminants like ammonium ions in water. The negatively charged functional groups, such as carboxyl and hydroxyl groups, are particularly important as they can attract and bind positively charged ions like ammonium through electrostatic interactions. The presence of these surface functional groups increases the surface area and porosity of biochar, providing more binding sites for contaminants. Moreover, the ball milling process not only boosted the adsorption capacity of BMBB but also accelerated the adsorption rate, making it a more effective adsorbent for ammonium removal from water [63]. The NaOH-modified ball-milled biochar

demonstrated a high ammonium sorption capacity of 8.93 mg/g [63]. Overall, the nanoporous biochars show promise as sustainable and environmentally friendly natural absorbents for ammonia removal in wastewater treatment.

Another biochar from rice straw produced at 550 °C with pH of Zero Point Charge was at pH 7.5 and had maximum ammonium adsorption capacity [99]. The adsorption-desorption of ammonium shown that rice straw biochar can run up to 5 cycles [99]. The fresh bamboo biochar (FBB) and degraded bamboo biochar (DBB) on NH_4^+ removal was also investigated and the adsorption capacity and percentage of NH_4^+ removal was 7.29 mg/g, 65.5% and 5.98 mg/g, 61.7%, respectively [100]. Both FBB, as well as DBB, can adsorb NH_4^+ . However, DBB may require a longer time to achieve equilibrium [100]. The adsorption of NH_4^+ on the FBB and DBB is a function of oxygen-containing functional groups, and physisorption is not the dominant mechanism [100].

Biochar produced by empty fruit bunch (EFB) also emerges as an efficient feedstock for producing biochar [101]. The adsorption capacity was found to be highest at a dosage of 0.05 g, achieving a capacity of 2.49 mg/g [101]. As the dosage increased to 0.25 g, the adsorption capacity significantly decreased to 0.46 mg/g [101]. This trend indicates that lower dosages of biochar provide more available active sites for ammonium adsorption. In comparison, higher dosages lead to competition among ammonium ions for these sites, ultimately reducing overall capacity. The optimal conditions for ammonium adsorption were identified as a biochar dosage of 0.05 g and a contact time of 200 minutes, with the best performance occurring at a neutral pH of around 7. Overall, the research demonstrates that EFB biochar is a promising and effective adsorbent for ammonium removal from wastewater, providing an environmentally friendly solution while enhancing the value of agricultural waste in Malaysia.

Biochar and activated carbon are both carbon-rich materials used in various environmental and industrial applications, but they differ significantly in their production processes, properties, and applications. Biochar is produced from organic materials such as agricultural waste, wood chips, and plant residues through pyrolysis at lower temperatures (400-700°C) without additional activation processes [102,103]. It has a lower surface area, is less porous, and contains more organic and volatile compounds. In contrast, activated carbon can be made from various carbon-rich materials, including wood, coconut shells, and peat [104,105]. It is produced by carbonizing the source material at high temperatures (600-1200°C)

followed by physical or chemical activation to enhance its surface area and porosity [106,107].

When comparing biochar and activated carbon for ammonia removal in wastewater, activated carbon is generally considered more effective. Activated carbon typically has a higher surface area, ranging from 500 to 1500 m²/g, which provides more sites for adsorption [108]. Its high porosity, with numerous micropores and mesopores, enhances its capacity to adsorb smaller molecules like ammonia [109]. The extensive surface area and porous structure make activated carbon highly efficient in removing ammonia and other contaminants from wastewater. In contrast, biochar has a lower surface area and is less porous, limiting its adsorption capacity. Additionally, the presence of organic and volatile compounds in biochar can interfere with its adsorption efficiency for ammonia [110]. Biochar is more commonly used for soil amendment, carbon sequestration, and immobilizing contaminants in soil rather than direct ammonia removal from wastewater [110]. Therefore, while both materials have their merits, activated carbon is better suited for ammonia removal in wastewater due to its superior adsorption capabilities. In contrast, biochar is more effective for applications related to soil health and environmental remediation.

Algae and microbial biomass

Algae are simple, photosynthetic organisms found in various aquatic environments. At the same time, microbial biomass refers to the mass of microorganisms, including bacteria, fungi, and algae, that can be utilized for various applications, including environmental remediation. Both algae and microbial biomass serve as effective biosorbents for the removal of heavy metals and other pollutants from wastewater. The biosorption process involves the uptake of metal ions through physical and chemical interactions with the biomass, utilizing reactive groups present on their surfaces, such as carboxyl and hydroxyl groups [111]. Studies have shown that non-living biomass, such as that derived from algal species, can exhibit significant metal binding capacities, outperforming traditional adsorbents like activated carbon [112,113]. Additionally, the use of waste biomass enhances the economic feasibility of biosorption processes, making it a sustainable option for treating contaminated water [112,114].

