



Engineering principles and process designs for phosphorus recovery as struvite: A comprehensive review

Nageshwari Krishnamoorthy^{a,1}, Baishali Dey^{a,1}, Yuwalee Unpaprom^b, Rameshprabu Ramaraj^b, Gaanty Pragas Maniam^c, Natanamurugaraj Govindan^c, Sivaraman Jayaraman^a, Thirugnanam Arunachalam^a, Balasubramanian Paramasivan^{a,*}

^a Department of Biotechnology and Medical Engineering, National Institute of Technology Rourkela, Odisha 769008, India

^b Maejo University, Chiang Mai 50290, Thailand

^c Faculty of Industrial Sciences and Technology, Universiti Malaysia Pahang, 26300, Malaysia

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ABSTRACT

The rising population relies heavily on expensive phosphate (P) fertilizers for its agricultural productivity that qualms the national food security. Globally, there is a growing concern for the profound reliance on finite phosphorite reservoirs for commercial phosphate fertilizers' qualitative and quantitative production. On the contrary, uncontrolled discharging of nutrient-rich wastewaters into natural streams, affecting the aquatic ecosystem. These critical situations have caused a scientific threat that has forced the researchers to explore other opportunities for the conservation of nutrient resources, recovery, and recycling. This review examines the nutrient recovery paradigms for struvite production by curtailing the nutrient gap between wastewater treatment and agricultural productivity. It comprehends the fundamental chemistry, thermodynamics, and factors influencing the struvite production process with its detailed mechanisms. Further, it deliberates the possible struvite engineering strategies, including process designs to enhance P recovery at the lab, pilot, and commercial scale. Also, it emphasizes the applications of nutrient-loaded struvite as a slow-release fertilizer and the challenges associated with economic feasibility and scaling up of the process in recent decades.

1. Introduction

Phosphorous is the eleventh most copious element on earth's crust (0.1%), occurring in the form of calcium phosphate rocks [98]. It is a finite, non-renewable resource and a critical essential nutrient (NPK) for primary producers. It plays an indispensable role in ensuring global food security and is used in commercial fertilizer production. It has a highly immoderate use having reserves distributed erratically across the globe. However, much of this phosphorus often end up in water bodies as agricultural runoff leading to eutrophication and algal bloom, plummeting their suitability for recreational use and drinking water, thus necessitating expensive treatment processes to make it fit for human consumption. With the increasing population, increasing amounts of phosphorus are ending in water bodies disrupting the natural phosphorus cycle; on the other; there have been rising concerns of

phosphorus supply worth only 100 years' left [82].

The paradigm shifts from disposing to recovery and reuse of phosphorus from these nutrient-rich wastewaters can solve the problems mentioned earlier. Phosphorus (P) and nitrogen (N) can be of industrial origin, mainly from semiconductor wastewater, beverage wastewater, swine wastewater, leachates from municipal landfill, anaerobically digested sewage sludge, human urine and runoff from waste disposal sites, which are all point sources. Agricultural and pasture runoff, human activities like logging and expansion of land and waterways, deposition from the atmosphere are a few examples of non-point sources [74]. High concentrations of salts, $\text{NH}_4^+\text{-N}$, PO_4^{3-} and organic matter are present in leachates percolating from solid waste, making it one of the most impactful wastewater types, causing significant environmental effects [52]. The reduction of eutrophication in freshwater and coastal ecosystems can be brought about by eco-recycling nutrients from

* Corresponding author.

E-mail address: biobala@nitrrkl.ac.in (B. Paramasivan).

¹ Authors contributed equally.

² <http://orcid.org/0000-0002-3821-5029>.

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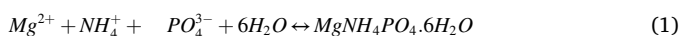
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source-separated urine and maximizing its recovery [74]. Formerly regarded as one of the critical polluting sources in livestock farms, swine wastewater contains low magnesium but is rich in total orthophosphate, total ammonia nitrogen (TAN), and organic substances, becoming a significant reason for eutrophication in water bodies [53]. Therefore, for effective pollution control, sustainable nutrient recovery technologies are required. Recently, researchers recommend NH_4^+ -N recovery in the form of struvite to curb these problems [74].

Many other wastewater sources for phosphorus recovery have been proposed for struvite recovery on a lab-scale level but are yet to be upscaled (Table 1). Previous review works have focussed only on specific findings, influencing parameters of precipitation of struvite, recovery of struvite from a specific wastewater source or on the various available technologies from a techno-economic point of view. This review paper starts from the very basics of struvite chemistry and structure to critical analysis of various wastewater sources and technologies developed so far for struvite recovery. It also focuses on the pros and cons of each technology while also highlighting the techniques that are yet to be upscaled for commercialization. It throws light on the influence of multiple factors that regulate the perspective of struvite formation along with respective reactor design and costs incurred. Lastly, the broad spectrum of struvite application, challenges and future aspects will be explored, making this review exceptional. The aim of this review is to analyse the developments that have taken place in this sector over the years, while emphasizing the research gaps.

2. Basic physicochemistry of struvite

Struvite stands for magnesium ammonium phosphate ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$, MAP). In a supersaturated solution, a chemical reaction among free Mg^{2+} , NH_4^+ , and PO_4^{3-} results in the formation of struvite as demonstrated in Eq. (1) [138].



According to Eq. (1), equimolar quantities of Mg^{2+} , NH_4^+ , and PO_4^{3-} are required to form $\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$. Over the years, several articles have addressed the nutrient burden in wastewater and suggested the recovery of N or P as struvite as a feasible solution. Since the first identification of struvite in the year 1939, structural accumulation of struvite in wastewater pipes was reported as a common problem [151]. Since then, studies on spontaneous deposition of struvite have been investigated. It creates a nuisance in pipe-work and associated machinery (particularly at joints and in pumps and aerators), leading to costly maintenance and repair of wastewater treatment plants. However, other chemical species might also form due to the common ion effect. The reaction is highly pH-dependent. The precipitation reaction occurs in alkaline conditions, with pH varying from 7.0 to 11.5 [69]. In further research, by fitting the first-order kinetic model, an estimated rate constant of 4.2 h^{-1} was obtained, and this association well-suited the loss of ortho-phosphate from bulk solutions [62]. Various process

parameters such as pH, temperature, degree of supersaturation, foreign ions, mixing energy, Mg/P molar ratios, seeding, hydraulic retention time (HRT), Ca/Mg molar ratio, and feeding sequence are likely to influence the struvite precipitation process. The physicochemical properties of struvite are shown in Table 2.

The occurrence and development of struvite crystals are preceded by nucleation (crystal birth) and crystal growth [3], as shown in Fig. 1. New crystallization centers must exist in the solution before crystals can grow. The crystals form at high supersaturation (primary nuclei) conditions, followed by lower supersaturation (secondary nuclei). A combination of several physicochemical parameters like pH, temperature, mixing energy, supersaturation, presence of foreign ions, as well as factors like reaction kinetics, crystal state of initial compounds, matter transfer between solid and liquid phases, and thermodynamics of liquid-solid equilibrium influence the struvite forming process, making it very difficult to predict or control [13].

Magnesium, ammonium, and phosphate are the significant ions forming struvite, which defines its ion activity product $\text{IAP} = \{\text{Mg}\} \{\text{NH}_4^+\} \{\text{PO}_4^{3-}\}$. Struvite nucleation and growth occur when the system is supersaturated, and the IAP is higher than the solubility product (K_{sp}), returning the system to equilibrium [114]. Depending on precipitation conditions, struvite crystals may have different morphologies, but supersaturation majorly defines crystal size and morphology

Table 2

Summary of the general physicochemical properties of struvite crystals.

Characteristics	Nature of struvite	Ref.
Chemical name	Magnesium ammonium phosphate hexahydrate	[114]
Formula	$\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$	
Molecular weight	245.43 g mol ⁻¹	
Size	15 μm –3 mm	[4, 160]
Composition	Magnesium 10.73% Phosphorus 12.54% Hydrogen 1.64% (NH_4) ₂ O Hydrogen 4.93% (H_2O) Nitrogen 5.71% Oxygen 65.20%	
Name origin	Named after Heinrich Christoph Gottfried Struve (1772–1851)	
Color	White, Yellowish white, Brownish white.	
Luster	Vitreous	
Specific Gravity/ Density	1.705 g/cm ³	[86]
Hardness	1½–2 (Mohs scale)	
Aspect	White glowing crystal	
Structure	Orthorhombic (space group $Pmn21$) Presence of regular PO_4^{3-} octahedral, distorted Mg (H_2O) ₆ ²⁺ octahedral, and NH_4^+ groups	[1]
Type of bonds	Hydrogen bond	
Tenacity	Brittle	
Solubility constant	$10^{-13.26}$	[100]

Table 1

Sources used for phosphorus recovery reported in the existing literatures.

