



# A two-stage treatment process for the management of produced ammonium by-products in ureolytic bio-cementation process

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## Abstract

Since the last decade, microbial-induced carbonate precipitation has been proposed as an environmentally friendly technique to improve the engineering properties of soil. Despite the considerable progress on ureolytic bio-cementation, one of the major concerns that has not been solved yet is related to the production of ammonium. This study aimed to manage ammonium ions to attain a sustainable pathway for bio-cementation treatment. To this end, a two-stage treatment including rinsing of ammonium from soil combined with a recovery of ammonium was considered herein for the first time. In the rinsing process, the followings were studied to optimize ammonium removal from soil: the effects of pH, concentration, and the single salts of the rinse solution. In the subsequent ammonium recovery process, the effects of phosphate source, pH, molar ratio, and  $\text{Ca}^{2+}$  ions were extensively investigated. The results revealed that at neutral pH conditions, ammonium removal was the lowest (68.82%). The  $\text{MgCl}_2$  solution was found to have the greatest ability to remove ammonium followed by  $\text{CaCl}_2$ ,  $\text{NaCl}$ , and distilled water (98.54%, 96.47%, 89.95%, and 74.77%, respectively). The ammonium recovery results showed that 86.8% of ammonium ions could be recovered as a high purity struvite (~94%), and that the optimum recovery was achieved at the following conditions:  $\text{Na}_2\text{HPO}_4$  as a phosphate source, the  $\text{Mg}^{2+}:\text{NH}_4^+:\text{PO}_4^{3-}$  molar ratio of 1.2: 1: 1, and a pH of 8.5. Overall, it was demonstrated that the proposed method could be an effective way for sustainable ammonium by-products management.

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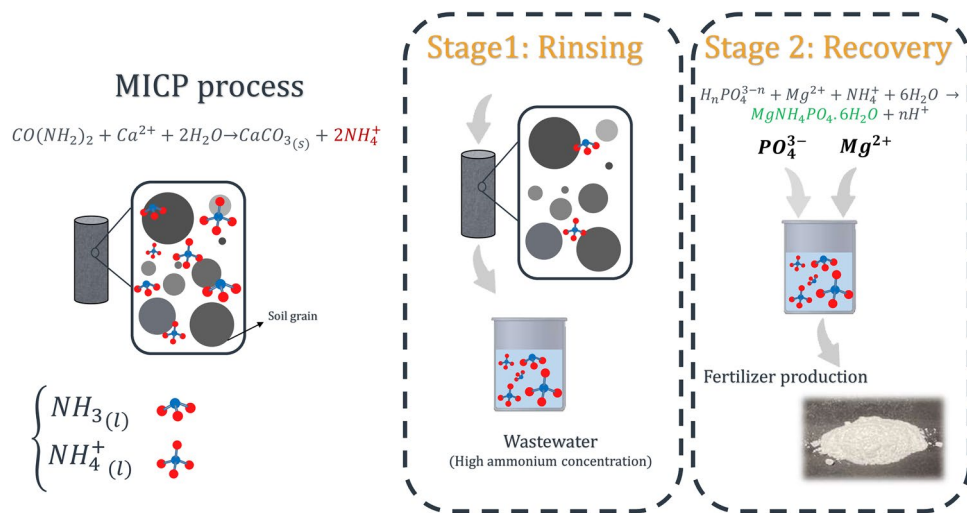
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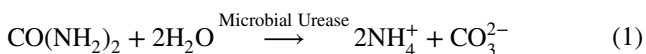
## Graphic abstract



**Keywords** Ammonium by-products · Desorption · Environmental management · Microbial induced carbonate precipitation · Recovery

## Introduction

Recent developments in the transition toward innovative and sustainable development in the engineering field have taken place by emerging multidisciplinary approaches aiming at reducing embodied energy and carbon (cutting greenhouse gas) emissions (DeJong et al. 2013). One such prominent technology in civil and environmental engineering is to use the process of bio-mineralization, more specifically microbial induced carbonate precipitation (MICP). This process utilizes the mediation of biological substances to induce the cementation in situ, thereby enhancing the strength and stiffness of soils (DeJong et al. 2010; Kakelar and Ebrahimi 2016). In MICP by urea hydrolysis, urease-producing bacteria are employed to catalyze the hydrolysis of urea into carbonate and ammonium (Eq. 1). Produced carbonate ions immediately react with existing or supplied calcium ions and form calcium carbonate crystals in the reaction medium (Eq. 2) (Whiffin 2004).



Following the success of laboratory level experiments of MICP with different purposes, such as increasing strength and stiffness (Amarakoon and Kawasaki 2018; DeJong et al. 2010; van Paassen et al. 2010), enhancing slope stability (Gowthaman et al. 2019, 2020), mitigating liquefaction (Montoya et al. 2013; Moosazadeh et al. 2019), and

preventing wind erosion and coastal erosion (Daryono et al. 2020; Maleki et al. 2016), the application of MICP is becoming increasingly popular in civil engineering. Despite the enormous potential of MICP technology as a sustainable soil-improvement technique, developing a widely successful application requires a detailed understanding and managing of complexities, especially in the case of by-products (e.g., ammonium ions) (DeJong et al. 2013; Ivanov et al. 2019).

Ammonium products (ions and/or free ammonia) are often regarded toxic, and their release could lead to various environmental issues, including eutrophication, depletion of dissolved oxygen (Ye et al. 2018), blue baby syndrome, and cancers (Shah and Mitch 2012). Mainly, living organisms essentially require nitrogen compounds (Zheng and Wang 2009). Nevertheless, exceeding the detoxify capacity for the ammonium nitrogen's aqueous concentrations of ( $\text{NH}_4^+$ ) would result in severe health problems in living species like animals and humans (Paerl 1997). Untreated MICP effluents remaining in soils possibly pollute the groundwater and water bodies like rivers and lakes by runoff (Massoudinejad et al. 2019). Higher aqueous ammonia in surface water improves toxic algae blooms, reduces dissolved oxygen, and yields aquatic toxicity; the procedure is denoted as eutrophication (Paerl 1997). The highest reported concentrations for aquatic life are 17 mg/L (about 1 mmol/L) and 1.9 mg/L (about 0.1 mmol/L) total  $\text{NH}_4^+$ , respectively, for chronic and acute exposure (Huff et al. 2013). From the reaction stoichiometry (Eq. 1), it is expected to produce ammonium with a concentration twice the urea concentration. Based on former reports, ammonia production over MICP was regularly



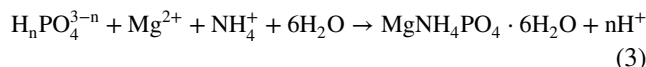
within the range between 50 and 500 mmol/L (Lee et al. 2019a, b; Martinez et al. 2013). For instance, sandy soil bio-cementation needs 62 kg of calcium carbonate per 1 m<sup>3</sup> of sand, causing the release of ~22 kg of ammonium (Ivanov et al. 2019). This indicates that MICP effluents need proper post-treatments to eliminate or recover ammonium and to satisfy water or wastewater quality standards. Bio-cementation technology faces a significant barrier that is the limited comprehension of post-treatment NH<sub>4</sub><sup>+</sup> removal, hindering environmental benefits and field-scale applications. Thus, to improve the efficiency of MICP application in geotechnical engineering, it is essential to manage and recover produced ammonium in the soil treatment process.

