



Enhancing phosphorus release from waste activated sludge containing ferric or aluminum phosphates by EDTA addition during anaerobic fermentation process



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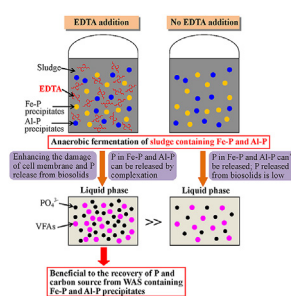
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HIGHLIGHTS

- EDTA addition can enhance P release from biosolids, Fe-P and Al-P during fermentation.
- P in Fe-P is much easier to be released than P in Al-P during MS fermentation.
- EDTA addition facilitates the production of STOC and VFAs, as well as solid reduction.
- Proper EDTA dose for both P release and sludge fermentation process is obtained.

GRAPHICAL ABSTRACT



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ABSTRACT

The effect of ethylene diamine tetraacetic acid (EDTA) addition on phosphorus release from biosolids and phosphate precipitates during anaerobic fermentation was investigated. Meanwhile, the impact of EDTA addition on the anaerobic fermentation process was revealed. The results indicate that EDTA addition significantly enhanced the release of phosphorus from biosolids, ferric phosphate precipitate and aluminum phosphate precipitate during anaerobic fermentation, which is attributed to the complexation of metal ions and damage of cell membrane caused by EDTA. With the optimal EDTA addition of 19.5 mM (0.41 gEDTA/gSS), phosphorus release efficiency from biosolids was 82%, which was much higher than that (40%) without EDTA addition. Meanwhile, with 19.5 mM EDTA addition, almost all the phosphorus in ferric phosphate precipitate was released, while only 57% of phosphorus in aluminum phosphate precipitate was released. This indicates that phosphorus in ferric phosphate precipitate was much easier to be released than that in aluminum phosphate precipitate during anaerobic fermentation of sludge. In addition, proper EDTA addition facilitated the production of soluble total organic carbon and volatile fatty acids, as well as solid reduction during sludge fermentation, although methane production could be inhibited. Therefore, EDTA addition can be used as an alternative method for recovering phosphorus from waste activated sludge containing ferric or aluminum precipitates, as well as recovery of soluble carbon source.

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1. Introduction

Phosphorus (P) is a non-renewable resource and an irreplaceable nutrient element for animals and plants. However, phosphate rock was reported to be depleted in 50–100 years, which will necessitate its recovery in the coming century (Cordell et al., 2009). On the other hand, about 1.3 Mt P/year is being treated in municipal wastewater treatment plants (WWTPs) on a global scale (Van Vuuren et al., 2010), and almost all the P is ultimately converged to the sludge phase during wastewater treatment processes. Therefore, P recovery from waste activated sludge (WAS) is an important alternative way to obtain P resource.

Chemical P removal process using Fe or Al salts is widely used in WWTPs to meet with the strict P discharge regulations for wastewater (Wilfert et al., 2015). Even in plants using enhanced biological P removal process, chemical flocculants are often dosed to guarantee the steady P removal (Wilfert et al., 2015). The chemical flocculants are usually dosed before or after the biological treatment units, and phosphate precipitates such as ferric phosphate (Fe-P) and aluminum phosphate (Al-P) often mix with the biosolids. It is estimated that approximately 40% of P is bound biologically and around another 50% exists as inorganic P (IP) in WAS (Xu et al., 2015). In nitrifying aerobic granular sludge, a higher percentage of IP was observed due to the accumulation of metal ions (Huang et al., 2015a). Therefore, P in phosphate precipitates is another important P resource pool, and it is essential to recover P from both biosolids and phosphate precipitates.

It is well known that P can be released during anaerobic digestion due to the decay of cells, resulting in a high P concentration in the supernatant (Chen et al., 2007; Latif et al., 2015). Consequently, P in the supernatant can be recovered as valuable products such as struvite and hydroxyapatite (Huang et al., 2015b; Tong and Chen, 2009). However, P in Fe-P and Al-P precipitates is difficult to be released during the conventional anaerobic digestion process (Braak et al., 2016; Latif et al., 2015). What is worse, chemical flocculants are commonly dosed excessively during wastewater treatment process. The excessive chemical flocculants can precipitate the P released from biosolids during anaerobic digestion, resulting in a low P concentration in the supernatant. This influences the recovery of P resource. Therefore, it is of interest to enhance the release of P from biosolids and phosphate precipitates using some advanced technologies so as to recycle more P resource from WAS. Pretreatments such as low-temperature thermal, alkali and ultrasonic treatments have been proved to be effective methods for improving the P release from WAS by promoting the sludge disintegration (Kim et al., 2015; Kuroda et al., 2002; Wang et al., 2010). However, these pretreatments are nearly useless to the release of P in Fe-P and Al-P precipitates. Although acidic treatment can improve the P release from phosphate precipitates to a certain extent (Chen et al., 2007; Xu et al., 2015), it has an adverse effect on the WAS treatment due to its inhibition to the activity of microorganisms below pH 4.0 (Hwang et al., 2004).