Sources of wastewater	pH	Mg: PO_4^{3-}	$\text{PO}_4\text{-P}$ (mg L ⁻¹)	$\text{NH}_4\text{-N}$ (mg L ⁻¹)	P recovery (%)	NH_4^+ recovery (%)	Ref.
Leather tanning	8.1–8.4	1:1	12.3	2405	90	85	[165]
Raw Swine	7.9	1:1	845	31	92	81	[121]
Semiconductor	2.9	1:1.2 (NH_4 :Mg)	286	100	70	98	[68]
Dairy industry	6.035 \pm 0.06	–	70	45	93	89	[72]
Coking/coke oven wastewater	9.3	1.6 (Mg: NH_4)	56	3500	–	95	[75]
Dairy (anaerobically digested)	8.3 \pm 0.3	1:1	1060	450	–	> 90	[130]
Fertilizer industry	7.86 \pm 0.1	1:1	–	1197	–	97	[153]
Baker's yeast industry effluent	7.7	1.5:1	529	10.8	97	92	[143]
Cola beverage	–	1:1	415	–	97	–	[39]
Poultry (anaerobically digested)	8.58	1.5:1	–	9356	–	97	[152]
Cattle urine	6.5	1:0.5 (urine:brine)	305	7732	–	–	[112]
Human urine	8	1.7:1	450	300	70	–	[139]

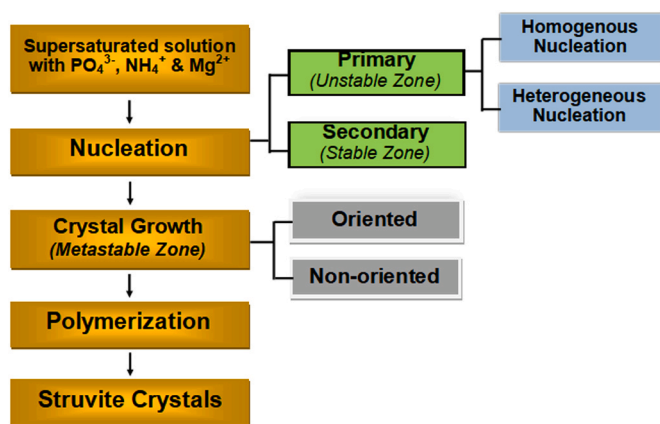


Fig. 1. Schematic representation of mechanisms involved in struvite crystal growth.

[127]. He et al. [50] established the thermodynamic equilibrium diagrams for $Mg^{2+} - PO_4^{3-} - NH_4^+ - H_2O$ system at 298 K. The theoretical analyses showed that the optimum pH for phosphorus recovery decreased from 9.8 to 8.8 when Mg concentration increased from 0.01 to 1.0 mol/L. Increasing the total ammonia concentration increased phosphorus removal but affected Mg concentration significantly. The estimated pH range for struvite formation was between 5 and 10 at high ammonia concentrations and between 7 and 10 at low ammonia concentrations [138].

According to nucleation theory, the rate of nucleation (J) or the number of struvite nuclei formed per unit time (second) and unit volume (cm^3) follows the following general equation (Eq. (2)):

$$J = A \exp \left[\frac{-16\pi\gamma^2}{3k^3} \frac{v^2}{T^3 (\ln\Omega)^2} \right] \quad (2)$$

where A is a kinetic factor, k is the Boltzmann constant (1.38 J K^{-1}), γ is the interfacial tension between a crystal and the solution (mJ m^{-2}), v the molecular volume (cm^3), T is the absolute temperature (K), and Ω is the supersaturation ratio [13]. Thus, the nucleation rate directly depends directly on supersaturation Ω at any solution pH due to rapid mass transfer from liquid to a solid phase [13]. The dynamics of the interactions and ion clustering are affected by the presence and concentrations of other co-existing ions, causing a change in the ion speciation profiles of H_3PO_4 and ammonia (NH_3) with respect to pH, as seen in Fig. 2 [138]. Similarly, Ravikumar et al. [116] reported that depending on their availability; other ions can substitute NH_4^+ (e.g., K^+ , Rb^+ , Cs^+) or Mg^{2+} (e.g., Ca^{2+} , Zn^{2+} , Cd^{2+}), resulting in the formation of crystals

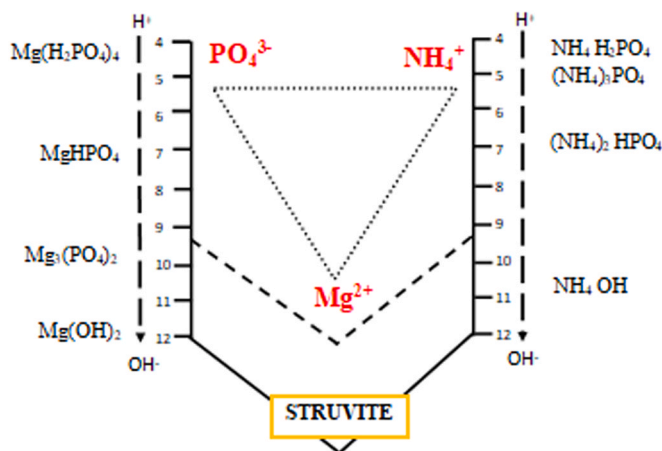


Fig. 2. Variation in salts and crystals formed with respect to pH.

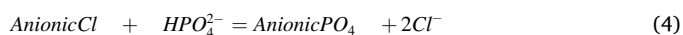
similar to struvite in appearance but different in composition. Different crystal characteristics were also found in the crystal-hydrates $MgNH_4PO_4 \cdot nH_2O$, where $n = 1, 3, 6$, and 7 [138].

3. Methods of struvite production

Research on struvite recovery started majorly as a method to treat wastewater. It recently found its use as a potential replacement of the fast depleting phosphorus source for commercial fertilizer production. Based on the underlying mechanism of struvite crystallization, alternative techniques have been developed over the years (as shown in Fig. 3). Chemical precipitation has been the most successful in terms of cost and energy demands. These techniques have been discussed in detail below.

3.1. Ion-exchange

This method is based on the principle of selective exchange of ions from wastewater (NH_4^+ , PO_4^{3-}) for struvite precipitation. They can be either sulfonic/carboxylic-based anionic exchanger or zeolite-based cationic exchanger, as seen in Fig. 4a. Sodium chloride is used as a regenerating solution in the columns where ammonium and P ions are replaced with sodium and chloride ions, respectively, according to Eqs. (3) and (4). The addition of $MgCl_2$ to the regenerates in a stoichiometric ratio of $Mg^{2+}:NH_4^+:PO_4^{3-} = 1:1:1$ results in struvite formation.



Generally, wastewaters do not contain balanced proportions of N and P [130]. Thus, efficient precipitation does not take place spontaneously until stoichiometric amounts of chemicals are added. The major factors influencing this process are the functional groups present in the ion exchange resin, the concentration of the wastewater source being used, and the ion exchange matrix [66,67]. The use of ion-exchange isothermal supersaturation has further modified the conventional ion-exchange technique. The principle was to simplify the precipitating solution concentration at a given temperature beyond its solubility level to enable spontaneous crystallization of struvite [102]. This approach is relatively faster but requires the regeneration of resins from time to time. The availability of specific anion exchangers for P sorption is limited with the added disadvantage of impurity co-precipitation.

3.2. Chemical precipitation

In this method, the external addition of Mg is required to precipitate struvite from nutrient-rich wastewater sources in a mechanically agitated manner (Fig. 4b). Most commonly used Mg salts are magnesium sulfate, magnesium oxide, and magnesium chloride. Still, some non-conventional renewable sources like seawater, bittern, wood ash, and struvite pyrolysate are also under study, which make this overall approach less cost-intensive and viable [122,53,86]. pH is the most important influencing factor that may be adjusted using NaOH (most commonly used), MgO, KOH, NH_3 , or CO_2 stripping to maintain the alkaline conditions required for struvite crystallization [163]. In lab-scale studies, agitation in a stirred batch reactor is provided, which is easy to install and operate [39]. In contrast, at a larger scale, fluidized bed design is preferred as they provide greater surface area, better turbulence, and reduced crystallization induction time [19]. The main advantage of this approach is its simplicity of operation. However, mechanical agitation produces unrecoverable fines, which can be recycled for seeding material [66,67]. Though this method is widely preferred, there are certain limitations, including the external addition of chemicals, that make the process cost-intensive, and labor-dependent. Among these chemicals, few are insoluble and energy-intensive and due to mechanical agitation, loss of ammonia might occur [66,67]. This method is relatively easy to install and operate. It does not require the

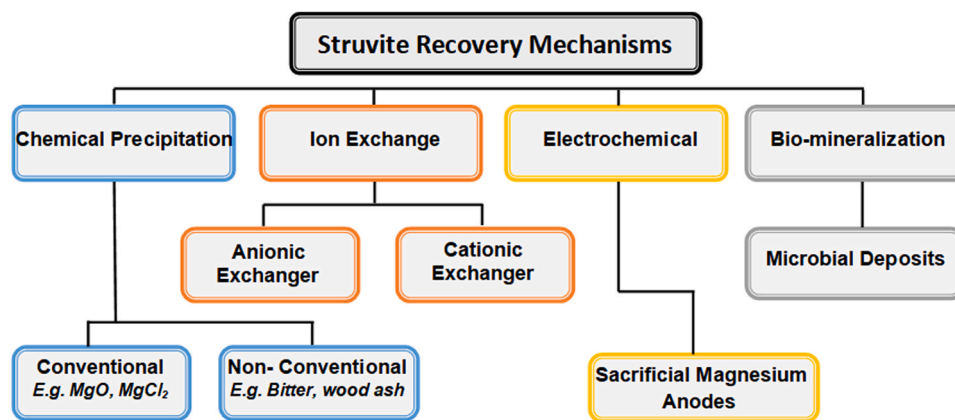


Fig. 3. Struvite recovery mechanisms.

use of a sophisticated instrument; however, it results in co-precipitation of impurities due to ineffective crystallization in the absence of optimum conditions.