Various biological (Ma et al. 2016) and physicochemical treatment mechanisms such as ion exchange, nitrification–denitrification, air stripping, and adsorption were proposed in the past for ammonium nitrogen removal in different industries (Huang et al. 2018; Rahmani et al. 2004; Ye et al. 2018). However, these mechanisms are laborious, likely impractical, and operating cost-intensive in geotechnical engineering projects as they require considerable modifications in the condition of oxygen availability in treatment zones and chemical factors. Gat et al. (2017) revealed that the in situ oxidation of NH<sub>4</sub><sup>+</sup> could impose detrimental impacts on bio-cementation integrity resultant from the increased acidity in aqueous solutions. Supplementary strategies are required to address produced ammonium ions over bio-cementation.

Currently, only very few studies have assessed possible approaches to remove ammonium by-products following MICP. Zeolite reagents have been commonly used to adsorb ammonium ions from the effluent of treated areas with consequent relevant remediation methods. For example, Putra et al. (2017) conducted a series of trials, in which they mixed natural zeolite with prepared grouting solutions. They found that the addition of 10 g natural zeolite/L solution could

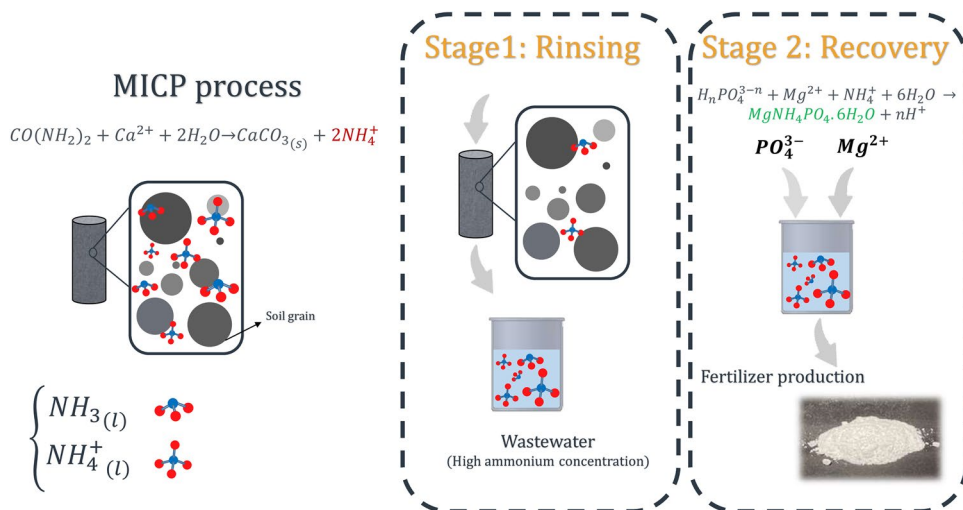
remove 75% and 45% of ammonium in reagent concentrations of 0.5 mol/L and 1.0 mol/L, respectively. Keykha et al. (2018) used the natural aluminosilicate treatment to remove NH<sub>4</sub><sup>+</sup> from the aqueous solution of CO<sub>3</sub><sup>2-</sup> and achieved the standard ammonium concentration level in a cement solution. Recently, Lee et al. (2019b) have suggested that the ionic strength of the rinse solution is the main factor to determine the removal of NH<sub>4</sub><sup>+</sup> and the pH of the rinse solution has only a minor effect. However, only very limited works concentrated on possibilities of removing ammonium from the MICP system (effluents and also the cemented area), especially by focusing on ammonium recovery. To promote the MICP completely environmental-friendly, possible novel mechanisms should be addressed in further studies to manage or remediate ammonium from the reaction system.

Hence, in this work, experiments were performed to assess the struvite (MgNH<sub>4</sub>PO<sub>4</sub>·6H<sub>2</sub>O) precipitation's applicability for ammonium removal from the reaction effluent system. The struvite mineralization, also called magnesium ammonium phosphate, is a potential nutrient recovery technique that can be effectively used to remove NH<sub>4</sub><sup>+</sup> and phosphate wastes (Jia et al. 2017).



It should be noted that this struvite technology has not been assessed so far for the MICP-ammonium recovery purpose. Thus, the simulated-MICP effluent (SME) was prepared synthetically and used in this investigation. According to Fig. 1, the study included two stages: first, washing ammonium from sand, and second, the precipitation of ammonium as struvite. In the first stage, rinsing conditions including the pH level, the type of salts, and the rinse solution's concentration were evaluated for optimizing ammonium removal from soil. In the second stage, the effects of pH, calcium concentrations, and molar ratio were investigated on the struvite

**Fig. 1** The conceptual illustration of the two-stage treatment for the recovery of produced ammonium by-products in the MICP process



precipitation. It should be mentioned that all the experiments were carried out at the Biotechnology for Resources Engineering Laboratory of the Hokkaido University between December 2019 and June 2020.

## Materials and methods

### Soil material

For this research work, soil obtained from Onuma (Hokkaido, Japan) was used (42.388532N; 140.284762E). Soil can be classified as poorly graded fine sand in accordance with the unified soil classification system (USCS), with the mean particle diameter of 0.23 mm (ASTM 2011). The grain size distribution curve and summary of the XRF analysis results of soil are presented in Fig. S1A (refer supplementary file) and Table 1, respectively.

### Soil columns

To check the efficiency of different solutions in rinsing ammonium ions from soil, small-scale sand column tests were conducted. Columns were prepared using 50-mL standard syringes with a filter paper at bottom. The predetermined weight of soil was added to the columns, and then, soil was packed using a custom-made, controlled-drop hammer fitting inside the column to achieve a desired compaction density of around  $1.5 \pm 0.1 \text{ g/cm}^3$ .

### Chemicals

In this initial study, the effluent solution was prepared synthetically to reasonably simulate the MICP effluent, which is hereafter referred to as SME. It should be noted that due to complexity in effluent compositions, the direct use of MICP effluents was avoided, and instead, SME was used to clearly demonstrate all the effects in the post-treatment of ammonium by-products with reduced complexity and errors. Based on previous reports, effluents generated in the MICP process of each cycle contain ammonium ions in a range between 0.05 and 1 mol/L (Ivanov and Stabnikov 2017; Martinez et al. 2013). SME was prepared by mixing the predetermined amount of the  $\text{NH}_4\text{Cl}$  reagent (0.25 mol/L) in deionized water. Required stock SME was prepared 15 min prior to each experiment to reduce the possible evaporation of  $\text{NH}_3$  (Capdevielle et al. 2013). To prepare solutions,

depending on the tests, variable amounts of  $\text{NH}_4\text{Cl}$ ,  $\text{CaCl}_2$ ,  $\text{MgCl}_2$ ,  $\text{Na}_2\text{HPO}_4$ ,  $\text{K}_2\text{HPO}_4$ , and  $\text{NaCl}$  powder were dissolved into deionized water. All the used reagents were of analytical grade.