Algae and microbial biomass have shown significant potential as adsorbents for ammonia nitrogen removal in wastewater. For instance, the freshwater microalga *Chlorella* sp. demonstrated a high removal efficiency of total nitrogen (17.04-58.85%) and total phosphorus (62.43-97.08%) from municipal wastewater, indicating its effectiveness in nutrient

assimilation [115]. Similarly, *Arthrospira platensis* was evaluated for its ability to remove ammonium ions, achieving removal rates of 64.24% and 89.68% in synthetic solutions, and lower rates in real aquaculture effluents [115]. Additionally, duckweed (*Lemna gibba*) in waste stabilization ponds effectively absorbed nitrogen, contributing to its removal alongside sedimentation and denitrification processes [116].

Geopolymer materials

The study on ammonium (NH₄⁺) removal using a metakaolin-based geopolymer material incorporating granite wastes reveals several significant findings regarding its effectiveness in Malaysia's wastewater treatment [117]. The research demonstrated that the geopolymer achieved a remarkable maximum removal efficiency of 97% for NH₄⁺ during initial lab-scale experiments. Furthermore, in full-scale pilot plant tests, it achieved an 81% removal rate of NH₄⁺ from real wastewater streams, underscoring its practical applicability in the region. The tests were conducted on leachate from a landfill, specifically at the Xiloga S.L. landfill [117]. The leachate contained a high concentration of NH₄⁺ (1978 mg/L) and was characterized by a significant presence of suspended solids, organic matter, and possible competitive ions, making it a challenging wastewater stream for treatment. The adsorption capacity of the geopolymer was found to be 25.24 mg/g for NH₄⁺ [117], which is competitive with that of natural and some synthetic zeolites, indicating its potential for effective ion binding in wastewater applications. The adsorption mechanism was elucidated through the Redlich-Peterson isotherm model, which suggested that NH₄⁺ was adsorbed onto the geopolymer surface as a monolayer, characterized by strong physical-chemical interactions [117]. Kinetic analysis revealed that the process followed the Weber-Morris rate equation, with intraparticle diffusion identified as the limiting step [117], highlighting the importance of the geopolymer's internal structure in facilitating ion exchange.

However, several challenges must be addressed to implement this technology effectively in Malaysia's industrial wastewater treatment sector. One significant challenge is the issue of alkali leaching from the geopolymer [117], which can elevate pH levels in aqueous media, potentially leading to regulatory compliance issues. Additionally, the variability in wastewater composition, including the presence of suspended solids, organic matter, and competitive ions, may affect the performance of the geopolymer in the wastewater treatment plant [118]. Furthermore, the initial costs associated with scaling up the technology and the need for proper infrastructure to support its

implementation could pose financial barriers. Strategies to mitigate alkali leaching and adapt the geopolymer for diverse wastewater conditions will be crucial for enhancing its viability in Malaysia's industrial context.

Methods of ammonia removal treatment

Addressing ammonia pollution in wastewater poses significant challenges, necessitating the exploration of various methods for its removal. These methods can be broadly classified into three categories: physical, chemical and biological processes. Physical processes focus on non-chemical interactions and the inherent properties of materials to separate ammonia from wastewater. Chemical processes involve reactive mechanisms that alter chemical structure of ammonia and facilitate its removal. Finally, biological processes utilize the capabilities of microorganisms to degrade ammonia through natural metabolic pathways.

Physical processes

Adsorption

Adsorption technology has emerged as a promising method for ammonia removal from wastewater, leveraging various materials with distinct adsorption characteristics. The process of adsorption is when molecules of a gas or liquid (adsorbate) accumulate on the surface of a solid (adsorbent) as shown in **Figure 5**.

In this process, the adsorbent provides active sites that attract and hold the adsorbate molecules. Initially the adsorbate molecule moves randomly in the surrounding phase. When they come close to the adsorbent surface, they experience attractive forces that cause them to adhere. These forces can be Van der Waals forces (physisorption) or chemical bonding

(chemisorption). As more molecules settle, they form an adsorbed layer on the surface of the adsorbent. Depending on conditions such as pressure and temperature, the adsorption process can result in a monolayer (a single thick layer) or multilayer (several layers of molecules stacking up). The kinetics of adsorption describe how quickly and efficient adsorbate molecules accumulate on the adsorbent surface. This behaviour is commonly explained using adsorption isotherms which relate the quantity of adsorbed molecules to their concentration in the surrounding phase. The Langmuir isotherm describes monolayer adsorption assuming that all adsorption sites on the surface are identical and independent and that each site can hold only one molecule [119,120, 121]. The Langmuir equation expresses adsorption in Equation (4) where q_e is the amount of the adsorbate adsorbed per unit mass of adsorbent, Q_m is the maximum adsorption capacity, K_L is the Langmuir constant and C_e is the equilibrium concentration of the adsorbate. This model is commonly used for applications where adsorption reaches saturation at a certain concentration.