Ethylene diamine tetraacetic acid (EDTA) is the most commonly used chelating reagent. Recently, some studies have reported that EDTA used in the sludge anaerobic fermentation has a considerably positive effect on the production of methane and volatile fatty acids (VFAs) (Liu et al., 2016; Nguyen et al., 2014; Vintiloiu et al., 2013). Furthermore, EDTA could obviously improve the release of polyphosphate from sewage sludge during thermal pretreatment (Zhang and Kuba, 2014). Zhang et al. (2010) also reported that most P (91%) can be released from the digested dairy effluent after adding EDTA. However, specific studies related to the enhancement of P release from both biosolids and phosphate precipitates by EDTA addition during anaerobic fermentation are still rarely

reported.

This study aimed at understanding the effect of EDTA addition on P release from both biosolids and phosphate precipitates (Fe-P and Al-P) during anaerobic fermentation process. Moreover, sludge hydrolysis, acidification and methane production potential were also investigated to reveal the effect of EDTA addition on anaerobic fermentation process. The results presented in this study would help to enrich the method of P recovery from WAS containing phosphate precipitates.

2. Materials and methods

2.1. Source of the waste activated sludge

The WAS was obtained from the secondary sedimentation tank of a municipal WWTP in Shanghai, China. This plant was designed to treat municipal wastewater using an anaerobic-anoxic-aerobic activated sludge process. Chemical flocculants for P removal were not added. The sludge was filtered using a 1 mm × 1 mm screen and concentrated by settling for approximately 24 h at 4 °C prior to use. The main characteristics of the concentrated sludge are as follows: pH 6.8 ± 0.2, suspended solids (SS) 18.6 ± 0.8 g/L, volatile suspended solids (VSS) 12.5 ± 0.6 g/L, total P (TP) content 29.6 ± 0.8 mg/gVSS. In order to identify the P released from biosolids and phosphate precipitates, FePO₄·4H₂O or AlPO₄ was dosed in the WAS, forming the mixed sludge containing Fe-P (denoted as MS-Fe) or Al-P (denoted as MS-Al) precipitates. FePO₄·4H₂O (chemical pure, ≥98.0%) and AlPO₄ (chemical pure, 85 ± 5%) were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China).

2.2. Experimental procedure

Firstly, the effect of EDTA on P release from Fe-P or Al-P was investigated using identical serum bottles (V = 600 mL) containing 400 mL of EDTA solution. One mM of P as FePO₄·4H₂O or AlPO₄ was dosed in each bottle. Three EDTA concentrations (1, 5, 10 mM) were investigated at 35 °C and neutral pH. Moreover, in order to investigate the effects of temperature and pH on P release from the phosphate precipitates, three temperatures (0, 35 and 50 °C) and three pH values (5, 7 and 9) were tested, respectively, at the initial EDTA to metal ion (Fe or Al) molar ratio of 5:1. All experiments were conducted in an air-bath shaker (200 rpm) for 6 days, and each experiment was performed in triplicate.

Secondly, the effect of EDTA on P release from biosolids, Fe-P and Al-P precipitates during sludge fermentation was investigated using identical serum bottles (V = 600 mL). 12 serum bottles were divided into three groups (four bottles in each), receiving WAS, MS-Fe and MS-Al, respectively. In each bottle, 300 mL of WAS was dosed, and then 100 mL of EDTA stock solution was added. Four initial EDTA concentrations (0, 6.5, 19.5 and 32.5 mM) were tested. The initial concentrations of SS and VSS in each bottle were 13.9 and 9.4 g/L, respectively. The dosages of FePO₄·4H₂O in MS-Fe and AlPO₄ in MS-Al were 1440 and 787 mg/L, respectively, equivalent to 200 mg P/L (6.5 mM-P). All bottles were purged with nitrogen gas for 2 min to remove oxygen, and then were capped with rubber stoppers, sealed and placed in an air-bath shaker (200 rpm, 35 ± 1 °C) for anaerobic fermentation. The pH values in each bottle varied arbitrarily without any control during anaerobic fermentation, and they were in the range of 6.1–7.1 (Fig. S1). The biogas and mixed liquor were sampled every day, and the total gas volume was measured according to the method described by Zhao et al. (2010). The mixed liquor samples were centrifuged at 8000 rpm for 15 min, and then the supernatant was filtered through a 0.45 μm cellulose membrane and used for chemical analysis. The solid samples were

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