## Experimental procedures

### Rinsing of ammonium ions from sample

**Effect of rinse solution** The experiments were performed to evaluate the role of rinse solution chemistry, including solution pH, solution concentration, and different ions ( $\text{Na}^+$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$ ), in ammonium removal from the soil samples. The experimental procedures involved the following steps: (1) adding 25 mL (1.1 PV) of SME to each sample and then allowing it to stay in the column for 22 h until soil was completely saturated with  $\text{NH}_4^+$  ions (this time period was considered the same as that in each cycle in the MICP process) and (2) subjecting each column to 130 mL rinse solution (~6 PV) to promote the desorption and removal of ammonium ions. It should be noted that the columns were gravity drained.

To measure how many ammonium ions were washed in each cycle, outlet solutions were collected in 10-mL portions and mass balance calculations were used to determine the percentage of removing ammonium.

### Recovery of ammonium ions by struvite

**Effect of  $\text{Na}^+$  and  $\text{K}^+$  in struvite precipitation tests in different pH** Batch experiments were conducted to evaluate the effectiveness of  $\text{Na}^+$  and  $\text{K}^+$  ( $\text{Na}_2\text{HPO}_4$  and  $\text{K}_2\text{HPO}_4$  as a phosphate source) and the effect of pH on ammonium recovery by struvite precipitation from SME. Three replicate runs of struvite crystallization were carried out for each variable pH of 6.5, 7.5, 8.5, 9.5 and 10.5. In each struvite precipitation test, magnesium ( $\text{MgCl}_2$ ) and phosphate were added to 50 mL of SME at  $\text{Mg}^{2+}:\text{NH}_4^+:\text{PO}_4^{3-}$  molar ratios of 1:1:1. The mixed solution (total volume = 50 mL) was continuously stirred at 300 rpm for approximately 15 min using a magnetic stirrer until a stable pH value was achieved. A pH-meter probe was immersed in the solution to continuously monitor and measure pH values. During the precipitation process,  $\text{NaOH}$  and  $\text{HCl}$  were utilized to adjust the pH of the samples at the desired value. Then, the formed struvite was allowed to precipitate in the next 30 min. Afterward, 1.5 mL supernatant was filtered using a 0.22- $\mu\text{m}$  membrane filter

**Table 1** The mineralogical composition of Onuma sand

Onuma sand	Oxide										
	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	P <sub>2</sub> O <sub>5</sub>	SO <sub>3</sub>	K <sub>2</sub> O	CaO	TiO <sub>2</sub>	V <sub>2</sub> O <sub>5</sub>	MnO	Fe <sub>2</sub> O <sub>3</sub>
Percentage (%)	1.99	25.6	57.5	0.302	0.672	1.05	3.68	0.583	0.0278	0.137	8.1

for the composition analysis and the precipitates were collected. Subsequently, the samples were subjected to freezing at  $-20\text{ }^{\circ}\text{C}$  until further analysis. All the experiments were triplicated for each case and carried out at constant room temperature conditions in accordance with the procedures defined above.

**Effect of molar ratio** To evaluate the effect of  $\text{Mg}^{2+}$ ,  $\text{NH}_4^+$ , and  $\text{PO}_4^{3-}$  concentrations on struvite precipitation, batch experiments were carried out. In addition to the stoichiometric ratio of struvite ( $\text{Mg}^{2+}:\text{NH}_4^+:\text{PO}_4^{3-} = 1:1:1$ ), the experiments were performed with different chemical dosages at pH 8.5 (Table 2). As per the procedures described in Sect. 0, the mixed solution was treated.

**Effect of calcium in SME** The wastewater collected from the outlet of the MICP process contains calcium at different concentrations. The cation ( $\text{Ca}^{2+}$ ) presenting in the MICP effluent might substantially influence the formation of struvite, as  $\text{Ca}^{2+}$  can compete with  $\text{NH}_4^+$  and  $\text{Mg}^{2+}$  to form calcium phosphate compounds. To figure out the effect of  $\text{Ca}^{2+}$  on struvite precipitation,  $\text{CaCl}_2$  was added to SME so that SME could be

**Table 2** Molar ratios of  $\text{Mg}^{2+}:\text{NH}_4^+:\text{PO}_4^{3-}$  for ammonium recovery

Effect of $[\text{Mg}^{2+}]$	Effect of $[\text{PO}_4^{3-}]$	Effect of $[\text{Mg}^{2+}]$ and $[\text{PO}_4^{3-}]$
0.8:1:1	1:1:0.8	0.8:1:0.8
1:1:1	1:1:1	1:1:1
1.2:1:1	1:1:1.2	1.2:1:1.2
1.4:1:1	1:1:1.4	1.4:1:1.4

**Table 3** Different chemical compounds observed during struvite crystallization

Name	Formula	Jia et al. (2017)	Bayo et al. (2015)	Kumari et al. (2020)	Hu et al. (2020)	Liu and Wang (2019)
Struvite	$\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$	+	+	+	+	+
Struvite—Na	$\text{MgNaPO}_4 \cdot 6\text{H}_2\text{O}$	—	—	—	+	—
Struvite—K	$\text{MgKPO}_4 \cdot 6\text{H}_2\text{O}$	+	—	—	+	—
Monenite	$\text{CaHPO}_4$	+	—	—	—	—
Magnesite	$\text{MgCO}_3$	—	—	—	—	—
Hydroxyapatite	$\text{Ca}_5(\text{PO}_4)_3(\text{OH})$	—	—	—	—	+
Dolomite	$\text{CaMg}(\text{CO}_3)_2$	—	+	—	—	—
Octacalcium phosphate	$\text{Ca}_8(\text{HPO}_4)_2(\text{PO}_4)_5 \cdot 5\text{H}_2\text{O}$	—	—	—	—	+
Brucite	$\text{MgCO}_3$	—	—	+	—	—
Trimagnesium phosphate	$\text{Mg}_3(\text{PO}_4)_2 \cdot 22\text{H}_2\text{O}$	—	—	—	—	+
Newberyite	$\text{MgHPO}_4 \cdot 3\text{H}_2\text{O}$	—	—	—	—	+
Tricalcium phosphate	$\beta - \text{Ca}_3(\text{PO}_4)_2$	—	—	—	—	+
Dicalcium phosphate anhydrous	$\text{CaHPO}_4$	—	—	—	—	+

prepared at varying  $\text{Ca}^{2+}:\text{NH}_4^+$  molar ratios of (0, 0.1, 0.2 0.3, and 0.4): 1. As per the procedures explicated in Sect. 0, the mixed solution was treated.

## Analysis

The pH of the solution was monitored using a Horiba F-71 pH meter. Total ammonium was measured using the colorimetric method according to the standard methods of the American Public Health Association (Jenkins 1982). The  $\text{Ca}^{2+}$  concentration was also measured using a LAQUA-twin  $\text{Ca}^{2+}$  meter (HORIBA Advanced Techno Co., Ltd.).