$$q_e = \frac{Q_m K_L C_e}{1 + K_L C_e} \quad (4)$$

In contrast, the Freundlich isotherm describes multilayer adsorption on a heterogeneous surface, where different adsorption sites have varying affinities for the adsorbate [121,122]. This model assumes that adsorption strength decreases as more molecules accumulate. The Freundlich equation is expressed in Equation (5)

$$q_e = K_F C_e^{1/n} \quad (5)$$

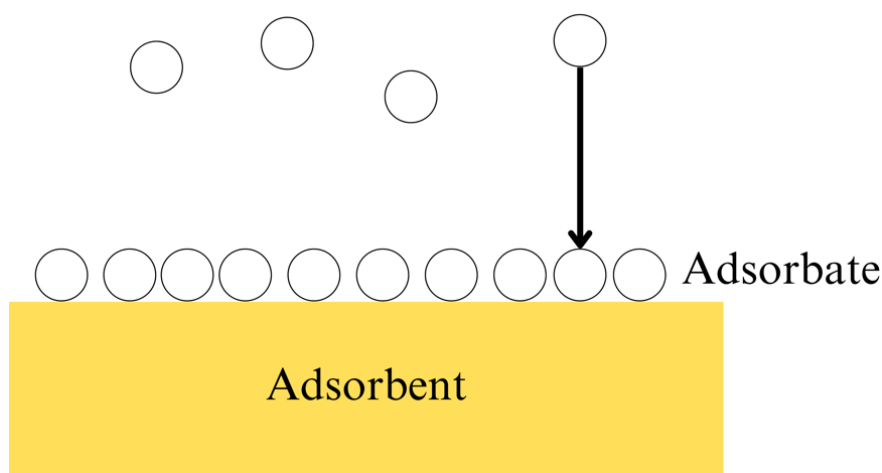


Figure 5. Adsorption mechanism

where K_F and n are constant that indicate the adsorption capacity and intensity. The Freundlich model is particularly useful for describing real-world adsorption processes where surface properties vary. A more advanced model, the BET isotherm (Brunauer Emmett Teller) extends the Langmuir model to account for multilayer adsorption. Unlike the Langmuir model, BET theory assumes that the first adsorbed layer behaves like monolayer adsorption but additional layers form through molecular interactions. The BET equation is expressed in Equation (6)

$$q_e = \frac{Q_m C_e C_B}{(C_s - C_e) \left(1 + \frac{(C_B - 1) C_e}{C_s}\right)} \quad (6)$$

where C_B is the BET constant and C_s is the saturation concentration. This model is widely used in surface science and materials engineering to determine surface area and pore size distribution. For instance, activated rice husk and magnetically modified excess sludge demonstrated high adsorption capacities with the former achieving over 90% removal efficiency [119,120]. Kinetic models, particularly the pseudo-second-order model, were frequently validated, suggesting that the rate of ammonia adsorption is influenced by the availability of active sites and the concentration of ammonia in solution [119,121].

Clinoptilolite, one of the most widely studied natural zeolites, exhibits a relatively moderate adsorption capacity with reported maximum value of 2.8 mg/g. In contrast, pre-treated Sarulla zeolite demonstrates an exceptionally high adsorption capacity, reaching up to 400 mg/g under optimized conditions [123,124,125]. This stark difference in adsorption performance underscores the influence of both intrinsic material properties and surface modifications in enhancing ammonia uptake. Kinetic analyses provide essential insights into rate-controlling mechanisms and adsorption dynamics. A predominant observation is that the adsorption of ammonium ions onto zeolites follows pseudo-second-order kinetics, suggesting that the adsorption rate is primarily governed by chemical interactions rather than simple physical diffusion [123,124,125]. The adherence to pseudo-second-order kinetics implies that the adsorption capacity is highly site-dependent, as the number of available active sites decreases, the adsorption rate slows down [71]. This means that when all active sites are occupied, the adsorption process reaches saturation, which is why the performance of zeolites can be significantly improved through surface modifications that introduce additional active sites or enhance existing ones. The presence of exchangeable cations (such as Na^+ , K^+ , and Ca^{2+}) in zeolite frameworks plays a crucial role in ion-exchange adsorption, further

confirming the chemisorption mechanism. One of the most effective ways to enhance the adsorption capacity of natural zeolites is through chemical modifications. Studies have demonstrated that ethylenediaminetetraacetic acid (EDTA) treatment can significantly improve adsorption efficiency by modifying the surface charge and increasing the number of available active sites [138]. EDTA, a strong chelating agent, interacts with the metal cations in the zeolite framework could increase the ion exchange capacity as the removal of certain metal ions creates vacancies that can be occupied by ammonium ions thereby enhancing adsorption [138]. Secondly, the reactivity is improved as the introduction of functional groups via EDTA treatment modifies the surface chemistry of the zeolite [138]. Lastly, the structural stability of the zeolite is preserved unlike some acidic or thermal modifications that may degrade the crystalline framework [71]. The adsorption kinetics of natural zeolites provide valuable insight into their potential for wastewater treatment. The pseudo-second-order model supports the hypothesis that ammonium adsorption is a chemisorption-driven process where the availability of active sites governs the overall efficiency.