3.3. Electrochemical

In the case of decentralized reactors, high mechanization of the dosage and small reactor sizes make the struvite precipitation process quite challenging [55]. A novel approach has been developed where Mg is sacrificially corroded from the anode to act as the source for precipitation of struvite. According to the literature, this technology is an excellent substitute for conventional wastewater treatment. The major problem lies in transportation of source-separated resources from one place to another. Thus decentralized or small onsite reactors would better suit large centralized treatment plants [47]. Electrolytic treatments are more advantageous due to simple equipment design, brief retention times, and smooth operation, thus reducing the operating cost in large-scale applications [140].



During electrolysis, there is increase in the diffusion rate of oxygen towards the cathode. As a result of this, the interfacial pH near the cathode increases (up to 7.5), leading to the generation of OH^- ions, as illustrated by Eqs. (5) and (6). These OH^- formed are immediately consumed by the chemical reactions to form precipitates [145]. According to the experiments conducted by Kruk et al. [73], electrolytic Mg release was found to be an efficient method for obtaining highly pure crystals and greater phosphorus removal efficiencies. The treatment of fermented waste activated sludge supernatant achieved a phosphate removal efficiency of 98% at an electric current density of 45 A m^2 within two hours. Wang et al. [146] and Hug and Udert [55] concluded that sacrificial Mg electrodes could easily compete with soluble Mg salts such as MgCl_2 and MgSO_4 in terms of cost but are more expensive than MgO . Energy costs for the electrochemical process were insignificant. pH, current density, and reaction time are the major influencing factors in the electrochemical precipitation of struvite [66,67]. These experiments need to be scaled-up and investigated to determine the key influencing parameters affecting the formation of struvite through electrochemical precipitation and to develop new reactor designs with cheaper and stable anodes and cathodes. pH optimization is not required, and hydrogen released at the cathode can be harvested as a potential biofuel. The efficiency gets limited by scale formation on the cathode, costly cathode material (e.g., platinum), and co-precipitation of impurities. Fig. 4c represents the schematic diagram of a typical electrochemical process.

3.4. Biomineralisation

Biomineralisation is the natural process by which microorganisms (e.g., *Myxococcus xanthus*, *Staphylococcus aureus*) cultured in a medium containing PO_4^{3-} , Mg^{2+} and NH_4^+ , harden their structural tissue with mineral deposits such as struvite [132]. The underlying mechanism is the metabolism of nitrogenous compounds in the medium, causing a rise in pH and thus favoring precipitation of struvite [67]. Fig. 4d represents typical biomineralisation occurring in a reactor. The precipitation of struvite is said to start during the exponential growth phase and reach the highest in the stationary phase. Jimenez-Lopez et al. [61] reported a link between species of microbes used, the morphology of struvite, and physicochemical environments of the culturing media. Similar to chemical precipitation, Ca^{2+} ions in wastewater play an inhibiting role in biomineralization [131]. Struvite crystallization occurs by heterogeneous nucleation due free surfaces on dead and disrupted cells or isolated bacterial structures (e.g., cell membranes) [66,67]. The presence of negatively charged molecules like proteolipids, phospholipids, glycoprotein, proteoglycan in the organic matrix of disrupted bacterial cells gets released into the medium. It attracts positive ions like Mg^{2+} , resulting in struvite precipitation. The culturing media contains several metabolites released by the microbes (for example, osteopontin, citric acid, and succinic acid), including the components of the media itself, which have been shown to influence the process of crystallization and the morphology of struvite [84]. The advantage of this approach lies in the fact that external pH adjustments are not required. However, the process is relatively slow, and many a times, results in the co-precipitation of impurities.

3.5. Microbial electrolysis cell

Microbial electrolysis cell (MEC) is an upcoming chemical-free technology that can be used to precipitate struvite from organic-rich wastewater with simultaneous Hydrogen production. Electrochemically active microbes present in the cathode chamber, biologically oxidize the organic substances present in wastewater and release carbon dioxide, electrons, and protons into the chamber [9]. The protons travel from the cathode chamber through a protein exchange membrane placed between the two electrodes into the anode chamber. The electrons travel through an external circuit to the anode and combine with the free protons to release hydrogen gas, increasing the local pH [30]. This rise in pH helps in phosphorus recovery without any external addition of chemicals for gaining the alkaline pH range required for precipitation of struvite. The process is mainly affected by COD, pH, and aeration flow rate [8]. For fulfilling charge neutrality, ammonia moved from the anode department to the cathode, leaving a concentrated ammonia solution to be recovered later [15]. MECs can be both single-chambered or double-chambered (anode and cathode divided by

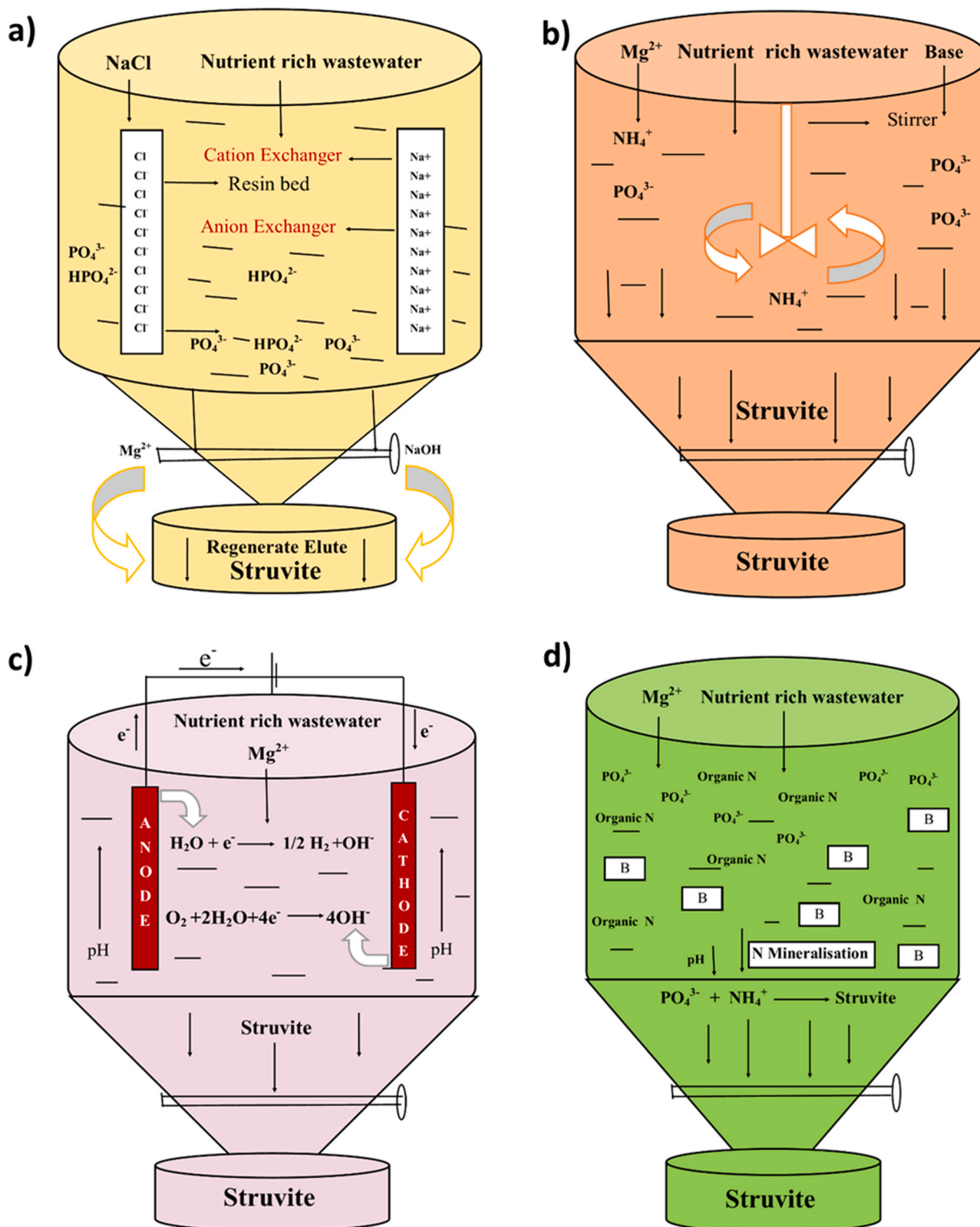


Fig. 4. Schematic representation of a typical a) ion exchange reactor b) precipitation reactor c) electrochemical reactor d) biomineralization reactor.

a separator like an anion or cation exchange membrane) depending on the requirement and optimizing the process to maximum efficiency. This technology can be used in a broad application area ranging from removal of pollutants (both organic and inorganic) to the production of value-added products like hydrogen, hydrogen peroxide, methane,

formic acid, and lastly, MECs can also be used as biosensors [54]. The challenges faced while using this technology are membrane fouling [156], scaling the electrodes due to deposition of phosphate salt crystals, and significant difference in the pH values between cathode and anode [9]. Life cycle assessment and Techno-economic analysis need to be

conducted to discover the true potential of this technology. MEC is still in its early stages, and the challenges regarding reactor designing, removal efficiencies, and power output still need to be overcome to obtain a sustainable technology for successful commercial application in the future.

4. Parameters affecting struvite production

Over the years, several reports regarding the efficacy of the struvite crystallization in recovering phosphorus from waste resources have been published. However, data concerning the kinetics of struvite precipitation is still inadequate. According to existing research, various physicochemical characteristics and process factors like pH, Mg: P ratio, temperature, supersaturation ratio, degree of mixing, the effect of HRT, presence of foreign ions such as Ca^{2+} , and seeding conditions are expected to affect the struvite formation process. Their extent of influence on crystallization and optimum conditions have been discussed below.