Struvite is the target component, and the percentage of the precipitated struvite out of total solid precipitate is defined herein as the product purity. The purity was assessed in accordance with the methodology suggested by Li et al. (2016). Most of the common struvite mineral impurities do not contain ammonia or ammonium. For a detailed explanation, potential precipitates are summarized in Table 3, along with the corresponding literatures.

For purity quantification, it was assumed that each mole of ammonium stands for one mole of struvite. For assessing the struvite purity, a certain quantity of the solid precipitate (oven dried) was dissolved into 1% concentrated nitric acid, followed by the determination of the  $\text{NH}_4^+$  concentration. The colorimetric method was used for the above measurement. Eventually, the struvite purity was computed using Eq. 4 (Li et al. 2016).

$$\text{Purity (\%)} = \frac{n_{\text{N}} * M_{\text{s}}}{m_{\text{p}}} * 100 \quad (4)$$



where  $n_N$ ,  $M_s$ , and  $m_p$  represent the moles of ammonium, the molar mass of struvite (245.4 g/mol), and the mass of the dissolved precipitates, respectively.

## Results and discussion

### Rinsing of ammonium ions from sample

#### Effect of pH and concentration

It was assumed that the pH of the rinse solution would significantly influence the ammonium removal rate. To determine optimum pH for the rinse solution, the ammonium removal test was conducted with distilled water (D.W) at pH values of 5, 7, and 9. As shown in Fig. 2a, the removal efficiency of ammonium slightly decreased from pH 5 to 7 and then increased in pH 9. This result may be explained by the fact that the concentration of  $H^+$  and ionic strength increase as pH decreases. Thus, high ionic strength caused more desorption of ammonium ions from the negatively charge surface compared to pH 7 (Huang et al. 2015; Li et al. 2012). The further increase in pH to 9 slightly increased the efficiency, which might be attributed to the expected transformation of ammonium ions into aqueous un-ionized  $NH_3$  (Abukhadra et al. 2020; Shaban et al. 2017). It is noteworthy that more increase in pH values causes volatilized ammonia (Duan et al. 2013).

In the next step, the impact of the selected concentrations (0, 0.05, 0.1, and 0.2 mol/L) of NaCl was assessed on ammonium removal. The results, as shown in Fig. 2b, indicated that the ammonium rinsing efficiency increased with increasing the concentration. As the rinse solution concentration increased from 0 to 0.2 mol/L NaCl, the ammonium removal efficiency correspondingly increased from 74.77 to 78.95%, 83.06%, and 89.95%. Mainly, increasing the rinsing solution concentration induces a high removal efficiency due to high ionic strength. Several reports have shown that ammonium removal from adsorbents (e.g., zeolite) increases with increasing the regeneration solution concentration (Lee et al. 2019b; Zhang et al. 2017b). To determine the optimum concentration of the rinse solution, further experimental investigations on different soils are needed.

#### Effect of $Na^+$ , $Ca^{2+}$ , $Mg^{2+}$ , and D.W.

To investigate the effect of single cation ( $Na^+$ ,  $Ca^{2+}$ , and  $Mg^{2+}$ ) on the rinsing of ammonium ions, the sand columns were rinsed with the 0.2 mol/L concentration of the NaCl,  $CaCl_2$ , and  $MgCl_2$  solutions and also with D.W. The pH of all the solutions was adjusted to  $9 \pm 0.1$ . In Fig. 2c, an overview of the ammonium removal efficiency is shown for each solution as a function of the rinsing volume. Rinsing

with the magnesium solution followed by calcium, sodium, and D.W exhibited the highest ability to remove ammonium, resulting in the ammonium removal of 98.54%, 96.47%, 89.95%, and 74.77%, respectively. Furthermore, the ammonium removal efficiency of  $MgCl_2$ ,  $CaCl_2$ , NaCl, and D.W was observed to rapidly reach 74.65%, 73.25%, 69.16%, and 73.14%, respectively, after rinsing with a 30 mL solution (~1.5 PV). However, a further rinsing with D.W resulted in only the ammonium removal of less than 1.5%. These results suggested that D.W. was ineffective in ammonium removal. Interestingly, the  $MgCl_2$  solution exhibited a better response than all the other solutions. A probable explanation for the apparent effectiveness of ions in ammonium removal can be the assumption that they are specifically affected by ionic strength and charge density. For instance,  $Mg^{2+}$  charge density ( $120 \text{ Cmm}^{-3}$ ) is higher than  $Ca^{2+}$  and  $Na^+$  with charge densities of 52 and  $24 \text{ Cmm}^{-3}$ , respectively. It is worth noting that rinsing the cemented samples by the solutions only caused slight changes in calcite contents (Lee et al. 2019a).

In Fig. 3, the ammonium concentration is plotted as a function of the rinsing solution volume. This provides essential information regarding the volume of the rinsing solution necessary to achieve an acceptable level of the ammonium concentration in effluents or soil. In fact, the required volume is expected to rely on soil physical (e.g., specific surface, porosity, and permeability) and chemical (e.g., cation exchange capacity) properties as well as the rinsing solution characteristics, and it should be derived based on specifications of each site.