Air stripping

Air stripping is a widely utilized technique for removing ammonia from wastewater by leveraging the natural process of volatilization. In this method, ammonia gas is transferred from the liquid phase to the gas phase and subsequently released into the atmosphere. Air stripping is particularly effective in treating wastewater with lower concentrations of ammonia such as that typically found in municipal effluent [127]. The efficiency of this method is influenced by several key operating parameters that must be carefully optimized to ensure effective ammonia removal. One critical factor in the efficiency of air stripping is the pH level of wastewater. Research has shown that higher pH levels specifically up to 12, significantly enhance ammonia volatilization [127]. At elevated pH, the equilibrium shifts favorably towards the gaseous form of ammonia (NH_3), resulting in increased volatility due to reduced solubility in water. Temperature is another important parameter affecting ammonia removal. Studies indicate that increasing the temperature, up to around $40.5 \pm 7.68^\circ\text{C}$, positively influences ammonia volatilization rates [127]. Higher temperatures reduce the solubility of gases in liquids thus favoring the transfer of ammonia from water to air. This relationship underscores the necessity of careful temperature management in wastewater treatment facilities to optimize operational efficiency.

The gas-to-liquid (G/L) ratio is also a critical factor that determines how effectively air can interact with the wastewater [128]. Optimal G/L ratios of 60:1 to 80:1 facilitate adequate contact between air and water, allowing for maximum ammonia transfer [128]. A well-adjusted G/L ratio ensures that sufficient air passes through the liquid, promoting effective removal of ammonia while minimizing unutilized air that could lead to energy inefficiencies [128]. Similarly, the flow rate of air introduced into the system has significant implications for both removal efficiency and energy use. Balancing air flow rates, typically between 5 L/min and 10 L/min, is essential to achieve an optimal removal rate while ensuring that energy consumption remains manageable [128]. Higher flow rates facilitate rapid ammonia stripping but can increase operational costs due to energy demands on blowers and fans. The air stripping process can achieve remarkably high removal efficiencies with studies indicating rates of up to 98% under optimal conditions [139,140]. This effectiveness is vital for addressing ammonia in wastewater prior to discharge, thereby protecting water quality and ecosystems from potential harms associated with nitrogen pollution. However, several challenges impede the widespread application of air stripping in wastewater treatment plants. One major concern is the significant energy consumption associated with operating blowers or fans that are necessary for air circulation. This energy requirement can amplify operational costs, particularly in large-scale facilities processing substantial volumes of wastewater. Moreover, the potential for stripping other volatile compounds alongside ammonia poses risks, as unintended pollutants may also be released into the atmosphere, necessitating strict regulatory compliance and monitoring. The applicability of the method is also constrained by the specific pH range (10-12) in which optimal operation occurs [127]. This requirement may limit its use in wastewater with more acidic or neutral pH levels presenting a considerable challenge for treatment facilities dealing with varying wastewater compositions.

To enhance the application and efficiency of air stripping in diverse wastewater treatment scenarios, future research should investigate broader operating conditions beyond the current pH and temperature limits. Exploring alternative configurations such as integrated systems combining air stripping with other treatment methods could facilitate improved ammonia removal while addressing operational challenges. Innovations in process control, including real-time monitoring and automation may further optimize the air stripping process, allowing for more responsive adjustments to changing wastewater characteristics.

Chemical processes

Coagulation/Flocculation

The coagulation process is initiated by the addition of coagulants, which are typically metal salts such as aluminum sulfate ($\text{Al}_2(\text{SO}_4)_3$), ferric chloride (FeCl_3) or calcium hydroxide ($\text{Ca}(\text{OH})_2$) [129], [130]. These coagulants work by neutralizing the surface charges of suspended particles and forming metal hydroxide precipitates that can trap ammonium ions. One of the key mechanisms involved in coagulation for ammonia removal is charge neutralization. Ammonia in wastewater exists in equilibrium between ammonium ions (NH_4^+) and free ammonia (NH_3) depending on the pH. When metal-based coagulants are introduced, they generate positively charged metal ions (Al^{3+} , Fe^{3+} or Ca^{2+}) that interact with negatively charged colloids and ammonium-associated compounds [129,130]. This interaction reduces the repulsion between particles allowing them to aggregate into larger settleable flocs.

Another important mechanism is sweeping coagulation where the hydrolysis of metal salts results in the formation of amorphous hydroxide precipitates, such as $\text{Al}(\text{OH})_3$ or $\text{Fe}(\text{OH})_3$ [129,130]. These precipitates create a sweeping effect, capturing fine suspended particles and ammonium compounds, thereby enhancing ammonia removal. Additionally, calcium-based coagulants such as lime ($\text{Ca}(\text{OH})_2$), promote the precipitation of ammonium ions as calcium ammonium phosphate, further reducing ammonia concentrations in wastewater. Interparticle bridging is also observed when polymeric coagulants are used [131,132]. Polymers with long molecular chains can bridge multiple particles together forming strong and stable flocs [131,132]. This mechanism enhances the removal efficiency of ammonia by ensuring that ammonium-containing particulates remain aggregated and are easily separated from the treated water.