4.1. pH

The pH at which struvite forms is an essential factor influencing crystallization and is linked to supersaturation and solubility [77]. According to the studies conducted by Stratful et al. [135], there was no detectable quantity of struvite present at pH 7, while at pH 7.5, minuscules of tiny struvite crystals were produced. As the pH increased to 8.5, approximately 92% of Mg was removed from the solution. However, only 85% of phosphorus was incorporated into the crystals. The studies also concluded that, with an increase in pH beyond 9, the Mg removed was > 97%, while the residual P concentration was 12% of the initial. This implies that to extract all the available P, excess addition of Mg may be necessary. Liu et al. [85] reported a minor decrease in P removal efficiency when the pH increased from 9.7 to 10.0; however, Le Corre et al. [76] said that an increase in pH from 10.0 to 10.5 could favor the formation of $\text{Mg}_3(\text{PO}_4)_2 \cdot 22\text{H}_2\text{O}$ over struvite from wastewater.

As struvite precipitation begins, hydrogen ions get released into the solution. Thus in an unbuffered system, the pH might decline below the required level, inhibiting the struvite precipitation reactions. Consequently, studies were undertaken to maintain optimum pH by adding NaOH, which helped to retain the crystallization process until the reactants became limiting [135]. Liu et al. [87] have reported that hydrolyzed urine has a strong buffering capacity and can serve as a reliable source for structural formation without any pH control.

pH variation can lead to the formation of phosphate precipitates other than struvite. For example, when pH is in the range 9.0–10.5, hydroxyapatite (HAP) begins to form in the metastable zone, and magnesium phosphate complexes other than struvite tend to get precipitated at pH above 10 [82]. According to reports by Hao et al. [49], neutral pH can favor the development of pure struvite crystals; but, the rate of precipitation is affected. Generally, a pH range of 8.0–10.5 is considered optimal and hence recommended to obtain high-quality struvite [158]. Whereas, according to Andrade and Schuiling (2001), at higher pH, the NH_4^+ concentrations might volatilize from solution in the form of gaseous NH_3 . This is the reason why struvite is barely formed at higher pH (> 10.5). However, the optimum pH depends on the type of wastewater and its composition, and their specific pH recommendations are still limited in the literature.

Furthermore, pH affects the growth rate of struvite crystals and their sizes. The tiniest crystal size was observed at pH 10.5, whereas the most significant crystals (> 100 μm) were obtained at pH 9.5 [82]. Most wastewaters have a slightly acidic environment, due to which large amounts of alkali need to be added for pH improvement, which sometimes leads to 90% of the total operational cost [52]. Chemicals commonly used for pH adjustment are NaOH, MgO, $\text{Mg}(\text{OH})_2$, KOH, and K_2CO_3 . Dosing with $\text{Mg}(\text{OH})_2$ can serve the purpose of both pH control and act as an Mg source. Due to its weak alkaline nature and low solubility, large dosing is required [82]. An alternative to chemical dosing is

aeration; it can reduce the caustic addition by at least 50%, depending on the operating conditions.

Talboys et al. [137] with their studies confirmed that with decrease in pH, the solubility of struvite crystals increased without affecting final P concentration. The maximum allowable pH represents the maximum degree of saturation that the precipitation reactor can handle, beyond which the rate of primary nucleation dominates. When the operational pH exceeds, P removal is still potential, but it cannot be conducive for proper reactor operation [7].

4.2. Temperature

Zeng et al. [157] has stated that the optimum temperature for the process of struvite crystallization lies between 15 °C and 35 °C. Lee et al. [78] reported an increase in phosphorus removal efficiency from 63% to 78% with an increase in temperature from 5 °C to 50 °C. A similar study on an anaerobic digestion system showed that the solubility of struvite increased with an increase in temperature from 0 °C to 20 °C. Le Corre et al. [77] showed that higher temperatures could promote struvite crystals' growth and alter their structure. However, Capdevielle et al. [166] have reported that high temperatures accelerated the crystallization of amorphous calcium phosphate, leading to decreased dissolution rate and product purity. Adnan et al. [5] made a completely contrasting observation, stating that low temperatures (< 15 °C) favored struvite crystallization. Song et al. [133] documented that diffusion-controlled growth occurred during high crystallization temperatures (< 35 °C), whereas surface integration-controlled growths occurred at low temperatures. Such variances in observations may be due to different reaction mechanisms occurring in different kinds of wastewaters. Temperature differences during day and night, winter and summer, or diverse geographical regions may affect the struvite quality and process cost, and hence future investigation on this aspect is essential [82].

4.3. Degree of supersaturation

At a fixed pH, the solution supersaturation influences the crystallization rate through crystal induction time [77]. The ratio of conditional solubility product (P_s) to the equilibrium conditional solubility product (P_{seq}) results in the supersaturation ratio (SSR), as shown in Eq. (7). P_s and P_{seq} represent the solubility product in actual condition and at equilibrium state, respectively [115]. Therefore,

$$SSR = P_s / P_{seq} \quad (7)$$

The degree of supersaturation for struvite can also be represented by the Saturation Index value (SI_{MAP}). Eq. (8) establishes the stability order of precipitation or dissolution of solids. If SI_{MAP} is negative, the system is under-saturated for struvite precipitation. If SI_{MAP} is positive, the solution is supersaturated [105].

$$SI_{MAP} = \log \left[\frac{[\text{Mg}^{2+}] \cdot [\text{NH}_4^+] \cdot [\text{PO}_4^{3-}]}{K_{SMAP}} \right] \quad (8)$$

where $[\text{Mg}^{2+}]$, $[\text{NH}_4^+]$, $[\text{PO}_4^{3-}]$ are magnesium, ammonium and phosphate concentrations (mol/l); γ_{Mg} , γ_{NH_4} and γ_{PO_4} are the activity coefficients of $[\text{Mg}^{2+}]$, $[\text{NH}_4^+]$ and $[\text{PO}_4^{3-}]$ species, respectively; K_{SMAP} is the struvite solubility product. SI_{MAP} affects the solution pH directly as it dictates the solubility of struvite [104]. At constant supersaturation level, the growth rate increased by seven times, from 8 h to 40 h of precipitation. But inside a reactor, operational parameters such as temperature, Mg dosing, and recycle ratio also affect supersaturation. There is variation in saturation level for a particular pH due to wide variation in the composition of the centrate being fed [19].

4.4. Presence of foreign ions

Different wastewaters vary in ionic species and concentrations (e.g., Ca^{2+} , Zn^{2+} , Cu^{2+} , Al^{3+} , CO_3^{2-} and SO_4^{2-} etc.), affecting the process negatively by hindering the efficiency of nutrient recovery, product purity, morphology, and reaction speed. In this way, the purity is affected by metal ions that compete for P and co-precipitate as complexes along with struvite [33]. The adsorption of such ions in the active sites on the crystal surface further affected the crystal growth by blocking [63]. In the presence of Ca^{2+} , Fe^{2+} , and NO_3^- , the crystal size decreased by up to 46% [57]. The presence of such non-participating cations increased the crystal induction time by accumulating around the anionic species of struvite. However, this phenomenon is insignificant in the presence of carbonate (CO_3^{2-}), sodium (Na^+), and sulfates (SO_4^{2-}) [63]. The role of calcium (Ca) during struvite crystallization has been extensively studied [52,66]. Ca is known to prolong the induction time, and precipitate simultaneously as phosphate salts (calcium phosphate, hydroxyapatite) along-side struvite and affect struvite particle size, purity, and fertility [52]. The appearance of irregular crystals occurs when Ca concentration is high [133]. The size of the struvite particles decreased from 34.2 to 18.4 μm when there was an increase in Ca concentration [35].

4.5. Mixing energy or turbulence

Mixing or turbulence can also influence the struvite precipitation process. At high mixing rates, CO_2 liberation causes an elevation in solution pH, favoring struvite crystallization [77]. Mixing rates can affect crystal size and shape; crystals were smaller at low turbulence due to lower dissipation leading to higher saturation [120]. Indeed, high mixing can quicken the nucleation rate and, on the other hand, crystal breakage [86]. Li et al. [81] illustrated that stirring promotes the mass transfer of solute, enhancing the nucleation process and, thus, the crystal growth rate. P removal efficiency increased from 72.7% to 97.3% under non-stirring and stirring (160 rpm) conditions, respectively, but at rates > 240 rpm, the P removal did not vary significantly. The efficiency was more than 97.3% within 5 min of initial mixing, whereas at around 30 min, the efficiency reached as high as 99.0%. These studies indicate that mixing time is more effective than mixing speed for P removal [86].

4.6. Molar ratios of Mg/P

Mg is an essential constituent of struvite crystallization. It is added as an external source as most of the nutrient-rich wastewaters lack the stoichiometric amount of Mg required to precipitate struvite. Theoretically, struvite formation takes place in the $\text{Mg}^{2+}:\text{NH}_4^+:\text{PO}_4^{3-}$ molar ratios of 1:1:1. Ali et al. [7] illustrated that increase in the Mg:P ratio increased the degree of saturation at a given pH and, in turn, promoted phosphorus removal. In a study conducted by Rahaman et al. [115], Mg: P ratio in the range of 1.00–1.60 was investigated and found that better removal rates were achieved at a higher the Mg: P ratio. Thus, higher molar ratios had a strong influence on decreasing P concentrations.