### Recovery of ammonium ions by struvite

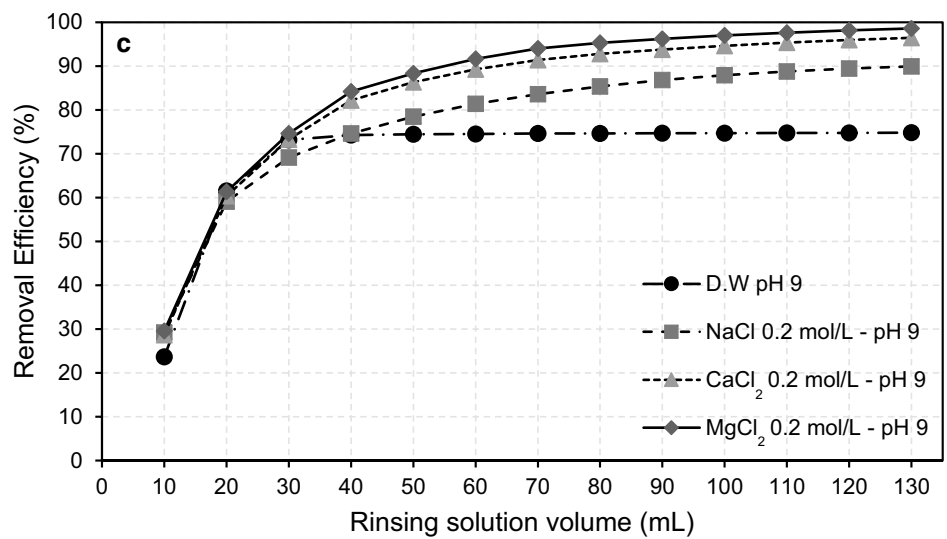
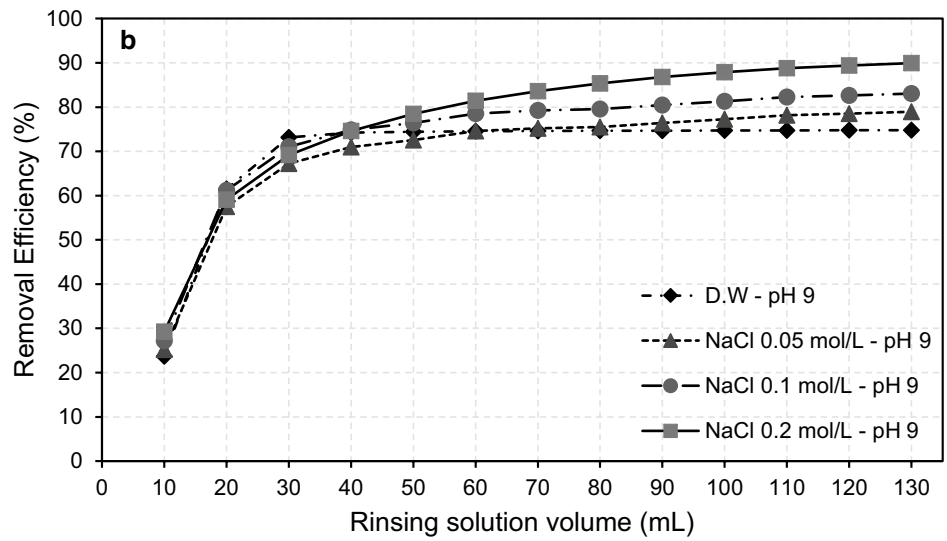
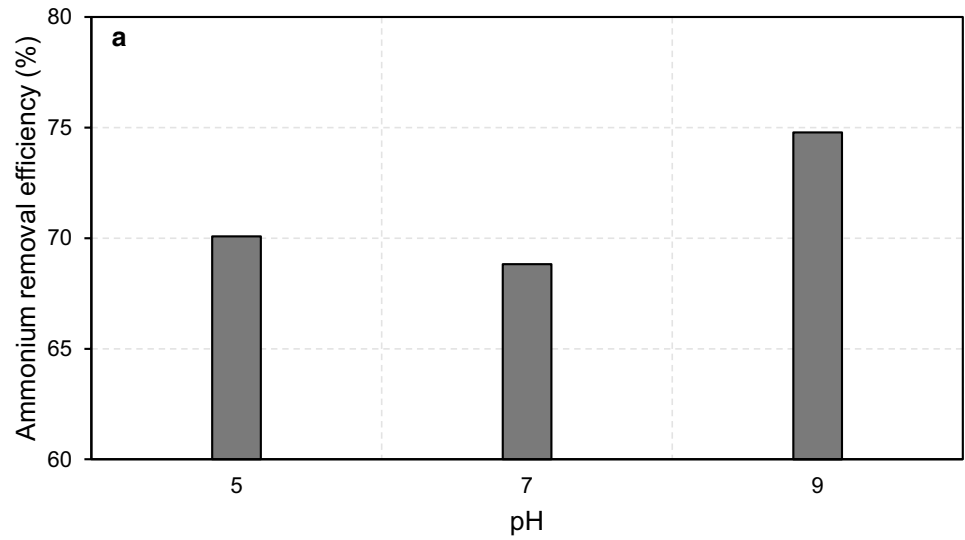
#### Effect of $Na^+$ and $K^+$ in struvite precipitation tests in different pH

The phosphate source and pH were considered as factors affecting the ammonium removal efficiency by struvite precipitation. To obtain the maximum recovery rate of ammonium, struvite precipitation experiments using  $Na_2HPO_4$  and  $K_2HPO_4$  as the phosphate source were conducted at pH 6.5–10.5 with the  $Mg^{2+}:NH_4^+:PO_4^{3-}$  molar ratio of 1:1:1. The results of the experiments are shown in Fig. 4. It can be clearly observed that a higher ammonium removal rate was achieved at all pH levels using  $Na_2HPO_4$ , as compared with  $K_2HPO_4$ .

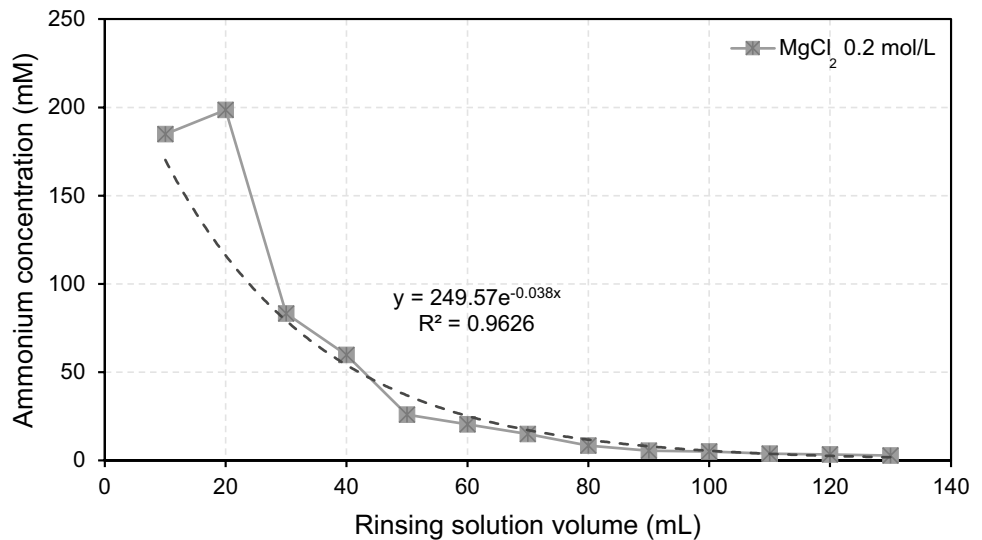
Struvite formation has been previously reported (Abbona et al. 1982). As struvite, K-struvite ( $MgKPO_4 \cdot 6H_2O$ ), and Na-struvite ( $MgNaPO_4 \cdot 6H_2O$ ) are isomorphous (the general chemical formula is  $MgXPO_4 \cdot nH_2O$ , where X can be the following cations:  $K^+$ ,  $Na^+$ ,  $Rb^+$ ,  $Cs^+$ , or  $NH_4^+$  and n ranges between 6 and 8) (Banks et al. 1975), their crystallization mechanisms are considered to be analogous (Huang et al. 2019a). Therefore, in light of the experimental conditions