Following coagulation, flocculation is employed to enhance the aggregation of destabilized particles into larger flocs [129]. This process involves slow and controlled mixing to facilitate particle collisions and floc growth. Two primary mechanisms drive flocculation: perikinetic and orthokinetic flocculation [133]. Perikinetic flocculation occurs at a microscopic level due to Brownian motion, where small particles randomly collide and form weakly bound microflocs. This process is effective for fine particles but requires additional mixing to encourage further aggregation. Orthokinetic flocculation, on the other hand, is driven by mechanical or hydraulic mixing, which induces controlled collisions between particles [133]. This method is particularly important for ammonia removal as it ensures the efficient aggregation of ammonium-

associated flocs into larger and more settleable masses. Flocculation also plays a crucial role in enhancing the removal of organic matter that binds with ammonium ions. Many wastewater streams contain dissolved organic compounds that form complexes with ammonia, which makes them difficult to remove through conventional precipitation. The use of flocculants such as polyacrylamides or natural biopolymers thereby helps to aggregate these organic-ammonium complexes into settleable flocs and improves overall ammonia removal efficiency [133].

Several factors influence the effectiveness of coagulation and flocculation in removing ammonia from wastewater. The type and dosage of coagulants play a critical role as insufficient coagulant addition may lead to incomplete charge neutralization. At the same time, excessive dosing can result in secondary pollution due to excess sludge formation. The pH of the wastewater is also a key factor, as it determines the equilibrium between ammonium ions (NH_4^+) and free ammonia (NH_3) [129]. For example, alkaline conditions ($\text{pH} > 9$) favor the conversion of ammonium to free ammonia, which can be more effectively removed through volatilization or adsorption onto coagulant precipitates [134]. Temperature influences the kinetics of coagulation and flocculation, with higher temperatures generally enhancing the rate of hydrolysis and floc formation [134]. However, in colder climates, the efficiency of these processes may decrease, necessitating longer retention times or higher coagulant dosages. Mixing intensity is another crucial factor, as rapid mixing during coagulation ensures uniform dispersion of coagulants, while gentle mixing during flocculation allows for the gradual formation of strong flocs. The presence of competing ions and natural organic matter (NOM) in wastewater can also affect ammonia removal. Organic matter may interfere with coagulant efficiency by forming stable metal-organic complexes, reducing the availability of coagulant ions for ammonium precipitation [134,135]. Pre-treatment methods, such as oxidation or adsorption can help mitigate this issue and improve coagulation performance.

A study also investigated the combined technique of enhanced coagulation and adsorption using nano-adsorbent (ACZ) for the efficient removal of ammonia from raw water [136]. The initially pre-acidified raw water to a pH of 6.2 to optimize the coagulation process. Enhanced coagulation with acidified alum was employed, reducing sulfate addition to the water and sludge production, achieving significant ammonia removal. The process of adsorption on ACZ was also utilized, achieving a remarkable 58% removal of ammonia in just 30 minutes [136]. The combined

treatment approach showcased high removal efficiency, with a noteworthy 76% removal of ammonia in a short treatment time of 20 minutes [136]. Additionally, the high performance in reducing dissolved organic carbon (DOC) levels underscores the potential of this method for drinking water treatment. The study concludes that the utilization of ACZ adsorption capacity within the short detention time provided by conventional water treatment plants offers a promising solution for efficient ammonia removal. The use of acidified alum not only reduces the sulfate addition but also aids in sludge reduction which is beneficial for overall water quality and sludge management. The reported 58% ammonia removal within 30 minutes of ACZ adsorption [136] is impressive, demonstrating the efficiency of the nano-adsorbent. However, the combined approach's ability to achieve 76% removal in just 20 minutes is particularly noteworthy, suggesting a synergistic effect when both processes are employed together. This high removal efficiency, coupled with significant DOC reduction, indicates a dual benefit of the treatment process, making it highly suitable for drinking water applications. Another study used natural zeolite (NZ01) as an adsorbent, where the adsorption capacity is influenced by pH, with NZ01 showing the highest NH_4^+ adsorption compared to other zeolites [137]. The addition of NZ01 into the system significantly enhances the NH_4^+ removal efficiency in synthetic NH_4^+ -kaolin suspension. The coagulation process, facilitated by alum, aids in the flocculation of destabilized solid particles, contributing to turbidity removal. The adsorptive coagulation process effectively removes ammonia by combining the adsorption capacity of NZ01 with the coagulation properties of alum, resulting in improved water quality through simultaneous removal of NH_4^+ and turbidity [137].