4.7. Seeding

According to experimental results reported by Liu et al. [87], seed crystals could significantly decrease the induction and equilibration time significantly. It was found that the correlation between crystallization time and reactant concentration was a second-order power function and exponential function, without and with seed addition, respectively. The equilibration time shortened from 90 to 15 min when the seed concentration was 0.422 g L^{-1} . During the crystallization process, the seed materials act as a template for a further build-up of ions. These seeds control the nucleation process by providing sufficient surface area for the ions to bind, thereby reducing the crystal induction time. Some of the desirable features of a seed material include inertness,

large surface area, and isomorphism with precipitating crystals [6]. Among the reported seed materials, struvite fines from spontaneous precipitation are the most extensively studied [115,153,87]. Some other seeding materials used in struvite recovery are sand, pumice stone, stainless steel mesh, and borosilicate glass [76]. The use of non-isomorphic non-struvite seeds alters the type of nucleation from homogeneous to heterogeneous. Also, the grain size, surface roughness, dose, and supersaturation of crystallizing solution play a direct role in affecting seed's effectiveness [95]. Freshly precipitating struvite nuclei cannot precipitate upon non-isomorphic seed to grow into larger crystals; thus, the unstable clusters break down or redissolve, thereby increasing the induction time [6]. However, most of the reported studies on seeding used are based on synthetic waste and require further investigation for real-time application.

4.8. Effect of organic substances

Wastewaters generally contain a significant amount of organic substances. These materials cause a reduction in the rate of struvite formation; however, the size of the crystals seemed to increase [26]. Increasing the Mg: P ratio can regulate humic substances, transforming the struvite crystals from prismatic to pyramid structure [164]. Similarly, it was also found that both citrate and phosphocitrate were effective growth inhibitors, as they got adsorbed to the surface of struvite crystal and successfully blocked the active sites for growth, thereby increasing the induction time of crystallization. This can harm human health and the environment when used as fertilizer. Meanwhile, a glycoprotein called Tamm-Horsfall present in feline urine was seen to promote struvite crystallization. Humic and succinic acid can both inhibit struvite crystal formation and growth, while acetic acid had a slight influence on the crystal forming process. On the contrary, high glucose concentrations in the solution promoted P removal efficiencies and structural formation [159]. In general, organic substances do have a slight influence on struvite composition while not affecting purity.

4.9. Effect of Ca/Mg molar ratio

With the increase of Ca concentration (Ca/Mg molar ratio of 0–1.8) in the influent while keeping pH, Mg, and other ion concentration constant, the recovery and precipitation efficiencies hugely increased from 58% to 92% and 60% and 93% respectively. This was due to P precipitation as struvite and Calcium phosphate [105]. At a Ca:Mg molar ratio of 1:1, the amorphous content formed was very high for all NH_4 concentrations, and it decreased with an increase in Mg concentration. This is an indication of the role of Ca-containing materials in crystallization [94]. Several phosphates of Ca can be found to form in aqueous solutions, namely HAP, amorphous calcium phosphate (ACP), dicalcium phosphate dehydrate (DCPD), and octacalcium phosphate (OCP) [105].

As the ammoniacal nitrogen concentration increased in the absence of Ca, the percentage of P removal ranged between 45% and 51%. Similarly, Mg removal from solution ranged from 86.9% to 91.7% [94]. Bouropoulos and Koutsoukos [23] established that Mg ions influence the struvite particle micro electrophoretic mobility at an isoelectric point of 1.75, which advocated that it's higher supersaturation was beneficial for crystallization. Acelas et al. [3] has reported that $\text{Mg:Ca} < 0.2$ delayed HAP formation, and $\text{Mg:Ca} > 2$ counterbalanced the undesirable effect of Ca during struvite crystallization.

4.10. Effect of feeding sequence

Kim et al. [70] first investigated the effect of the feeding sequence of reagents added during struvite crystallization by testing different combinations of pH, Mg^{2+} and PO_4^{3-} . He observed that $\text{NH}_4\text{-N}$ removal efficiencies reduced to less than 50% when buffering reagents for pH control were supplemented prior to Mg and orthophosphate. Also, after

the reaction, the $\text{PO}_4\text{-P}$ concentration in the solution was unexpectedly high. The reason probably being that when struvite is formed, hydrogen is released, which reduces the solution pH. Due to this, the dissolution of struvite inhibits further precipitation of struvite, and thus, the residual concentration of $\text{PO}_4\text{-P}$ would have increased. In cases where buffering reagents and orthophosphates were added after Mg, $\text{NH}_4\text{-N}$ removal efficiencies were yet as low as 60%, and a similar scenario for $\text{PO}_4\text{-P}$ as before resided. However, a prior addition leads to removal efficiencies as high as 90%. The reason for this can be the reaction taking place between Mg and OH^- ions, leading to the formation of amorphous products such as $\text{Mg}(\text{OH})_2$. This showed that relatively pure struvite could be attained by reducing the occurrence of unexpected reactions.

4.11. Magnesium source

Magnesium needs to be added externally for struvite crystallization to occur. Davis et al. [32] reported that when Mg is supplemented into the system, it competes with Ca for phosphorus and promotes precipitation of struvite. As dosing with Mg economically affects the process (75% of the total operational cost), various other Mg salts have been investigated in laboratory and pilot-scale to lessen the cost while guaranteeing the product quality. MgCl_2 was found to be more effective than MgO and $\text{Mg}(\text{OH})_2$ as it is non-corrosive and non-toxic with high solubility and short dissolution time; however, it is costly and requires alkali supplementation for pH improvement [78]. MgO (N removal efficiency of 60–70%) and $\text{Mg}(\text{OH})_2$ (less solubility compared to other salts, phosphorus removal efficiency – 94%) offer not only Mg ions but also elevate the solution pH [5,78]. Zeng et al. [157] investigated the phosphorus removal efficiencies of various Mg salts at pH 9 with an Mg:P ratio of 1.75 and found the decreasing order of efficiency to be $\text{MgCl}_2 > \text{MgSO}_4 > \text{MgO} > \text{Mg}(\text{OH})_2 > \text{MgCO}_3$. MgCO_3 is the least effective due to its poor solubility Gunay et al. [48]. Seawater has also been studied due to its low cost and found to be useful for P recovery from urine [90, 154]. Few other Mg sources that have also been studied so far with not much successful yield results are magnesite [48], brucite [52], wood ash [122], bone meal [130] and reject saline water [38].

5. Wastewater treatment technologies for struvite production

The global population excretes 3 million tons of P per year through urine and feces at the rate of 1.2 g per person every day. Recovery of such P is critical because of the waning resource and rising demand for fertilizer. In contrast, nitrogen removal from wastewater streams is essential in preventing eutrophication [109]. There are currently various technologies available for the direct recovery of nutrients (mainly phosphorus and nitrogen) from wastewater; yet, the application depends on the source, recovery efficiency, scale of the treatment plant, operational complexity, restraints on chemical usage, and financial scenarios [37,113]. Table 3 enlists the reported technologies, along with working principles and recovery efficiencies for phosphorus and nitrogen.

5.1. Commercialization of struvite technology

Several review papers [82,108] have discussed various aspects of P recovery in the form of struvite. Though they have established several advantages of struvite for wastewater remediation and organic fertilizer production, very few large-scale installations of the available technologies have been implemented all around the world [25]. Table 4 consists of descriptions of some commercially available struvite recovery technologies spread across the globe in countries like Canada, Japan, North America, Netherlands, and Germany. Apart from these, few other large-scale implementations that have not been tested yet. MFC is a device that simultaneously treats wastewater and recovers nutrients in the form of struvite (deposited on cathode surface). It uses conventional electrochemical technology to convert microbial metabolic or enzyme

catalytic energy into electricity with the help of electrogenic microorganisms [74]. Suzuki et al. [136] has developed a pilot-scale crystallization reactor along with an accumulation device for the removal of struvite from swine wastewater and achieved > 95% P removal. Crutchik and Garrido [29] modified the crystallizer into an airlift reactor with a settler for treating wastewater released from fish processing industries by struvite recovery. However, the highest P removal was only 64%. Bhuiyan et al. [18] has optimized phosphorus recovery from pilot-scale crystallizer at Lulu Island Wastewater Treatment Plant (WWTP), Richmond. Apart from these scattered approaches, there have also been a few integrated studies. Kumar and Pal [75] have used the Membrane-Integrated Hybrid Treatment (MIHT) process for struvite recovery from treating coke wastewater using membrane integrated bioremediation which makes the process economically more feasible.

6. Process design

6.1. Reactor design

The composition of wastewater varies depending on the source as reported in the literature over the decades. Total ammonia content ($\text{NH}_3\text{-N}$) varies from 129 mg/L (dairy manure) to 2260 mg/L (swine slurry); total phosphorous varies from 8.5 mg/L (landfill leachate) to 740 mg/L (fresh urine); total Ca varies from 14 mg/L (stored urine) to 245 mg/L (landfill leachate); and Mg concentration varies between 6.7 mg/L (swine wastewater) to 252 mg/L (dairy manure) [81]. Thus based on effluent composition, the downstream process may require different operational supplementary steps (e.g., Ca removal or step-by-step precipitation) as a pre-treatment, which leads to a potential increase in the overall procedure cost.

Diverse materials have been used for the fabrication of struvite reactors. Struvite accumulated most rapidly on stainless steel than PVC (polyvinyl chloride) and least on acrylic [76]. Various reactors have also been developed, of which the most common ones are FBR (Fluidized Bed Reactors), MSR (Mechanically Stirred Reactors), and MFCs. FBRs are the most commonly investigated, while MSRs have been widely applied in industries with careful considerations regarding the speed, as it is a determining factor in struvite crystal size. Mechanically stirred reactors are simple in operation, and the P removal efficacies are relatively high. Regy et al. [118] attained P removals over 60% from synthetic liquors, whereas Seco et al. [125] achieved up to 90% from anaerobic digestion supernatant. However, Seco et al. [125] concluded that higher removal efficiencies in the liquors were due to the presence of Ca, resulting in the co-precipitation along with struvite (76.9%) and calcium phosphate (23.1%).