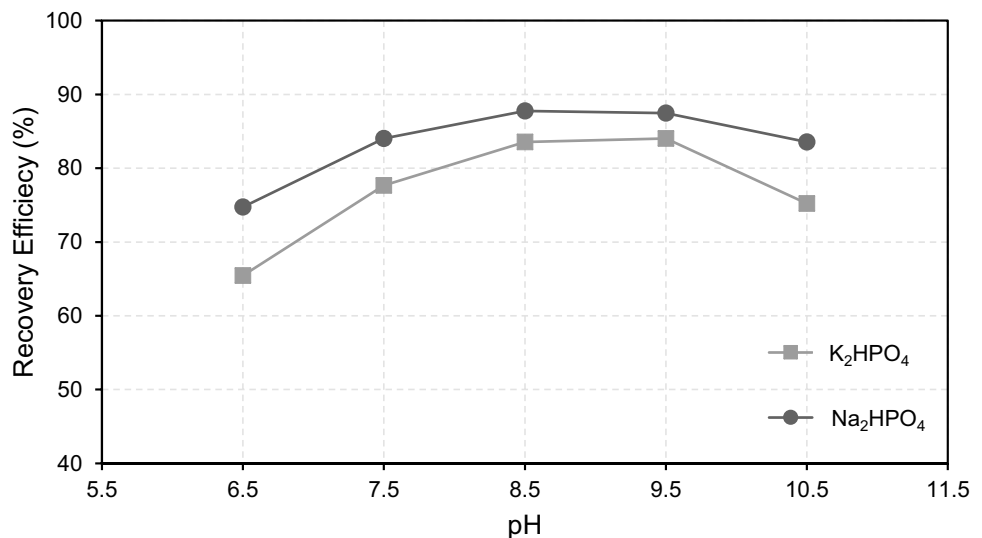
**Fig. 2** The  $\text{NH}_4^+$  removal efficiency from the soil columns: **a** the effect of pH, **b** the effect of the rinsing solution concentration, and **c** the effects of NaCl,  $\text{CaCl}_2$ ,  $\text{MgCl}_2$ , and D.W.



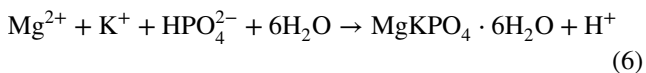
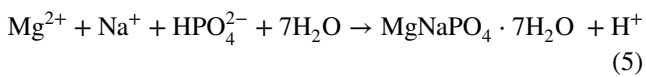
**Fig. 3** Ammonium concentrations in the outlet during ammonium removal by the 0.2 mol/L  $\text{MgCl}_2$  solution



**Fig. 4** The recovery efficiency of  $\text{NH}_4^+$  during struvite precipitation



and based on the literature review, it was possible to infer that the following reactions may occur:



A possible explanation for this might be that K-struvite precipitated more than Na-struvite. Thus, it is concluded that  $\text{K}^+$  has much more potential than  $\text{Na}^+$  for phosphate and magnesium. In accordance with the present results, previous studies have demonstrated that the solidity of  $\text{MgXPO}_4 \cdot n\text{H}_2\text{O}$  is highly associated with the ionic radius of X ions. A high ionic radius results in a high solidity of the compound (Banks et al. 1975; Gao et al. 2018). As  $\text{K}^+$  has a higher ionic radius than  $\text{Na}^+$ , the crystal stability of

$\text{MgKPO}_4$  is higher than that of  $\text{MgNaPO}_4$  (Huang et al. 2019b). The collected precipitates were analyzed with XRD to confirm the composition of struvite. The XRD analysis of the samples (Fig. S2, refer to the supplementary file) revealed that the patterns matched well with the peaks for pure struvite.

In Fig. 4, it is shown that the highest ammonium removal rate (around 88% with using  $\text{Na}_2\text{HPO}_4$  and around 83% with using  $\text{K}_2\text{HPO}_4$ ) occurred at pH 8.5–9.5. These results concur with previous findings. According to Table 4, various optimum pH ranges were reported for struvite crystallization principally based on ammonium removal.

By reviewing the literature, the following explanations were found on the relationship between struvite precipitation and optimum pH:





**Table 4** Optimal pH reported in the literature for struvite precipitation

Reference	Optimal pH	Mg source	Phosphate source	Removal rate
Maekawa et al. (1995)	7.5	MgCl <sub>2</sub>	KH <sub>2</sub> PO <sub>4</sub>	More than 90%
Li et al. (1999)	8.5–9	MgCl <sub>2</sub>		99%
Di Iaconi et al. (2010)	9	MgO	H <sub>3</sub> PO <sub>4</sub>	95%
Altınbaş et al. (2002)	9.2	MgCl <sub>2</sub>	NaH <sub>2</sub> PO <sub>4</sub> · 2H <sub>2</sub> O	68% to 72%
Zhang et al. (2009b)	9.5	MgCl <sub>2</sub>	Na <sub>2</sub> HPO <sub>4</sub>	88%
Huang et al. (2014)	8.5	MgO	H <sub>3</sub> PO <sub>4</sub>	83%

1. As the pH increased, Mg(OH)<sub>2</sub> and Mg<sub>3</sub>(PO<sub>4</sub>)<sub>2</sub> were formed instead of MgNH<sub>4</sub>PO<sub>4</sub>, which can reduce the Mg<sup>2+</sup> concentration to precipitate as MgNH<sub>4</sub>PO<sub>4</sub> (Li et al. 2012; Ryu et al. 2008).
2. According to the information available in the literature, the optimal pH values corresponding to Na-struvite and K-struvite are 12 and 11, respectively (Gao et al. 2018; Huang et al. 2019a, b).
3. As the phase change of ammonia (from ammonium ions to free ammonia) occurs at pH values greater than 8.5, ammonium ions cannot precipitate as struvite, and thus, a low ammonium removal rate can be obtained at a high pH range (Hu et al. 2020; Muhmood et al. 2019).
4. In the pH range lower than optimal, because of H<sup>+</sup> ions, the precipitation of struvite is inhibited (Li et al. 2012; Zhang et al. 2009a).

From the economic perspective, the pH value of 8.5 was taken to preserve chemical reagents.

### Effect of molar ratio

To improve the ammonium removal efficiency, the experiments were conducted at different magnesium, ammonium, and phosphate molar ratios. In Fig. 5, the variations of the ammonium recovery efficiency and the purity of sediments with different Mg<sup>2+</sup>:NH<sub>4</sub><sup>+</sup>:PO<sub>4</sub><sup>3-</sup> molar ratios are demonstrated. Moreover, Fig. 5a shows the effect of changing magnesium molar ratios on the ammonium removal efficiency and the purity of sediments when the molar ratio of Mg<sup>2+</sup>:NH<sub>4</sub><sup>+</sup>:PO<sub>4</sub><sup>3-</sup> was 0.8: 1: 1, 1: 1: 1, 1.2: 1: 1, and 1.4: 1: 1.

As indicated in Fig. 5a, the ammonium removal efficiency and the purity of sediments increased by increasing the Mg<sup>2+</sup> molar ratio from 0.8 to 1.2; however, a further increase in the magnesium molar ratio resulted in an insignificant increase in the ammonium recovery efficiency and the purity of sediments. This indicates that Mg<sup>2+</sup> is a pivotal parameter to optimize ammonium recovery. The result obtained herein is in a good agreement with the results obtained by Huang et al. (2017) and Song et al. (2007), revealing that the struvite formation efficiency increased by increasing the Mg<sup>2+</sup> molar ratio.

