



# Enhancing recovery of magnesium as struvite from landfill leachate by pretreatment of calcium with simultaneous reduction of liquid volume via forward osmosis

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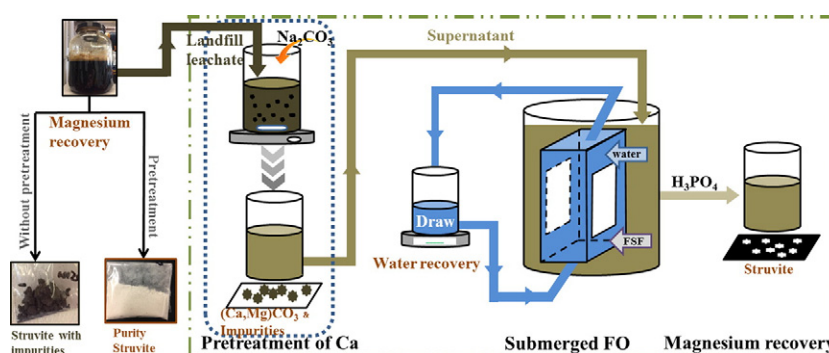
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## HIGHLIGHTS

- Pretreatment to remove calcium improves the quality of the recovered struvite.
- The  $\text{Ca}^{2+}:\text{CO}_3^{2-}$  molar ratio of 1:1.4 achieves low  $\text{Mg}^{2+}$  loss and high  $\text{Ca}^{2+}$  removal.
- Calcium removal decreases membrane fouling and thus benefits water recovery.
- The calcium pretreatment - FO - struvite recovery is the optimal configuration.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Landfill leachate contains substances that can be potentially recovered as valuable resources. In this study, magnesium in a landfill leachate was recovered as struvite with calcium pretreatment; meanwhile, the leachate volume was reduced by using a submerged forward osmosis (FO) process, thereby enabling significant reduction of further treatment footprint and cost. Without pretreatment, calcium exhibited strong competition for phosphate with magnesium. The pretreatment with a  $\text{Ca}^{2+}:\text{CO}_3^{2-}$  molar ratio of 1:1.4 achieved a relatively low loss rate of  $\text{Mg}^{2+}$  ( $24.1 \pm 2.0\%$ ) and high  $\text{Ca}^{2+}$  removal efficiency ( $89.5 \pm 1.7\%$ ). During struvite recovery,  $98.6 \pm 0.1\%$  of magnesium could be recovered with a significantly lower residual  $\text{PO}_4^{3-}\text{-P}$  concentration ( $<25 \text{ mg L}^{-1}$ ) under the condition of  $(\text{Mg} + \text{Ca}_{\text{residual}}):\text{P}$  molar ratio of 1:1.5 and pH 9.5. The obtained struvite had a similar crystal structure and composition (19.3% Mg and 29.8% P) to that of standard struvite. The FO process successfully recovered water from the leachate and reduced its volume by 37%. The configuration of calcium pretreatment - FO - struvite recovery was found to be the optimal arrangement in terms of FO performance. These results have demonstrated the feasibility of magnesium recovery from landfill leachate and the importance of the calcium pretreatment, and will encourage further efforts to assess the value and purity of struvite for commercial use and to develop new methods for resource recovery from leachate.

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## 1. Introduction

Landfill leachate is a liquid waste by-product generated from municipal solid waste (MSW) landfill via decomposition of landfill materials

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and drainage of local precipitation (Yan et al., 2015). It is a hazardous waste due to its major pollutants such as dissolved organic matter, xenobiotic organic compounds, and heavy metals (Tigini et al., 2014; A et al., 2017). Direct discharge or unexpected leaking of landfill leachate can lead to contamination of both groundwater and surface water, causing environmental and health concerns (Butt et al., 2017); thus, proper treatment of landfill leachate would be required (Renou et al., 2008). Biological treatment offers a cost-effective and reliable approach for efficient removal of organic contents but the refractory compounds such as humic and fulvic acids are not easily biodegradable (Kulikowska and Klimiuk, 2008). Moreover, humic and fulvic substances could inhibit the biodegradation of simple organics such as acetate due to its toxicity to microbes and thus decrease biological treatment efficiency (Zhao et al., 2017). It has been reported that the presence of heavy metals, such as Cd(II) and Ni(II), was harmful to microbes in a biological treatment process and could depress nitrification and biological phosphorus removal (Chen et al., 2014; Sun et al., 2017). Therefore, the removal of metal ions from landfill leachate is also a major challenge because of their high toxicity and non-biodegradability feature. Physicochemical methods/technologies, e.g. coagulation precipitation (Li et al., 2016; Liu et al., 2012), activated carbon adsorption (Foo and Hameed, 2009; Xu et al., 2012), and advanced oxidation (Cortez et al., 2011; Kurniawan et al., 2006), have been extensively investigated. Nonetheless, notable drawbacks are observed, such as excess chemical sludge production (Al-Hamadani et al., 2011), limited pollutant removal (e.g., molecular weight <1000), and elevated operation costs (Wiszniewski et al., 2006).