When it comes to the application at full scale, few problems have been observed, which leads to lower efficiencies. When operated in continuous mode, reactors developed fouling in the impellers. Whenever a high mixing speed (600 rpm) was used to keep particles in suspension and homogenize the solution, the production of a considerable amount of fines was observed, leading to growth limitation [77]. Bowers and Westerman [24] developed a cone-shaped FBR, which can be operated at lower flow rates as a continuous process for swine wastewater treatment. This improved mixing conditions in the reactor as the conical zone created plug flow conditions for the liquid. Warmadewanthi and Liu [147] used an internal self-seeding mechanism by designing a new internal recycling seeding reactor, which improved phosphorous recovery efficiency and reduced the energy required for external recycling. A new technology, MFC, has been developed to treat wastewater and produce electricity simultaneously. The electrogenic microorganism converts the energy stored in chemical bonds of organic compounds to electrical energy [74]. As no precious metals are required, the process is cost and energy-effective compared to a water electrolyzer. In addition, hydrogen gas recovery can be used. However, the phosphorus recovery rate is meager compared to other reactors. Commercial struvite recovery units for treating municipal wastewater sludge

Table 3

Wastewater treatment technologies, their principles, efficiencies, benefits and bottlenecks for recovering phosphorus and nitrogen.

Nutrient recovered: Phosphorus					
Method	Principle	Desorption/ Recovery efficiency (%)	Benefits	Bottlenecks	Ref.
<i>Ion Exchange/ Adsorption</i>	PO_4^{3-} is adsorbed on to a P-selective media and replaced with cations such as Cl^- to produce P-depleted effluent	~ 90	<ul style="list-style-type: none"> High-quality effluent No pH alteration required No additional sludge generated 	<ul style="list-style-type: none"> Media regeneration and treatment before disposal required Leaching out of metal cations which affects the purity 	[109, 126]
<i>Reactive filtration</i>	Multiple mechanisms, including co-precipitation, adsorption and physical filtration to remove P	90.3	<ul style="list-style-type: none"> Orthophosphates can also be removed 	<ul style="list-style-type: none"> Dosing of chemicals such as FeCl_3 is required Recovery of nutrients from backwash requires further treatment 	[99, 109]
<i>Source-separated urine</i>	P recovery by struvite precipitation	77	<ul style="list-style-type: none"> Reduces nutrient load on treatment plants Allows separation of pharmaceuticals Recovery of high concentrations of nutrients (670 mg/L-P and 8180 mg/L-N) 	<ul style="list-style-type: none"> Possible health risks due to incomplete pathogen inactivation Recovery requires chemical addition Public acceptance of onsite nutrient recovery required 	[109, 141, 150]
<i>Struvite precipitation</i>	Struvite is crystallized when the concentrations of PO_4^{3-} , NH_4^+ and Mg^{2+} exceed struvite solubility index	85–99	<ul style="list-style-type: none"> An efficient slow-release fertilizer can be formed with lower impurities. Minimum leaching and lack of fertilizer burn of crops even at high application rates Less-frequent application to fields 	<ul style="list-style-type: none"> Requires chemical addition and pH adjustment The cost of P removal from dilute solutions increases due to chemical requirement 	[109, 156]
<i>Electrodialysis</i>	Anions (e.g., PO_4^{3-} , SO_4^{2-} , and Cl^-) and cations (e.g., NH_4^+ , K^+ , Na^+ , Ca^{2+} , Mg^{2+} and so on) are separated using exchange membranes in the presence of electric field	93	<ul style="list-style-type: none"> Achieves eight times higher concentrated P recovery Used prior to struvite recovery to concentrate effluent for reducing chemical requirements 	<ul style="list-style-type: none"> Heavy metal accumulation in catholyte Requires precipitation with chemical addition for recovery 	[109, 162]
<i>Chemical precipitation</i>	Chemical precipitation by the addition of metal ions (Fe^{3+} , Al^{3+} and Ca^{2+})	–	<ul style="list-style-type: none"> Meets low P discharge limits Easy to operate 	<ul style="list-style-type: none"> Requires chemical dosage Affects preceding biological processes Heavy metal contamination 	[12, 109]
<i>Enhanced Biological Phosphorus removal (EBPR)</i>	Removes P based on the accumulation by organisms (PAOs), resulting in enrichment in the sludge	–	<ul style="list-style-type: none"> Economical compared to chemical precipitation Achieves 1 mg/L-P concentration in effluent at appropriate conditions Simultaneous P and N removal 	<ul style="list-style-type: none"> Sensitive to influent water characteristics Operational complexity Expensive and unstable 	[97, 109]
<i>Electrochemical</i>	Electrochemical nutrient recovery methods can be divided into two categories: 1. Processes with sacrificial anodes 2. Processes with dimensionally stable anodes (DSAs)	90	<ul style="list-style-type: none"> Economical compared to chemical dosing in struvite precipitation Used as an alternative to chemical pH adjustment H_2 generated at the cathode is used as a potential fuel 	<ul style="list-style-type: none"> Co-precipitation of CaCO_3 with shift in pH The energy requirement is inversely proportional to wastewater conductivity 	[64, 109]
Nutrient recovered: Nitrogen (N)					
<i>Ion Exchange/ Adsorption</i>	Zeolites as adsorbent for in NH_4^+ removal	–	<ul style="list-style-type: none"> Effluent meets narrow N discharge limits Operationally simple process 	<ul style="list-style-type: none"> Mainly used for N removal rather than nutrient recovery Adsorption capacity impaired by competing ions Limited resin life Requires chemicals and reagents for media regeneration. 	[71, 109]
<i>Electrochemical</i>	The process involves concentration of ammonia in the cathode compartment of an electrochemical cell	95	<ul style="list-style-type: none"> Produced H_2 can be used as a potential fuel and passive means of NH_3 stripping 	<ul style="list-style-type: none"> Reaction time limited by anode pH N recovery requires stripping and absorption into an acid solution 	[89, 109]
<i>Bioelectrochemical-Microbial Fuel Cell (MFC)</i>	Same principle as electrochemical. In addition, it generates energy required for the process using organic matter	–	<ul style="list-style-type: none"> Generates energy necessary for the process Concentrates N in the cathode compartment for post-recovery Eliminates chemical pH adjustment due to OH^- generation at air cathode H_2 generated at the cathode can be used as potential fuel 	<ul style="list-style-type: none"> Electricity production limited by microbial activity on the anode Sensitive to pH, influent toxicity and substrate loading. 	[109, 148]

(continued on next page)

Table 3 (continued)

Nutrient recovered: Phosphorus					
Method	Principle	Desorption/ Recovery efficiency (%)	Benefits	Bottlenecks	Ref.
Struvite precipitation	Struvite precipitation carried out by adding both Mg^{2+} and PO_4^{3-}	–	<ul style="list-style-type: none"> Lower evaporative losses compared to other N rich fertilizer 	<ul style="list-style-type: none"> Both Mg^{2+} and PO_4^{3-} need to be added for N recovery High operational costs due to chemical requirement 	[109]
Stripping and absorption	Ammonia is stripped from pH elevated concentrated solutions (> 1000 mg/L) and recovered into acid solution	94	<ul style="list-style-type: none"> Direct recovery of N in fertilizer form Easy operation and installation Energy requirement for NH_3 stripping and adsorption from urine is economical 	<ul style="list-style-type: none"> Requires pH adjustment Typically used for high strength wastewater Energy requirement for stripping is inversely proportional to influent NH_3 concentration Not suitable for cold climates 	[79, 109]
Gas permeable membranes	NH_3 is recovered as $(NH_4)_2SO_4$ by recirculating H_2SO_4 in tubular membranes submerged in ammonia-containing wastewater or vice versa	95	<ul style="list-style-type: none"> Direct recovery of N in fertilizer form is possible pH shifting by low rate aeration is sufficient to run the process 	<ul style="list-style-type: none"> Typically used for high strength wastewater < 50 mg/L NH_3 High capital costs are expected when effluent NH_3 is < 1 mg/L 	[31,42, 109]

have been operating in many countries, as explained in Section 3. Recovery of struvite from urine has been demonstrated at the pilot scale in Nepal without the need for any pre-treatments [46].

6.2. Pre-treatments to improve struvite recovery efficiency

Pre-treatments play an essential role in removing the interfering ions from wastewater. If P can be mobilized in available form and released from the solid fraction, its recovery can be maximized. Though the feasibility of pre-treatments has been studied experimentally, its economic implications on industrial applications need to be investigated.