Next, the experiments focused on the effect of the phosphate concentration on the ammonium recovery efficiency and the struvite purity. Different molar ratios of phosphate were considered, as in the previous test. The purity of sediments declined from 92.3 to 75.1% with the increase in the phosphate molar ratio from 0.8 to 1.4. However, the removal efficiency reached the maximum value of 85.2% at the 1:1:1 molar ratio and then gradually decreased with a further increase in the phosphate molar ratio. Moreover, the results showed that with an increase in the phosphate concentration more than the stoichiometric molar ratio of struvite (1:1:1), the purity of sediments and the ammonium recovery ratio decreased (Fig. 5b). Consequently, adding a higher excess phosphate is not advantageous, in particular when the struvite purity is considered.

As observed in Fig. 5c, with increasing the Mg<sup>2+</sup>:NH<sub>4</sub><sup>+</sup>:PO<sub>4</sub><sup>3-</sup> molar ratio from 0.8:1:0.8 to 1.4:1:1.4, the purity of sediments decreased from 98.1 to 73.4%. Although the ammonium removal efficiency swiftly increased with the increase in the Mg<sup>2+</sup>:NH<sub>4</sub><sup>+</sup>:PO<sub>4</sub><sup>3-</sup> molar ratio, the low purity of sediments was also obtained. It was inferred that the other magnesium phosphate component precipitated (MgPO<sub>4</sub>·3H<sub>2</sub>O, Mg<sub>3</sub>(PO<sub>4</sub>)<sub>2</sub>·8H<sub>2</sub>O, or Mg<sub>3</sub>(PO<sub>4</sub>)<sub>2</sub>·22H<sub>2</sub>O), apart from struvite (Desmidt et al. 2013; Li et al. 2016).

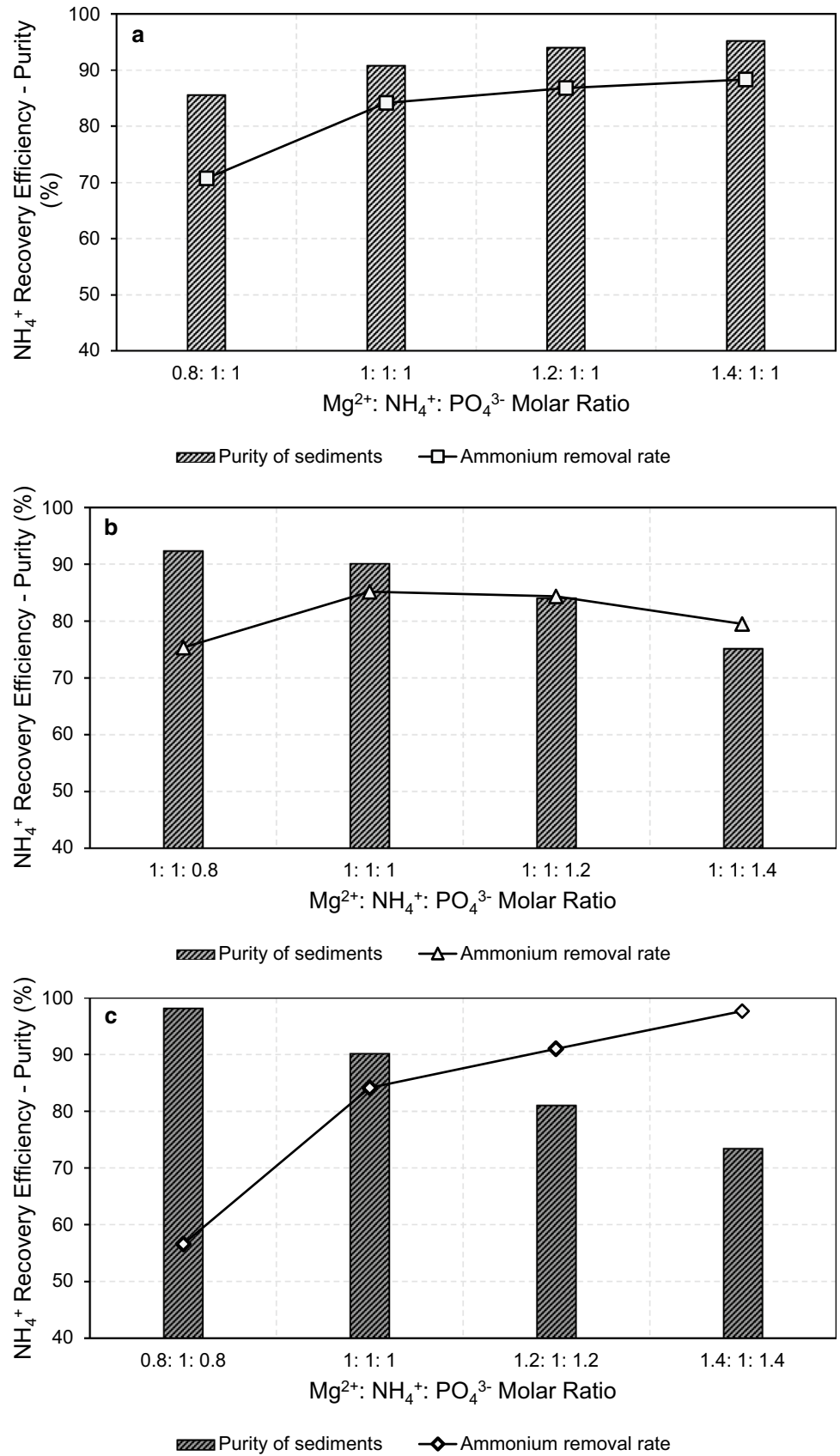
Thus, considering both ammonium removal and the purity of sediments, the molar ratio of Mg<sup>2+</sup>:NH<sub>4</sub><sup>+</sup>:PO<sub>4</sub><sup>3-</sup> at 1.2:1:1 was effectively chosen to be the optimal ratio for ammonium recovery. However, this molar ratio is optimal from both economic and environmental points of view, as overdosing of phosphate and/or magnesium would possibly generate high concentrations of ions at the effluent. In addition, this molar ratio could eliminate the leaching of phosphate at high concentrations in the effluent.

### Effect of calcium ions in SME

For the effective and economic recovery of ammonium from SME, the quality of struvite is highly significant. Calcium is a common component in the effluent of the MICP process. To investigate the effect of calcium ions on ammonium recovery and the struvite purity, a series of batch experiments with the following solution conditions were designed: the pH value was 8.5, the Mg<sup>2+</sup>:NH<sub>4</sub><sup>+</sup>:PO<sub>4</sub><sup>3-</sup>

