



# Removal of ammonia from landfill leachate by struvite precipitation with the use of low-cost phosphate and magnesium sources



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## ABSTRACT

This paper presents a study concerning ammonia removal from landfill leachate by struvite precipitation with the use of waste phosphoric acid as the phosphate source. The results indicated that the  $\text{Al}^{3+}$  ions present in the waste phosphoric acid significantly affected the struvite precipitation, and a removal ratio of ammonia close to that of pure phosphate salts could be achieved. Nevertheless, large amounts of NaOH were necessary to neutralize the  $\text{H}^+$  present in the waste phosphoric acid. To overcome this problem, a low-cost magnesium source was proposed to be used as well as an alkali reagent in the struvite precipitation. The ammonia removal ratios were found to be 83%, with a remaining phosphate of 56 mg/L, by dosing the low-cost MgO in the Mg:N:P molar ratio of 3:1:1. An economic analysis showed that using waste phosphoric acid plus the low-cost MgO could save chemical costs by 68% compared with the use of pure chemicals. Post-treatment employment of a biological anaerobic filter process demonstrated that the high concentration of  $\text{Mg}^{2+}$  remaining in the effluent of the struvite precipitation has no inhibitory effect on the performance of the biological treatment.

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## 1. Introduction

Landfill leachate is one of the wastewater types exerting the greatest environmental impact, as it contains high concentrations of organic matter, inorganic ions and ammonia nitrogen ( $\text{NH}_3\text{-N}$ ). If this is not properly collected and treated, the discharge of landfill leachate into the environment may cause serious pollution to the groundwater aquifers as well as to the adjacent surface waters (Shultz and Kjeldsen, 1986). Thus, the rigorous removal of organic material and ammonia nitrogen from the leachate is the normal prerequisite before the leachates are discharged into the natural waters.

Various physical/chemical and biological treatment methods have been applied to treat the landfill leachate because of its complex composition and high contaminant strength (Chou et al., 2013; Kulikowska and Bernat, 2013; Niu et al., 2013; Singh et al., 2012). Among these methods, the biological processes have undoubtedly been considered as the best cost-effective and reusable ones. However, they were not found to be satisfactory in the performance of a conventional activated sludge on treating the landfill

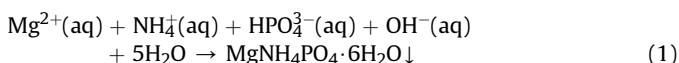
leachate, because of the high concentration of ammonia nitrogen and the lack of sufficient electron donors in the leachate (Anglada et al., 2011; Dogen et al., 2001). For example, Li and Zhao (1998) confirmed the chemical oxygen demand (COD) removal by a conventional activated sludge process which declined from 95 to 79%, when the  $\text{NH}_3\text{-N}$  concentration in the wastewater increased from 50 to 800 mg/L. Although the ammonia nitrogen removal greater than 90% was reported in the recently published literature, the total nitrogen removal efficiency was rather low (Yusof et al., 2010; Wei et al., 2012; Wang et al., 2013a). Therefore, discharging this effluent may still cause a serious pollution problem, such as eutrophication, to the receiving waters. A feasible solution to the resulting problem is the use of a physical–chemical process for the removal of ammonia nitrogen from landfill leachate prior to the employment of biological treatment methods.

Among the alternative processes, struvite (magnesium ammonium phosphate, MAP,  $\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$ ) precipitation has been proven to be a promising method for the removal of ammonia nitrogen because of the high reaction rate and removal ratio (Kochanya and Lipczynska-Kochanyb, 2009). Struvite is a white insoluble crystalline compound and can occur naturally, when the combined concentrations of  $\text{Mg}^{2+}$ ,  $\text{NH}_4^+$ ,  $\text{PO}_4^{3-}$  in solution are greater than their solubility limit (23 mg per 100 mL  $\text{H}_2\text{O}$ ) (Li and

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Zhao, 2003; Wang et al., 2013,b). The reaction equation is given below:



In general, landfill leachate lacks the  $\text{Mg}^{2+}$  and  $\text{PO}_4^{3-}$  used for the struvite precipitation. To achieve effective ammonia removal, large amounts of magnesium and phosphate salts need to be added in the precipitation. This leads to a high treatment cost, which deters the widespread application of the struvite process. At present, one of the solution proposals for this problem is the use of low-cost materials containing magnesium in struvite precipitation, such as MgO-containing byproducts (Chimenos et al., 2003; Quintana et al., 2005, 2008), magnesite pyrolyzate (Chen et al., 2009; Huang et al., 2010), magnesite mineral (Gunay et al., 2008), bittern (Lee et al., 2003; Ye et al., 2011), seawater (Crutchik and Garrido, 2011; Kumashiro et al., 2001) and seawater NF concentrate (Telzhensky et al., 2011; Lahav et al., 2013). The findings in these published papers emphasized that using the low-cost magnesium sources in the struvite precipitation could achieve high ammonia removal efficiency, which may be close to that achieved by using pure magnesium reagents. For example, Gunay et al. (2008) reported that 90.9% of the ammonia in the leachate was removed using magnesite instead of  $\text{MgCl}_2$ , with a resultant drop of 18% in the operation costs. Huang et al. (2010) found that ammonia removal of approximately 90% could be achieved using the pyrolyzate of magnesite as the magnesium source, saving 34% of the operation costs compared with that of using the pure magnesium salt. Lahav et al. (2013) used nanofiltration concentrate as the source of  $\text{Mg}^{2+}$  for struvite precipitation, and a phosphate recovery of 90% was achieved with a corresponding 25% decrease in the cost. Although there is an obvious decrease in the cost of the ammonia removal by using low-cost materials containing magnesium in the struvite precipitation, the contribution is still limited. As seen in Eq. (1), the total chemical cost of ammonia removal from landfill leachate by struvite precipitation can be observed as being due to the magnesium salt, phosphate salt and an alkali reagent. In fact, the phosphate salt and alkali reagent may account for a great part of the cost. Nevertheless, this problem did not draw significant attention from the researchers. To the best of our knowledge, there has been no study available which has focused on the chemical, economical and environmental feasibility of using waste phosphoric acid (WPA) to provide phosphate ions to the struvite precipitation. A large quantity of waste phosphoric acid generated in the chemical production process and its low cost makes it an important phosphate source.