Biological treatment

Biological treatment methods such as nitrification and denitrification are also effective for ammonia removal from wastewater through the activities of various microorganisms [138,139]. The nitrification process involves ammonia-oxidizing bacteria and nitrobacteria converting ammonia nitrogen into nitrite nitrogen and nitrate nitrogen under aerobic conditions, typically at pH levels of 8.0 and 8.4 [138]. This process is highly stable, cost-effective, and operationally simple, resulting in significant aeration cost savings of about 50% and eliminating secondary pollution. Conversely, the denitrification process utilizes denitrifying bacteria, including heterotrophic and autotrophic microorganisms, to reduce nitrate and nitrite nitrogen into nitrogen gas in the absence of oxygen or under low-oxygen conditions, achieving a 90% reduction in operating costs and a 100% savings

on carbon sources, while maintaining high volumetric efficiency [138]. Despite these benefits, biological treatment methods are challenged by high land occupancy and large oxygen demand. The presence of harmful substances can inhibit microbial activity, and the long start-up time required for laboratory cultivation and practical application of these processes can be a hindrance. Anammox bacteria, although effective in converting ammonia to nitrogen gas have a low growth rate and are sensitive to substrate conditions like organic matter, nitrite nitrogen, phosphate, ammonia, and physical conditions such as oxygen and electromagnetism [139]. In lab-scale bioaugmentation experiments with microbial consortia, the cultures were mixed in a sterile conical flask to form a consortium thus demonstrating efficient ammonia removal and effective nitrite and nitrate conversion. Initial conditions included an ammonia concentration of $1.07 \pm 0.263 \mu\text{M/L}$, a temperature of $30 \pm 2^\circ\text{C}$, a pH of 7.8 ± 0.4 , and dissolved oxygen levels of $6.7 \pm 0.71 \text{ mg/L}$. The microbial consortium achieved high removal efficiencies and a survival rate of 97.2% [139], indicating the potential for creating healthier aquatic environments. However, the study's limited monitoring parameters, such as the absence of final microbial count, pH, and dissolved oxygen values, restrict comprehensive evaluation. Additionally, the long-term stability of the consortium over 15 days raises questions about its sustainability in real-world applications. While the practicality and cost-effectiveness of implementing microbial consortia in large-scale aquaculture facilities appear promising, further research is necessary to determine the feasibility and economic viability of this approach.

Comparative analysis of natural adsorbents

Efficiency in ammonia removal

Natural adsorbents have emerged as a promising solution for ammonia removal from wastewater, particularly in Malaysia, where industrial activities contribute significantly to nitrogen pollution. This analysis compares various natural adsorbents based on their cost-effectiveness, availability, environmental impact, and sustainability.

Availability in Malaysia and cost-effectiveness

The market for natural adsorbents in Malaysia is influenced significantly by the availability and cost-effectiveness of materials derived from the robust agricultural and industrial sectors of the country. Coconut husk which a by-product of the coconut industry is notably abundant in Malaysia, where it makes up to 6.7% of all agricultural waste generated in Malaysia [140]. The annual coconut production exceeds 80,000 tons thus its low procurement cost makes it a preferred option for wastewater treatment

and other applications as studies have shown that modified coconut husk can achieve ammonia removal efficiencies exceeding 90% [140]. This high availability and efficiency place coconut husk at the forefront of natural adsorbents in the market. Palm oil biomass is another highly available and cost-effective natural adsorbent due to Malaysia being the world's second-largest producer of palm oil contributing to over 75.61 million tons of solid biomass waste generated annually including empty fruit bunches, fronds and trunks, yielding a competitive advantage [140]. Certain studies highlight these materials achieving ammonia removal capacities of up to 3.58 mg/g [141].

Conversely, natural zeolites although effective in ammonia adsorption due to their unique porous structure, they are less favored in the Malaysian market. Access to natural zeolite in Malaysia is considered limited compared to more readily available agricultural by-products like coconut husk and palm oil biomass. The extraction and processing of zeolite often incur higher costs making them less appealing to smaller treatment facilities. According to a study, the costs associated with mining and processing natural zeolites can be up to 30% higher than those for processing agricultural waste materials leading to a preference for more economically accessible options [142,143]. Currently, the Malaysian market for natural zeolite is still developing. While industries are beginning to recognize the utility of zeolite in wastewater treatment, such as in palm oil milling, the market demand for zeolite has not yet reached the scale necessary to stimulate significant investment in its extraction or processing [144,145]. Moreover, emerging data indicates a growing interest in zeolite applications within certain sectors but the competition from other natural adsorbents remains strong. For instance, estimates show that the use of agricultural waste materials in wastewater treatment can achieve nitrogen removal efficiencies upward of 90%. At the same time, natural zeolites typically yield lower efficiencies due to the challenges [146,147].