Sustainable leachate treatment needs to lower the treatment cost and urges treatment transformation from removal-centered approaches to recovery-centered processes. In fact, many pollutants in landfill leachate can be reclaimed as reusable resources (Di Iaconi et al., 2011; Xu et al., 2012). For instance, organic compounds can be converted to biogas, which is then used as an energy source (Chan et al., 2002). Struvite (MAP as  $\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$ , or MKP as  $\text{MgKPO}_4 \cdot 6\text{H}_2\text{O}$  (Ikematsu et al., 2007)), a valuable slow release fertilizer (Antonini et al., 2012), can also be precipitated from landfill leachate to recover both ammonia (Xiu-Fen et al., 2011) and magnesium by adding an external phosphorus source (e.g.,  $\text{Na}_2\text{HPO}_4$  (Kabdaslı et al., 2008),  $\text{H}_3\text{PO}_4$  (Kochany and Lipczynska-Kochany, 2009), or  $\text{KH}_2\text{PO}_4$  (Kim et al., 2007)) and/or a magnesium source (e.g.,  $\text{MgCl}_2$  (Kabdaslı et al., 2008),  $\text{MgSO}_4$  (Li and Zhao, 2003),  $\text{MgO}$  (Krähenbühl et al., 2016),  $\text{MgCO}_3$  (Gunay et al., 2008), wood ash (Sakthivel et al., 2012), or sea salt (Huang et al., 2014)). The addition of relatively expensive chemicals and the product's low purity (mainly due to co-precipitation of other compounds such as calcium) (Kabdaslı et al., 2008; Uysal et al., 2010) impedes the broader application of leachate-originated struvite recovery.

In addition to struvite recovery, water recovery can also be achieved with simultaneous reduction of leachate volume. Membrane separation technologies, such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO) and forward osmosis (FO) have been developed to concentrate landfill leachate while recovering fresh water (Iskander et al., 2016; Qin et al., 2016). Among them, FO utilizes a natural osmotic pressure gradient across a piece of semi-permeable membrane to drive water migration from a feed solution (high-water potential) to a draw solution (low-water potential) (Zou and He, 2017). Comparing to pressure-driven processes, FO requires less energy input if proper draw regeneration is applied, and has a low fouling propensity (Zou and He, 2016; Zou et al., 2016). Selection of the draw solute should be carefully evaluated in an FO process treating landfill leachate (the feed). Because of leachate's high conductivity, desirable draw solution must have a high osmotic pressure to render a positive driven force. Currently, 40% of the leachate-related FO studies employed NaCl as a draw solute due to its high solubility, low cost, and relatively high osmotic pressure (Lutchmiah et al., 2014). The final high-quality fresh water could be obtained by isolation of NaCl

solute via low-pressure RO (LPRO), membrane distillation (MD), or electro dialysis (ED) technique with low energy consumption (Chung et al., 2012). Pretreatment of landfill leachate is preferred to avoid potential penetration of pollutants, such as humic acid, low-molecular-weight organic matter, and heavy metals, through FO membrane via forward solute flux (FSF, concurrent ion diffusion with water flux from the feed side to the draw side (Zou et al., 2017)), membrane fouling (both biofouling and inorganic scaling), and contamination of product water (Liu et al., 2013). However, the trade-off between increased ionic concentration via chemical addition for pretreatment (i.e. reduced osmotic pressure gradient) and water recovery capability in subsequent FO should be carefully evaluated.

Our previous study has demonstrated successful water recovery from leachate via FO treatment featuring low fouling and low energy consumption (Iskander et al., 2017). However, nutrient recovery from landfill leachate with simultaneous reduction of its volume using FO technology has not been investigated before. Herein, a submerged FO process linked to struvite precipitation was proposed with a focus on both water and magnesium reclamation. Pretreatment by adding carbonate was employed to enhance struvite purity via chemical precipitation of calcium. To evaluate the trade-off between the increased ionic concentration (i.e. reduced osmotic gradient) due to chemical pretreatment and water recovery capability, as well as resource loss to draw side via FSF effect, three configurations were proposed and investigated. The specific objectives of this study were to (1) investigate the struvite recovery from landfill leachate and the effects of chemical pretreatment on such recovery; (2) examine water recovery performance affected by the pretreatment in the FO system; and (3) evaluate the optimal arrangement of chemical pretreatment, struvite precipitation, and FO water recovery with a special focus on FO performance.

## 2. Materials and methods

### 2.1. Landfill leachate characterization

The landfill leachate used in this study was collected from a MSW landfill site located in Virginia, USA and stored at 4 °C before tests. The characteristics of the raw landfill leachate are listed in Table 1. It had a pH of 6.58 (weak acidic condition) and major ionic components including magnesium ( $722.0 \text{ mg L}^{-1}$ ), calcium ( $3254.7 \text{ mg L}^{-1}$ ), potassium ( $2000.0 \text{ mg L}^{-1}$ ), ammonia nitrogen ( $2753.0 \text{ mg L}^{-1}$ ) and a low content of orthophosphoric phosphorus ( $\text{PO}_4^{3-}\text{-P}$ ,  $3.9 \text{ mg L}^{-1}$ ). The landfill leachate was centrifuged at 10000 rpm and filtered by  $0.45 \mu\text{m}$  filter paper to remove suspended particles before the following experiments.

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

Parameters	Values	Unit
pH	6.58	
Conductivity	35.8	$\text{mS cm}^{-1}$
COD	69,470	$\text{mg L}^{-1}$
$\text{NH}_4^+\text{-N}$	2753.0	$\text{mg L}^{-1}$
Na	4205.0	$\text{mg L}^{-1}$
K	2000.0	$\text{mg L}^{-1}$
Mg	722.0	$\text{mg L}^{-1}$
Ca	3254.7	$\text{mg L}^{-1}$
Cl	5957.0	$\text{mg L}^{-1}$
Fe	882.1	$\text{mg L}^{-1}$
Si	94.2	$\text{mg L}^{-1}$
Zn	7.6	$\text{mg L}^{-1}$
Ni	0.2	$\text{mg L}^{-1}$
Cu	0.03	$\text{mg L}^{-1}$
P	3.9	$\text{mg L}^{-1}$

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