



Short communication

Recovery of organic carbon and phosphorus from wastewater by Fe-enhanced primary sedimentation and sludge fermentation



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ABSTRACT

A new chemically enhanced primary sedimentation (CEPS) and sludge fermentation process are developed for improved nutrient removal, energy saving and resource recovery in municipal wastewater treatment. The FeCl_3 -based CEPS with a dosage of 20 mg-Fe/L can remove 75.6% of organic pollutants and 99.3% of $\text{PO}_4\text{-P}$ on average from wastewater. Under natural fermentation conditions, the CEPS sludge undergoes effective hydrolysis and acidogenesis to produce volatile fatty acids (VFAs) and release phosphate as valuable resources. By using CEPS, around 27% of the organic carbon in wastewater influent can be recovered via sludge fermentation, mainly in the form of VFAs, and about 23% of phosphorus recovered for making vivianite fertilizer. In comparison, both the organic and phosphorus recovery ratios from wastewater are under 10% with conventional primary sedimentation and sludge fermentation. CEPS combined with side-stream sludge fermentation can be readily applied in new treatment plants or in a retrofit of existing treatment systems.

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1. Introduction

Nutrients, including phosphorus (P) and nitrogen (N), in wastewater discharge induce eutrophication and frequent algal blooms in natural waters [1]. Compared to organic degradation in wastewater treatment, nutrient removal is often difficult. Phosphorus removal can be achieved by either the enhanced biological P removal (EBPR) process or by dosing with chemicals to form P-incorporated precipitates, followed by sufficient sludge discharge [2]. Compared to EBPR, the chemical dosing approach is more reliable and effective. Chemicals such as FeCl_3 and alum have been widely used as coagulants in wastewater treatment, especially in a process named chemically enhanced primary sedimentation, or CEPS [3]. Compared to conventional primary sedimentation, CEPS can greatly improve the pollutant removals [4]. Stonecutters Island Sewage Treatment Works in Hong Kong is the largest CEPS treatment plant in the world, dosing FeCl_3 (10–12 mg-Fe/L) to treat wastewater at a flow of 1.7 million m^3/d [5]. About 1000 tons/d of dewatered CEPS sludge is produced, and current disposal of the sludge by landfilling requires land space and threatens the environment. On the other hand, the CEPS sludge contains large amounts of

organics and phosphorus, which are valuable resources that should be recovered, rather than being wasted.

Nitrogen removal in wastewater treatment is also difficult and costly due to the decreased organic carbon/nitrogen ratio in municipal wastewater [6]. To make up the deficit in organic for biological denitrification, an external organic carbon source, such as methanol, is often added to wastewater. The use of methanol increases both the cost and fire risk for the wastewater treatment system [7,8]. Recently, fermentation of waste sludge has drawn much attention, because the obtained soluble organic substances (mainly volatile fatty acids, VFAs) are good alternatives to methanol for denitrification [9,10]. For example, the total nitrogen removal by A^2/O wastewater treatment was improved from 27% without any organic carbon addition to 80% when supplemented with the fermented sludge liquor [10]. Besides, 83.2% of the released $\text{PO}_4\text{-P}$ could be recovered in the form of struvite by adding magnesium hydroxide to form $\text{NH}_4\text{MgPO}_4 \cdot 6\text{H}_2\text{O}$ [9]. As the major component of the excess sludge, the secondary sludge is much less degradable than the primary sludge, since it mainly consists of microbial cells that can be hardly ruptured [11]. Therefore, various pretreatment methods have been adopted on secondary sludge to disintegrate microbial cells and improve the VFAs yield [12], which makes the sludge fermentation costly and complex. Due to the limited amount of primary sludge from simple sedimentation, the amount of VFAs produced from its fermentation is often insufficient to meet the

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denitrification requirement [11]. In this regard, the high organic content in CEPS sludge would provide more VFAs for enhanced nitrogen removal or for other uses.

However, little research has been conducted on CEPS and its sludge fermentation performance. Until now, the fermentation potential of CEPS sludge and related VFAs production, especially in the presence of chemical coagulants, have remained unknown. The effectiveness of CEPS for phosphorus removal and recovery also has not been reported. In this study, FeCl_3 was used as the coagulant for CEPS to concentrate organic pollutants and phosphorus into the sludge. The CEPS sludge was then processed by simple fermentation to convert the pollutants to valuable resources, including VFAs for denitrification or other uses and phosphate for fertilizer. The effects of pretreatment of the sludge by ultrasonication or alkali treatment on organic hydrolysis and VFAs production were also tested.

2. Materials and methods

2.1. Iron-enhanced primary sedimentation and sludge fermentation experiments

Raw wastewater was collected from Stanley Sewage Treatment Works in Hong Kong. The wastewater was typical of domestic sewage, with the following average values for the influent parameters during the experimental period: pH 7.02 ± 0.02 , total chemical oxygen demand (TCOD) 451 ± 18 mg/L, total organic carbon (TOC) 147.3 ± 2.4 mg/L, suspended solids (SS) 368 ± 12 mg/L, total phosphorus (TP) 4.85 ± 0.4 mg/L, orthophosphate-phosphorus ($\text{PO}_4\text{-P}$) 3.93 ± 0.2 mg/L, total nitrogen (TN) 59.4 ± 1.0 mg/L and ammonia-nitrogen ($\text{NH}_4\text{-N}$) 17.1 ± 0.4 mg/L. Ferric iron-facilitated CEPS was conducted on the wastewater samples with FeCl_3 used as the coagulant. The CEPS sludge was then processed with acidogenic fermentation for organic carbon and phosphorus recovery.