Therefore, in this study, experiments have been conducted to remove the ammonia nitrogen from landfill leachate using waste phosphoric acid. The objectives of this study were to investigate the feasibility of using waste phosphoric acid as the phosphate source in the struvite precipitation, as well as to examine the performance of the biological anaerobic filter (BANF) on landfill leachate, pretreated with struvite precipitation. The specific study contents are summarized as follows. First, the effect of the  $\text{Al}^{3+}$  present in the waste phosphoric acid on the struvite precipitation was clarified. Second, the performance of the waste phosphoric acid as the phosphate source was performed under different conditions. Third, the waste phosphoric acid combined with the low-cost materials containing magnesium was utilized in the struvite precipitation to further reduce the cost of struvite precipitation. The solid precipitation thus obtained was studied under a Scanning Electron Microscope (SEM) and X-ray Diffraction (XRD). Finally, an economic analysis of using waste phosphoric acid was conducted and BANF treatment of the landfill leachate pretreated with struvite precipitation was performed.

## 2. Materials and methods

### 2.1. Landfill leachate

In this study, the landfill leachate used was collected from a municipal sanitation landfill site in Beijing, China. It was stored in a cool room before use. The characteristics of the landfill leachate are listed in Table 1.

### 2.2. Materials used in the experiments

The waste phosphoric acid used as the low-cost phosphate source was collected from a chemical plant in Wuxi, China. Its main composition in molar concentration was as follows:  $\text{H}_3\text{PO}_4$  4.5 M,  $\text{H}_2\text{SO}_4$  1.7 M,  $\text{Al}^{3+}$  0.4 M and  $\text{Na}^+$  0.3 M. Besides, to further reduce the struvite precipitation cost, low-cost MgO was used in the experiments. This was obtained from the calcination of magnesite, with a 53% Mg content (Huang et al., 2010). In addition, analytical grade  $\text{AlCl}_3$ ,  $\text{Ca}(\text{OH})_2$ , NaOH,  $\text{Na}_2\text{HPO}_4 \cdot 12\text{H}_2\text{O}$  and  $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$  were used in this study.

### 2.3. The influence of $\text{Al}^{3+}$ on struvite precipitation

Due to the high content of  $\text{Al}^{3+}$  present in the waste phosphoric acid, determining the influence of  $\text{Al}^{3+}$  on the struvite precipitation of landfill leachate, involved the use of pure  $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$  and  $\text{Na}_2\text{HPO}_4 \cdot 12\text{H}_2\text{O}$  as the magnesium and phosphate sources in the experiments, respectively. The experimental procedures included the following steps: First, 250 mL landfill leachate was fed into an airtight 500 mL jar. Subsequently,  $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$  and  $\text{Na}_2\text{HPO}_4 \cdot 12\text{H}_2\text{O}$  were added to the wastewater at the Mg:N:P stoichiometric molar ratio of 1:1:1, respectively, followed by the addition of  $\text{AlCl}_3$  solution (1.0 M) into the leachate (controlling the  $\text{Al}^{3+}$  concentration of the landfill leachate in the range of 0–400 mg/L). Finally, the landfill leachate was stirred using a magnetic stirrer for 30 min at the desired pH (8–9.5). The pH of the solution was adjusted by the addition of 0.1 M NaOH. After the reaction solution was precipitated for 10 min, the supernatant was filtered through a 0.45- $\mu\text{m}$  membrane filter for component analysis.

### 2.4. Experiments for the use of waste phosphoric acid

Using waste phosphoric acid as a phosphate source in struvite precipitation experiments were conducted at different Mg:N molar ratios (in the range of 0.9–1.3, N:P = 1:1). In these experiments,  $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$  was used as the magnesium source, and the pH was

**Table 1**  
Composition of the landfill leachate used in this study.

| Parameter                                      | Value         |
|--|---------------|
| pH   | 7.9 ± 0.2     |
| COD (mg/L)                                     | 4295 ± 520    |
| BOD (mg/L)                                     | 2086 ± 358    |
| $\text{NH}_3\text{-N}$ (mg/L)                  | 1750 ± 89     |
| TN (mg/L)                                      | 1980 ± 108    |
| Alkalinity ( $\text{Na}_2\text{CO}_3$ ) (mg/L) | 10,187 ± 1898 |
| TP (mg/L)                                      | 12.5 ± 1.3    |
| SS (mg/L)                                      | 2350 ± 180    |
| K (mg/L)                                       | 2100 ± 145    |
| Ca (mg/L)                                      | 125 ± 20      |
| Na (mg/L)                                      | 2990 ± 450    |
| Mg (mg/L)                                      | 387 ± 70      |
| Fe (mg/L)                                      | 5.9 ± 0.9     |
| Al (mg/L)                                      | 4.1 ± 0.6     |
| Cu (mg/L)                                      | 0.8 ± 0.4     |
| Zn (mg/L)                                      | 0.3 ± 0.1     |

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