Organic nutrients hydrolyze and mineralize in anaerobically digested pig, cattle, and poultry manure, resulting in increased availability of N, P, and Mg, thereby increasing the potential for struvite crystallization [10]. When the pre-treatment process followed by anaerobic digestion was used to treat raw swine wastewater, the phosphorus recovery increased from 92% to 98% [93]. Another pre-treatment method is acidification, which drops the ionic product below their equilibrium solubility product, causing protonation of P ions from bound phosphates (Ca/Mg/Fe phosphate). An advantage of this process is the higher release of particulate P into the solution, increasing its availability [161]; however, it may contaminate the crystallized struvite due to the release of metal ions. P availability can also be enhanced by treating with essential reagents to increase alkalinity, leading to an increase of 50–70% P in dried sewage sludge, as demonstrated by [134]. The inhibitory effect of Ca on struvite crystallization can be minimized by using chelating agents (e.g., ethylenediaminetetraacetic acid (EDTA) and oxalic acid) as a pre-treatment method [161]. The studies revealed that the ligands react with $Ca-PO_4^{3-}$ to form Ca-EDTA complexes, and PO_4^{3-} is released. However, the use of EDTA for chelating Ca and augmenting struvite formation needs optimization as additional EDTA might successively bind to Mg, thereby regulating Mg availability, a prerequisite for struvite formation. When microwave irradiation is used as a pre-treatment method, it leads to particulate P release, which increases struvite recovery [88].

The degree of P release depends upon the different forms present in the source effluent and its extent of solubilization depends upon the working temperature and duration of heating [83]. A temperature of $120^\circ C$ is considered optimum for the release of both NH_4^+ and P in sewage sludge [28]. Microwave treatment is advantageous over conventional heating as it involves uniform heating throughout the material, provides precise control over operating temperature, and prevents direct contact between heating source and materials [88]. This process has been further modified by combining chemical assistance with

microwave digestion using common chemicals like oxidants, acids and bases [103]. As demonstrated by Pan et al. [103] dairy slurry, when microwaved, the phosphorus release was 80% but increased to 85% when H_2O_2 treatment was also incorporated. Xiao et al. [149] showed that when activated sludge was microwaved for 1 h before anaerobic stirring, P release was maximized with a recovery efficacy of 95% as amorphous calcium phosphate and struvite. Lastly, EBPR is gaining popularity where sludge bacteria (PAOs) release and uptake large quantities of PO_4^{3-} due to alternating anaerobic and aerobic steps resulting in increased P removal [17]. As reported by Münch and Barr [96], precipitation of struvite from EBPR in wastewater treatment plants leads to 58–94% P recovery.

6.3. Costs incurred

Although struvite crystallization reduces phosphorus in wastewater, the usage of the product for commercial applications can only be successful depending on its economic sustainability, where the production cost and market price of struvite should be considered. According to [56], recovery of 1 kg of struvite from a wastewater treatment plant will reduce sludge handling and disposal and make the operational process economical by lowering costs by AUD (Australian dollar) 1.13. In the case of chemical precipitation of struvite, the chemical cost plays a significant part in the overall process [27,58]. The material costs in electrochemical precipitation are mainly governed by Mg dosage; the additional energy costs for the electrochemical process were negligible. In comparison, $MgSO_4$ and the Mg anode cost about the same (3.45 and 4.50€ kg^{-1} struvite), while MgO turns out to be the cheapest option (0.85€ kg^{-1} struvite), and $MgCl_2$ was the most expensive Mg source (6.80€ kg^{-1} struvite). Due to the variable nature of different wastewaters, sometimes pre-treatment might also be required, which increases the overall process cost (e.g., dairy wastewater always has a high Ca concentration, which unfavorably affects precipitation rate and product quality). Cost involving the development of reactors also affects the production cost of struvite [114]. Jardin and Pöpel [60] claimed that even if only 20% of the P (in mass) in sewage were recovered through struvite crystallization, it could save 640,000 USD (United States dollar) annually. When focusing only on P recovery, pH adjustment is costlier than Mg addition.

Most market price estimations when it comes to selling the recovered struvite are based on its application as a fertilizer. Geerts et al. [44] predicted a guaranteed discounted payback of 10 years for an installation at Leuven, Belgium, at a selling price of 430 US dollars per ton. The orthophosphate concentration also influences the cost of struvite in the

Table 4

Commercial installations for struvite recovery along with reactor design and process description.

Technology	Process description	Place of installation	P recovery (%)	Ref.
Pearl™ Technology (marketed under the commercial name Crystal Green)	Reactor type: Fluidized bed Source of WW: Effluent containing high PO_4^{3-} and NH_4^+ Final product: Struvite	North America, UK	80	[25, 67]
Ostara	Reactor type: Fluidized bed Source of WW: Dewatering liquor and centrate after anaerobic digestion Reagents used: MgCl_2 , NaOH Final product: Struvite	USA	85–95	[45]
Phospaq™	Reactor type: Stirred tank Source of WW: Sewage sludge effluent following dewatering, combined with potato factory effluent Reagents used: MgO Final product: Struvite	Netherlands, UK	80	[45, 106]
Phosnix	Reactor type: Fluidized bed Source of WW: Sewage sludge treatment effluent Reagents used: MgOH, NaOH Final product: Struvite	Japan	90	[45, 142]
AirPrex™	Reactor type: Stirred tank Source of WW: Digested sludge before dewatering Reagents used: MgCl_2 , NaOH Final product: Struvite	Germany, Netherlands	85–90	[45, 110]
Multiform	Reactor type: Fluidized bed Source of WW: Food processing and swine farm waste, agricultural waste streams, municipal wastewater Reagents used: MgCl_2 , caustic soda Final product: Struvite	America	80–90	[45, 107]
DHV Crystalactor	Reactor type: Fluidized bed Source of WW: PO_4^{3-} rich wastewater Reagents used: MgCl_2 , caustic soda Final product: Calcium phosphate	–	70–80	[45]
NuReSeys	Reactor type: Stirred tank	–	85	[45, 92]

Table 4 (continued)

Technology	Process description	Place of installation	P recovery (%)	Ref.
	Source of WW: Municipal wastewater Reagents used: MgCl_2 , NaOH Final product: Struvite			
ANPHOS	Reactor type: Stirred tank Source of WW: Anaerobic digester supernatant Reagents used: MgO/Mg(OH)_2 , NaOH Final product: Struvite	Netherlands	80–90	[45]
Seaborne	Reactor type: Stirred tank Source of WW: Wastewater slurry Reagents used: MgOH, NaOH Final product: Struvite	Germany	–	[45]

(WW-Wastewater).

wastewater. When the PO_4^{3-} -P concentration in the wastewater increased from 50 to 800 mg/L, the value of struvite dropped from 2800 to 520€/t MAP [36]. Recovery of struvite also reduced the landfill cost.

In conclusion, struvite production can be made feasible by searching for economic alternatives to add Mg and pH optimization. Costs can be further reduced by using the solar energy supply to power the reactors, and using wastewater sources that do not require much pre-treatment process can be collected and stored without incurring additional costs.

7. Uses of struvite

7.1. Struvite application as fertilizer

The accumulation of urine in the ecosystem causes eutrophication in the water bodies and leads to the suffocation of fish and aquatic organisms. Struvite precipitates naturally in aged urine when the pH reaches a certain level. As a result of the formation of struvite, a reduction in the weight and volume of urine can be identified. The nutrients are bio-available and hence can be readily absorbed by plants. The handling of the struvite is more accessible as it can be produced in powder form, and it acts as a slow-release fertilizer [161]. The low heavy metal content of struvite, when compared to other P-containing commercial fertilizers, supports its use as fertilizer [80].

Struvite utilized as agricultural fertilizer serves various advantages such as waste management, land reclamation, and lower water footprints through reclaimed disinfected water for irrigation. Struvite production improves the agricultural livelihood of farmers by reducing the expenditures associated with the help of chemical fertilizers and generating incomes through the saleable struvite agglomerates. It reduces the dependency on P rocks and provides a better livelihood for farmers and people in rural areas [161].

Struvite crystals produced from the wastewater nutrient source have the property of utilization as an organic fertilizer [87]. These crystals can be refined to obtain granules of an acceptable size that can be applied on land for fertilization purposes. Struvite-based fertilizers are of less harm to the plant roots, and they have a low content of heavy metal ions than commercially available fertilizers [80]. Struvite precipitation is technologically most accessible when it comes to nutrient

recovery from urine, as it does not necessarily involve any sophisticated operations [133]. In cases where direct application of urine is not a feasible option, and external sources of nutrients are often not economical or environmentally friendly, struvite may serve as an intermediary solution between on-farm nutrient recycling and global nutrient transfer [114]. Struvite serves as an eco-friendly fertilizer due to its slow nutrient loss by leaching and fertilizer quality. The acute shortage of rock phosphate can be overcome by connecting all the municipal and livestock wastewater with wastewater treatment plants for mass P recovery [66,67].

According to its chemical composition, struvite has an NPK-value of approximately 6:29:0:(Mg) 10. It has advantages over other fertilizers because of its properties like slow nutrient release and good phosphorus availability [133]. It was found that there is no significant dissimilarity between struvite and mono-calcium phosphate, which is a source of phosphorous that is water-soluble and also considered to be available to the plant completely [87]. However, for optimal growth, the plant-available nitrogen is not enough, and higher struvite application to the plants can significantly raise the soil pH level to a degree where nutrient availability and uptake by the plants are adversely affected [66, 67]. The average amount of nutrients that should be applied to the soil, in general, should follow the order $N > K > P > Mg$. A combination of other fertilizers, along with struvite, should be developed for optimal use that significantly has an increased nitrogen content [80]. Production of struvite from wastewater nutrient source and its utilization as chemical fertilizer for crop production has been shown in Fig. 5.