Another emergent competitor is biochar which has gained popularity for its multifunctional roles in environmental applications including soil enhancement and pollutant adsorption. Biochar availability in Malaysia is supported by the country's rich agricultural sector which generates a wealth of biomass resources that are suitable for biochar production. Materials such as palm oil waste, including Palm Kernel Shell (PKS) and empty fruit bunches alongside residues from bamboo farming are readily accessible [148]. The increasing focus on sustainable waste management practices has spurred interest in converting these agricultural residues into

biochar, which can be utilized in various applications such as soil amendment, carbon sequestration and pollution mitigation [148]. Recent market analyses indicate that Malaysia's biochar market is poised for growth. In 2021, the market for biochar in the Asia-Pacific region, including Malaysia, was valued at approximately USD 100 million, with an expected compound annual growth rate (CAGR) of around 13% from 2022 to 2028 [149]. This growth is largely driven by increasing awareness of biochar's environmental benefits and applications in agriculture, construction and waste management. As industries and government bodies seek to implement more sustainable practices, the demand for biochar is expected to rise significantly. The recent launch of the Bukit Selar Carbon Station by Carbon Plus marks a significant milestone in Malaysia's industrial biochar landscape [150]. This pioneering facility, located in Bukit Selar, Kelantan, is set to produce 500 tonnes of high-quality biochar annually through advanced gasification technology provided by Renewables Plus [150]. The availability of biomass feedstocks combined with innovative facilities like the Bukit Selar Carbon Station positions Malaysia as a key player in the regional biochar market. Moving forward, the biochar industry in Malaysia is expected to grow substantively, bolstered by advances in technology, increased investment, and supportive government policies aimed at environmental sustainability.

Moreover, chitin and chitosan, derived mainly from crustacean shells, present an environmentally friendly alternative in the adsorption market. With the seafood industry in Malaysia yielding approximately 1.5 million metric tons of shellfish annually, sourcing these materials becomes feasible and economically sound for wastewater treatment applications [151]. These natural polysaccharides have shown considerable adsorption capacity for various pollutants leading to an optimistic market outlook. Regarding fish scales, the potential remains largely underutilized in Malaysia despite the sizeable fish processing industry. Fish scales are typically discarded as waste, yet they have demonstrated effective properties in adsorption applications [152]. Statistical integration of fish waste into broader waste management strategies could enhance their market acceptance. At the same time, anise residues and peanut shells represent niche markets within the natural adsorbent landscape. Both materials are generally low-cost and readily available as by-products of agricultural activities [153]. However, their adoption relies heavily on consumer awareness and research into their effectiveness compared to more

established adsorbents like coconut husk and palm oil biomass. Currently, agricultural by-products are favored for their low environmental impact and contributions to a circular economy. In summary, the market for natural adsorbents in Malaysia showcases the strong positions of coconut husk and palm oil biomass owing to their abundance and cost advantages.

Environmental impact and sustainability

The environmental impact and sustainability of natural adsorbents are critical considerations in their application for nitrogen removal in wastewater treatment. Utilizing agricultural waste materials, such as coconut husk and palm oil biomass, not only aids waste management but also fosters a circular economy by valorizing by-products that would otherwise contribute to environmental pollution. By repurposing these materials, we can mitigate waste accumulation and promote resource efficiency. Additionally, these natural adsorbents are biodegradable and possess a lower environmental footprint compared to synthetic alternatives, which often involve energy-intensive production processes and can introduce harmful chemicals into ecosystems.

Moreover, the use of natural adsorbents can diminish the reliance on chemical treatments, which frequently have adverse environmental effects, such as the potential for chemical runoff and toxicity to aquatic life. This shift towards more sustainable practices aligns with global efforts to reduce chemical usage in wastewater treatment and promote eco-friendly solutions. However, it is essential to consider the environmental implications of all adsorbents. For instance, while natural zeolites are effective in nitrogen removal due to their unique properties, their mining and processing can lead to habitat disruption and other environmental concerns if not managed sustainably. The extraction process can impact local ecosystems, leading to soil erosion, loss of biodiversity, and changes in land use.

Therefore, when selecting an adsorbent for nitrogen removal, it is crucial to evaluate not only its effectiveness in treating wastewater but also its overall environmental impact. This holistic approach ensures that the chosen solution contributes positively to environmental sustainability while effectively addressing nitrogen pollution. By prioritizing natural and sustainable materials, we can enhance wastewater treatment practices while minimizing ecological harm.

Table 1. Comparison of key aspects of natural adsorbents for ammonia removal in Malaysia