An FeCl_3 dose of 20 mg-Fe/L was used in the CEPS treatment, which was determined based on the laboratory jar-test results shown in Fig. 1. A pre-determined amount of FeCl_3 solution was added to 50 L raw sewage in a water tank without pH adjustment. After the addition of the coagulant, mechanical mixing was applied at 200 rpm for 1 min and then 30 rpm for 15 min, followed by sedimentation for 1 h. The supernatant was then siphoned off, and the sediment, which was 18-times reduced in volume, was retained as the CEPS sludge (Fe-sludge). The control sludge (simple primary sludge) was obtained from wastewater without FeCl_3 addition after the same sedimentation and supernatant discharge procedure, and its solid and organic contents were significantly lower than that of the Fe-sludge.

The sludge samples obtained were fermented under anaerobic conditions for hydrolysis and material recovery. The fermentation experiments were carried out using 550-mL glass bottles as batch reactors, each filled with 470 mL of the sludge mixture and 30 mL seed sludge. The seed sludge was from a sludge fermentation reactor that had been operated for more than 6 months. The sludge bottles were sealed, and the headspace of the bottles was purged with nitrogen to ensure an anaerobic environment. The bottles were placed in a temperature-controlled air chamber (25 °C), with magnetic stirring for mixing. In addition to the Fe-sludge and primary sludge, the Fe-sludge samples after pretreatment was also tested for the fermentation performance. Based on previous studies by others [13,14], ultrasonication (20 kHz, 0.36 kW/L for 10 min) and alkali treatment (initial adjustment to pH 10.0 by 2 M NaOH) were used to pretreat the sludge before fermentation. The four sets of sludge samples (Pri, Fe, Fe-S and Fe-Na) were fermented in parallel for up to 11 d. For each sludge sample, the fermentation test was conducted in triplicate reactors. The sludge mixtures were sampled every two days to evaluate the fermentation results. To recover P

from the sludge, the fermented sludge was centrifuged at 4000 rpm for 10 min, and the supernatant was adjusted to pH 8.0 with 2 M NaOH to form P-containing precipitates.

2.2. Analytical methods

A pH meter (Starter 3100, Ohaus, USA) was used to measure the solution pH. The sludge samples from reactors were centrifuged at 8000 rpm for 10 min to separate the solids from the supernatants. The SS and volatile suspended solids (VSS), COD, TN, $\text{NH}_4\text{-N}$, TP and $\text{PO}_4\text{-P}$ were determined in accordance with Standard Methods [15]. As Fe(II) in solution might interfere with the COD measurement, the organic content in the sludge was measured using a TOC analyzer (Aurora 1030, OI Analytical, USA) for the overall TOC in the mixture and soluble organic carbon (SOC) in the supernatant. Analysis of the same sample was performed in triplicate, and the results reported below are the mean values of the measurements. VFAs in the supernatant, including acetic acid (HAc), propionic acid (HPr), n-butyric acid (n-HBu), iso-butyric acid (iso-HBu), n-valeric acid (n-HVa) and iso-valeric acid (iso-HVa), were quantified using gas chromatography (GC; 6890A, Agilent, USA) with an HP-FFAP (30 m \times 0.25 mm \times 0.25 μm) capillary column and a flame ionization detector, following the method detailed previously [16]. The recovered P-containing product was analyzed by X-ray diffraction (XRD; Bruker AXS GmbH, Germany).

3. Results and discussion

3.1. Enhanced organic carbon and phosphorus removal by CEPS

As shown in Fig. 1, simple primary sedimentation (0 mg Fe/L) only reduced TOC by 38.8% and $\text{PO}_4\text{-P}$ by 5.9% on average from the domestic wastewater. With FeCl_3 flocculation, the CEPS significantly enhanced both organic carbon and $\text{PO}_4\text{-P}$ removal. At a dosage of 20 mg-Fe/L, the TOC and $\text{PO}_4\text{-P}$ removal efficiencies increased to 75.6% and 99.3%, respectively. Compared to the primary effluent, the average TOC, TP and $\text{PO}_4\text{-P}$ concentrations of the CEPS effluent decreased from 90.1, 4.35 and 3.70 mg/L to 36.0, 0.56 and 0.03 mg/L, respectively. However, similar to previous reports [4], CEPS with prior chemical coagulation was not able to improve N removal from wastewater. According to the mechanisms of CEPS [3], dosing with Fe(III) induces Fe^{3+} -based charge neutralization and sweep coagulation, leading to enhanced removal of PO_4^{3-} and particulate organics. In contrast, the positively charged NH_4^+ cannot be neutralized and removed by the positively charged Fe^{3+} and Fe(III) hydrolysis species. Overall, more than 70% of total organics and 95% of phosphate were removed from wastewater by CEPS into the sludge. After sedimentation to the same sludge volume of 2.8 L, the Fe-sludge contained 1993.7 mg/L of TOC and 70.0 mg/L of TP on average, whereas the primary sludge had only 1029.3 mg/L of TOC and 15.6 mg/L of TP.

3.2. Organic release and recovery during sludge fermentation

Sludge fermentation under anaerobic conditions induces organic hydrolysis and acidogenesis. As shown in Fig. 2a, with the solubilization of particulate organics from the sludge, the SOC concentration increased gradually in the supernatants. Moreover, compared to the primary sludge, the Fe-sludge had a higher organic content and exhibited a much more significant increase in SOC during fermentation. Most of the organic hydrolysis in the two sludge samples took place in the first 7 d of fermentation. After 11 d, the SOC increased to 283 mg-C/L in the primary sludge mixture and to 725 mg-C/L in the Fe-sludge. Accordingly, the soluble organic yield, or the average hydrolysis efficiency (SOC/TOC), reached 27.5% and 36.4% for the primary sludge and Fe-sludge, respectively.

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