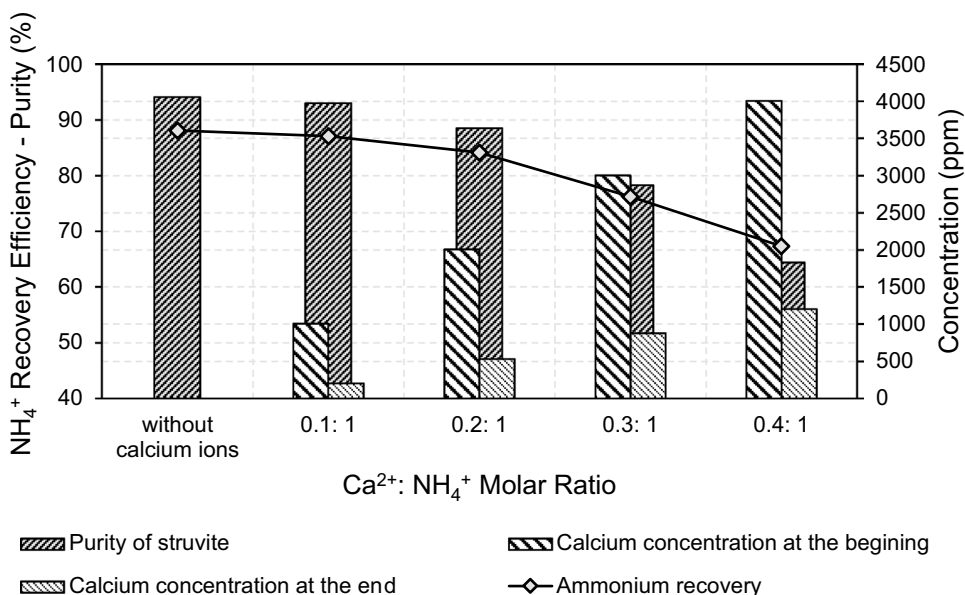


**Fig. 5** The variations of the ammonium recovery efficiency and the struvite purity with different  $Mg^{2+}:NH_4^+:PO_4^{3-}$  molar ratios



molar ratio was 1.2:1:1, and  $\text{Ca}^{2+}:\text{NH}_4^+$  molar ratios were 0, 0.1, 0.2, 0.3, and 0.4. According to the experimental data shown in Fig. 6, it was found that ammonium recovery and the struvite purity relied heavily on  $\text{Ca}^{2+}:\text{NH}_4^+$  molar ratios in the initial solution. These results are consistent with those reported earlier (Hu et al. 2020; Yan and Shih 2016). Liu and Wang (2019) reported that calcium ions highly competed with  $\text{MgNH}_4\text{PO}_4$  for phosphate ions and caused to form hydroxyapatite ( $\text{Ca}_5(\text{PO}_4)_3(\text{OH})$ ) and calcium phosphate ( $\text{Ca}_3(\text{PO}_4)_2$ ), thereby reducing ammonium recovery and the struvite purity. As shown in Fig. 6, it can be concluded that the effect of calcium ions on ammonium recovery was negligible at low  $\text{Ca}^{2+}:\text{NH}_4^+$  molar concentrations (i.e., 0.1:1 and 0.2:1) without significantly affecting the product quality.

**Fig. 6** The variations of the ammonium recovery efficiency, the calcium concentration, and the struvite purity with different  $\text{Ca}^{2+}:\text{NH}_4^+$  molar ratios



As shown in Table 5, there were some pretreatment methods employed to reduce the effect of  $\text{Ca}^{2+}$  during ammonium recovery by struvite crystallization. However, further experimental investigations are needed to estimate the feasibility and practicality of these pretreatment methods, especially from the environmental and economic perspectives.

**Application perspective**

As the MICP effluent is absent in  $\text{Mg}^{2+}$  and phosphate ions, extra chemicals need to be added, leading to high treatment cost. This remains as a major constraint of the struvite process. Few studies examined the cost reduction of struvite precipitation through the use of possible alternative materials.

**Table 5** The overview of different  $\text{Ca}^{2+}$  pretreatment methods used in struvite precipitation

	Chelating	Precipitate $\text{Ca}^{2+}$	Precipitate $\text{Ca}^{2+}$	Precipitate $\text{Ca}^{2+}$
Principle	Chelating agents react with calcium ions to form soluble [Ca-EDTA or Ca-oxalate] complexes with more phosphates available	Precipitate $\text{Ca}^{2+}$ as hydroxide	Precipitate $\text{Ca}^{2+}$ as $\text{CaCO}_3$	Precipitate $\text{Ca}^{2+}$ as $\text{CaCO}_3$
Material used	Ethylenediaminetetraacetic acid (EDTA) Nitrilotriacetic acid (NTA)	—	$\text{Na}_2\text{CO}_3$	Imported $\text{CO}_2$ gas
Remarks	- The process needs to be optimized, because excess chelates might bind to $\text{Mg}^{2+}$ - Environmental concerns - High cost	- Higher dose of phosphate is essential - Cost optimization	- The quantity of $\text{CO}_3^{2-}$ needs to be optimized as toward minimal $\text{Mg}^{2+}$ loss rate - High cost	- High pH - High $\text{Mg}^{2+}$ loss rate
Reference	(Sabbag et al. 2015; Zhang et al. 2010)	(Muster et al. 2013)	(Hu et al. 2020; Wu et al. 2018)	(Hu et al. 2020; Zhang et al. 2017a)

As documented, Bittern (Etter et al. 2011; Lee et al. 2003; Li et al. 2019) and sea water (Matsumiya et al. 2000; Shin and Lee 1998) are two  $Mg^{2+}$  sources with the  $Mg^{2+}$  concentration of 9220–32000 mg/L and 1300 mg/L, respectively. Moreover, the potential of struvite precipitation from various waste sources of industrial, farm, and municipal origin has been established at laboratory scale (Kataki et al. 2016). However, more studies should focus on eliminating economic challenges through the use of possible alternatives, particularly, the use of industrial waste sources for ammonium recovery.

Considering investigations performed to date, only few researchers (Keykha et al. 2018; Putra et al. 2017) have focused on removing ammonium ions produced in MICP and enzyme-induced carbonate precipitation (EICP) using zeolite. Zeolite with a negative charge was able to adsorb  $NH_4^+$  from the cementation solution to the desired level. However, the method proposed herein could recover ammonium ions in the form of a valuable fertilizer.

## Conclusion

The by-production of ammonium ions during biotreatment remains a massive hurdle, limiting the real-field applications and commercialization process of MICP. This study proposed a new post-treatment method, called a two-stage treatment process, for ammonium by-products. The results indicated that the proposed strategy was a simple, convenient, and reliable approach for ammonium management in geotechnical applications based on rinsing and precipitation instead of the complex nitrification process and then recovered materials can be used. We observed that the ionic strength of the rinse solution had a significant effect during the rinsing step. We realized that using high ionic strength was more suitable and the pH of the rinse solution alone showed a small effect. Secondly, the proposed strategy can recover as much as 86.8% of ammonium ions in the effluent as high purity struvite by adding  $Na_2HPO_4$  and  $MgCl_2$  at pH 8.5 with a  $Mg^{2+}:NH_4^+:PO_4^{3-}$  molar ratio of 1.2:1:1. Thus, the method can manage ammonium by-products by the recovery-centered approach instead of the remove-centered approach, which is vital in making the process profitable and sustainable.

The limitations of the two-stage treatment process are as follows. We did not examine responses of different types of soil as they have differences in composition as well as in physical and chemical properties. The effectiveness of this method was unclear in the meter-scale distance treatment. We also needed to add extra chemicals when ammonium precipitated as struvite, which could increase treatment cost. Therefore, further studies should focus on these aspects and enable the practical use of this method.

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