7.2. Other applications of struvite

Struvite has also been reported to be used for building coatings with simultaneous photocatalytic activity for clean-up of surface adsorbed pollutants as it can form a robust complex and has the ability to capture TiO_2 [21]. Başakçılardan-Kabakci et al. [16] used recovered struvite as pollutant removal to remove tetracycline. Due to its large surface area, Foletto et al. [39] documented the potential use of struvite as an adsorbent for dyes. It can also overcome the low volatility of radioactive cesium and be used as an excellent host for its immobilization [144]. The potential use of struvite as a substitute nutrient for microalgae culturing (*Chlorella vulgaris*) was tested [91]. Struvite recapture can be positively incorporated into the composting process to increase the agronomic value of compost. Due to high quality and negligible heavy metal content, the market value of struvite has become significant as compared to phosphorus ore [156]. Struvite satisfies the need for slow-release fertilizer and has many horticulture benefits, such as in nurseries, golf courses, and boutique fertilizers [6].

8. Research gap

This research area is relatively young, with the number of publications increasing only since the last decade. Even though struvite precipitation has been well studied, gaps and inadequacies in this field still exist, preventing its widespread acceptance by industry. In this extensive literature survey conducted, it was found that several factors (i.e., influent composition and foreign ions) and control variables (i.e., pH, temperature, and mixing) influence the struvite crystallization process. However, most of these publications mimicked wastewater compositions instead of using real-time wastewater sources, which creates problems in scale-up by hindering industrial applications. Lab-scale studies on using real-time wastewater such as urine have been reported to show better performance on struvite recovery than synthetic proxy [22,51,59]. Source-separated collection and storage of wastewater need more attention for effective processing and struvite recovery. Ways to prevent ureolysis during storage to prevent ammonia volatilization and enhance ureolysis to reduce the storage period without adding chemical reagents should be researched for large-scale application and decentralization [117]. Future research on finding the source of wastewater, which is best suitable for the process and the reagents with an economical price range for precipitation, is recommended. The incorporation of struvite crystallization with other techniques for pre-treatment or post-processing has improved nutrient recovery. Such aspects can be widely investigated to reduce the overall cost while uncompromising the efficiency. For example, drying of struvite crystals has been known to promote sterilization by means of bacterial inactivation [20]. In addition, its role in determining the composition and type of compounds formed (struvite and its analogs) requires further investigation [138]. The current research focusses on phosphorous removal/reduction to satisfy jurisdictional requirements, rather than on nutrient recovery for sustainable wastewater management or finding a potential renewable source of phosphorus fertilizers [109,114]. This ideology should be reformed and centered towards nutrient recovery, on the whole to satisfy the need for potential organic fertilizer. In this regard, struvite recovery from wastewater has gained limelight; however, the literature data available are mainly in the lab-scale level and insufficient for the scale-up [128,41,66]. Correspondingly, minimal research on product quality measurement and control (i.e., heavy metal reduction and process control) has been performed and requires profound exploration for industrial applications on developing a potential market base. This applies to struvite application on plant growth and field studies. The data on crops cultivated with struvite incorporation is minimal. The mechanism behind nutrient uptake from struvite by the plants is yet to be explored. Further research is encouraged on process

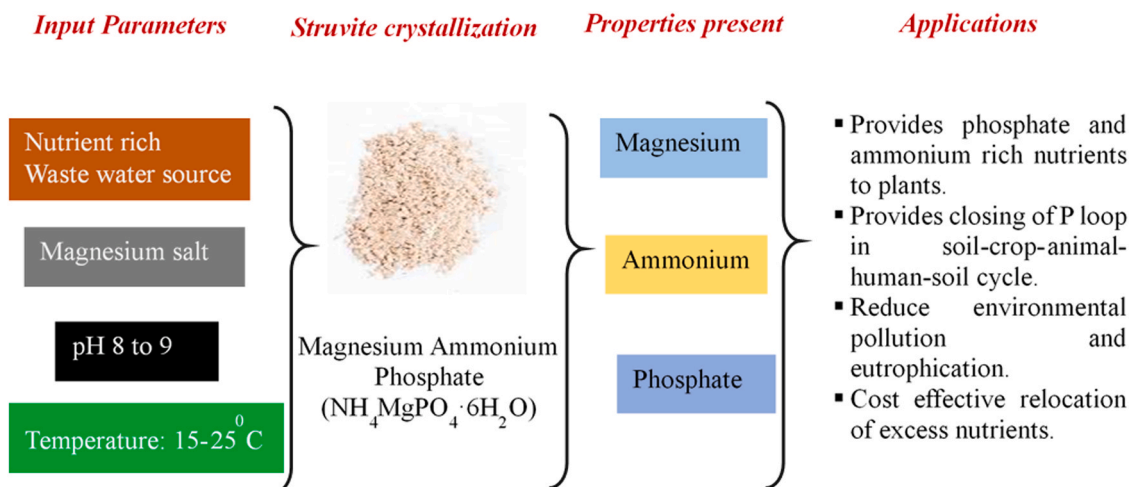


Fig. 5. Struvite precipitation and utilization as a chemical fertilizer.

optimization and cost analysis to advance the current understanding of process economics and viability. The biorefinery aspect of combining the cultivation of potential microbial strains along with struvite precipitation can be considered for advanced studies [2,65].

9. Summary and future perspectives

Struvite crystallization is a feasible management technique that can mitigate the depletion of phosphorus rocks. But current nutrient recovery processes are focused principally on individual recovery of P or N, with process selection dependent on a wide array of factors, including influent wastewater characteristics, recovery goals, operating constraints regarding the use of chemicals, the need for separate conveyance network, the requirement for onsite treatment, and the need for trained operators. Mg sources free of impurities with actual soluble content improve struvite quality, and augment its suitability as fertilizer. Among the currently developed nutrient recovery technologies discussed earlier, only a few produce fertilizer with minimal post-processing. Even though chemical precipitation separates P from solution, the precipitates are typically wasted in a treatment plant along with the sludge, creating a bulky product. In contrast, electrochemical processes without increasing the sludge volume can recover P either in the form of struvite or calcium phosphates. Further, the much purer product can be obtained using electrochemical methods after biological P accumulation by EBPR from anaerobic sludge digester supernatant. In addition, electrochemical processes serve the purpose as they minimize chemical use. All the recovery processes work efficiently when the nutrients are concentrated. The speed of operation can be dramatically increased by integrating chemical and biological treatments with modern membrane-based units. The overall process can be made economical by producing value-added struvite fertilizers from this recycled nitrogen and phosphorus with the addition of Mg chemically or electrochemically. Thus the major observations from this review are as follows:

- Despite the number of technologies studied, the demonstration of the same remains limited to lab-scale and are yet to be upscaled for commercial applications.
- Among the number of wastewater resources available, the one with the high nutrient concentration, requiring no pre-treatment and feasible with all technologies need to be chosen.
- Though a number of factors influence struvite precipitation, pH remains to be the most significant factor despite the technology used.
- Lack of data for scaling up the technologies to a commercial level is a major obstacle in adapting struvite precipitation as means of nutrient recovery and wastewater remediation.
- The additional costs involved in the struvite precipitation can be compensated by its commercial value and application in a variety of areas including hydrogen gas production as an alternate fuel option.

Through this extensive review conducted, it can be found that there is a lack of literature availability that focusses on the betterment of process design, data for scale-up of existing technologies, and product quality and control measurements for comprehensive industrial application. The role played by the different components of wastewater, which varies from one source to another, also falls in the gray area when it comes to the production of struvite. Further, not much light is shed on the collection of wastewaters and their storage for pilot-scale production.

The hazard of eutrophication in the water bodies can be significantly reduced by producing struvite from wastewaters by removing P and N. Small-scale and commercial struvite production plants should be established at the wastewater production sites across the world. The future prospects in the improvement of the overall struvite process and its commercialization lie in the following aspects:

- Addressing issues regarding the appropriate chemical inputs, reactor designing, and cost dynamics related to investments and payback time.
- Finding an alternate Mg source (replacing high-grade Mg input with cheap Mg-rich by-products) requires optimum strategies that will enhance the reaction's overall efficiency, including removal of non-participating ions (like Ca^{2+} , Al^{3+} , Fe^{3+} , etc.).
- Wastewater with significant phosphorus concentrations should be studied to widen its application. Seed material addition as a part of process can be used to increase particle size and for improvement of product purity. However, they increase the process complexity and overall cost.
- Further detailed investigation on the efficiency of struvite as fertilizer on various crops concerning soil conditions, climatic factors, and other biotic and abiotic components of the environment will play a significant role in developing the market for struvite as commercial fertilizer.
- Additional work also needs to be carried out to evaluate the product purity and particle size control for better product quality and economic benefit.

The strategy for market development of struvite lies in adopting a more holistic approach considering pricing, purity, size, storage, transportation, and distribution, taking into account the legal structure of contaminants and eco-toxicity. This will help develop a sustainable and eco-friendly P-rich fertilizer that can be used as a complement to the prevailing nutrient supply system. The impacts of such technological successes would be profound on global food security and closing the nutrient loop related to sanitation that is crucial because of the critical need to address the looming phosphorus crisis.

CRedit authorship contribution statement

Nageshwari Krishnamoorthy: Conceptualization, Data curation, Writing - original draft. **Baishali Dey:** Conceptualization, Data curation, Writing - original draft. **Yuwalee Unpaprom:** Visualization, Funding acquisition, Writing - review & editing. **Rameshprabu Ramaraj:** Visualization, Investigation. **Gaanty Pragas Maniam:** Project administration, Funding acquisition, Writing - review & editing. **Natanamurugaraj Govindan:** Project administration, Investigation. **Sivaraman Jayaraman:** Supervision. **Thirugnanam Arunachalam:** Supervision. **Balasubramanian Paramasivan:** Conceptualization, Funding acquisition, Writing - review & editing, Final approval.

Declaration of Competing Interest

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

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