Adsorbent	Ammonia Removal Technique	Key Parameters	Removal Efficiency	Advantages	Disadvantages	Availability in Malaysia	Reference
Coconut Husk	Adsorption	Dosage: 5-10 g/L; pH: 6-8; Contact Time: 30-120 min; Temp: 25-30°C; Initial NH ₃ : 10-100 mg/L	Up to 90%	Low-cost, abundant agricultural waste, effective adsorption capacity.	Limited adsorption capacity requires modification for optimal performance.	Widely available as agricultural waste.	[77,78]
Palm Oil Biomass	Adsorption	Dosage: 10-20 g/L; pH: 7; Contact Time: 60-180 min; Temp: 25-35°C; Initial NH ₃ : 50-200 mg/L	Up to 88.6%	Utilizes local agricultural waste, is cost-effective, good removal efficiency.	Variability in biomass quality may require pre-treatment.	Readily available from palm oil industry.	[18,21, 22,23,39]
Zeolite	Adsorption	Dosage: 1-5 g/L; pH: 6-9; Contact Time: 60-120 min; Temp: 20-30°C; Initial NH ₃ : 20-150 mg/L	70-90%	High affinity for ammonium ions, reusable and effective in various conditions: high regeneration capability.	Cost can be higher than other natural adsorbents and may require pretreatment/modifications	Available in natural deposits; used in various applications.	[23, 39,41,42]
Biochar	Adsorption	Dosage: 5-15 g/L; pH: 6-8; Contact Time: 30-120 min; Temp: 25-30°C; Initial NH ₃ : 10-100 mg/L	60-85%	Enhances soil quality, sustainability and can be produced from various biomass.	The production process can be energy-intensive and variable quality.	Increasingly produced from agricultural residues.	[102,103]
Algae	Biological Treatment	Dosage: 10-15 g/L; pH: 7-8; Contact Time: 24-72 hours; Temp: 20-30°C; Initial NH ₃ : 20-100 mg/L	70-96%	Eco-friendly, promotes nutrient cycling and can be cultivated locally.	Growth conditions can be sensitive and may require specific nutrients.	Available in natural water bodies and can be cultivated.	[12,35,111]
Chitosan	Adsorption	Dosage: 1-3 g/L; pH: 5-7; Contact Time: 30-120 min; Temp: 25-30°C; Initial NH ₃ : 10-50 mg/L	80-95%	Biodegradable, effective for heavy metals and ammonia and high adsorption capacity.	Higher cost compared to other natural adsorbents and sourcing can be limited.	Sourced from the seafood industry and limited availability.	[56]
Animal Bones	Adsorption	Dosage: 5-10 g/L; pH: 6-8; Contact Time: 60-180 min; Temp: 25-30°C; Initial NH ₃ : 20-100 mg/L	70-94%	Utilizes waste materials and effective for various contaminants; low-cost.	Requires pre-treatment and potential for odor issues.	Readily available as a byproduct of the meat industry.	[54,97]
Rice Husk	Adsorption	Dosage: 5-15 g/L; pH: 6-8; Contact Time: 30-120 min; Temp: 25-30°C; Initial NH ₃ : 10-100 mg/L	60-80%	Abundant agricultural waste, low-cost and renewable.	Limited adsorption capacity and may require modification.	It is widely available in rice-producing regions.	[79]
Neem Leaves	Adsorption	Dosage: 5-10 g/L; pH: 6-8; Contact Time: 30-120 min; Temp: 25-30°C; Initial NH ₃ : 10-100 mg/L	65-85%	Biodegradable, possesses antimicrobial properties and low-cost.	Limited research on effectiveness and variable quality.	Available in tropical regions; commonly used in traditional medicine.	[79]

Conclusion

The review of ammonia removal from wastewater using natural adsorbents underscores the pressing need for sustainable and cost-effective solutions to combat the escalating challenge of ammonia pollution in Malaysia's industrial effluents. The findings reveal that locally sourced materials, such as coconut husk, palm oil biomass, biochar and algae possess significant potential as effective adsorbents for ammonia removal. These materials not only enhance pollutant removal efficiency but also contribute to environmental sustainability by valorizing agricultural waste products that would otherwise exacerbate pollution issues. However, there is a critical need to optimize the adsorption conditions and understand the mechanisms that govern the efficacy of these natural adsorbents. Future research should focus on innovative approaches to combine natural adsorbents with existing or emerging treatment technologies. For instance, integrating natural adsorbents with biological treatment methods could enhance the overall removal efficiencies and reduce the reliance on chemical processes which often have detrimental environmental impacts. Additionally, exploring hybrid adsorbents that combine the properties of natural materials with engineered ones could significantly improve adsorption capacities and regeneration potentials, offering a dual benefit of efficiency and sustainability. From a policy perspective, industries in Malaysia should be encouraged to incorporate natural adsorbents into their wastewater treatment systems as part of a comprehensive strategy to adhere to the Environmental Quality Act standards. This transition could be supported by government incentives such as subsidies or grants for research and development on natural adsorbent technology as well as funding for pilot studies that demonstrate their effectiveness in real-world applications. Moreover, regulations should facilitate the establishment of environmentally friendly waste management practices that promote the utilization of agricultural by-products in wastewater treatment. Collaborative efforts between industry stakeholders, academic researchers and government agencies are essential to fostering innovation and the implementation of these sustainable practices. By prioritizing the use of natural adsorbents, Malaysia can advance towards eco-friendly wastewater management that not only mitigates ammonia pollution but also protects aquatic ecosystems and public health. Emphasizing a circular economy approach will not only enhance the sustainability of industrial practices but also contribute to Malaysia's long-term environmental goals. In conclusion, leveraging local resources through the adoption of natural adsorbents presents a viable path for improving wastewater treatment in Malaysia. Further

studies especially focusing on the scalability and long-term performance of these solutions are crucial for establishing a robust framework that can effectively address the multifaceted issues associated with ammonia pollution in industrial wastewater.

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