



Acetate favors more phosphorus accumulation into aerobic granular sludge than propionate during the treatment of synthetic fermentation liquor

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HIGHLIGHTS

- Ra- and Rp-granules were compared with respect to P recovery and bioavailability.
- Larger and more stable aerobic granules were achieved by mainly feeding acetate.
- Higher P removal capability and anaerobic P release were detected in Ra-granules.
- More cations were accumulated in Ra-granules while more organics in Rp-granules.
- More GAOs were found to exist in granules fed with propionate.

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ABSTRACT

Anaerobic digestion (AD) is an efficient biotechnology widely applied for energy and resource recovery from organic waste and wastewater treatment. The effluent from AD or fermentation liquor containing organic substances like volatile fatty acids (VFAs) and mineral nutrients (such as N and P), however, will trigger serious environmental issues if not properly dealt with. In this study two identical sequencing batch reactors (SBRs), namely Ra and Rp were used to cultivate aerobic granules for P recovery from synthetic fermentation liquor, respectively using acetate and propionate as additional carbon source. Larger and more stable granules were achieved in Ra with higher P removal capability (9.4 mg P/g-VSS-d) and higher anaerobic P release (6.9 mg P/g-VSS-h). In addition to much higher P content (78 mg P/g-SS), bioavailable P in Ra-granules increased to 45 mg P/g-SS, approximately 2-times those of seed sludge and Rp-granules. Microbial community analysis indicated that more GAOs were accumulated in Rp-granules.

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1. Introduction

Anaerobic digestion (AD) is regarded as one of the most important and effective stabilization, disposal and energy recovery processes for wasted activated sludge (WAS), livestock manure and other high-strength organic wastewaters or solid wastes. Through AD, organic carbonaceous substances can be converted into biogas to a greater extent, leaving all the other components such as mineral materials, non- or slowly biodegradable organics, and some intermediate products like volatile fatty acids (VFAs) in the diges-

tate or fermentation liquor (Barker et al., 1999; Ji and Chen, 2010). This huge amount of fermentation liquor, if not properly treated or discharged directly to the environment, will trigger serious environmental problems. Traditionally, fermentation liquor is either directly spread as liquid fertilizer or treated by solid-liquid separation, drying, filtration, etc., before land application (Möller and Müller, 2012). These practices sometimes encounter problems relating to land availability, long-distance transportation, and cost-effectiveness. Since fermentation liquor contains high levels of nutrients, especially nitrogen (N) and phosphorus (P), it is more applicable to firstly recover these resources and then treat it to meet the standards for final usage or disposal. Magnesium ammonium phosphate (MAP) precipitation can realize simultaneous

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recovery of $\text{NH}_4^+\text{-N}$ and ortho-P from WAS alkaline fermentation liquor, and after MAP precipitation the liquor can serve as additional carbon source for enhanced N and P removal from wastewater (Tong and Chen, 2009). However, due to the complex nature of fermentation liquor and the requirement for MAP formation ideally at $\text{Mg}^{2+}\text{:NH}_4^+\text{-N:PO}_4^{3-}\text{-P}$ of 1:1:1 (molar ratio), it is challenging to achieve stable nutrients recovery and high purity target product by using MAP method without adding other chemicals (like Mg^{2+}) and proper pretreatment processes. Other alternative and prospective processes are necessary for a better management of the fermentation liquor. Ammonia stripping has been proven to be effective for $\text{NH}_4^+\text{-N}$ recovery from digestate, and greater than 80–95% of total ammonia can be recovered after wet stripping at 35 °C and pH 10–11 for 3 h, leaving high levels of PO_4^{3-} and VFAs in the liquid phase (Huang et al., 2016). Further work is still demanding for VFAs utilization and P recovery from fermentation liquor.

Aerobic granule sludge (AGS) is considered to be one of the most promising biotechnologies for wastewater treatment. Up to the present, AGS with high treatment efficiency and much lower investment and operation costs has been successfully applied in large-scale domestic and industrial wastewater treatment plants (WWTPs) (Pronk et al., 2015; Nereda, 2016). Still, little information is available for AGS application in the treatment of fermentation liquor.

As reported, AGS can be used for effective P accumulation from wastewater treatment, achieving stable N and P removal (Schönborn et al., 2001; Wu et al., 2010). Solovchenko et al. (2016) claimed that P is always present in a form that does not meet the specifications for agriculture use when recovered chemically or biologically from wastewater. A previous work (Huang et al., 2015b) achieved P-rich granules with 93–95% of P bioavailability (i.e. the proportion of organic P and non-apatite inorganic P (NAIP) to the total P stored in granules) through enhanced P removal AGS process. Specifically, if stable P-rich granules with high P bioavailability could be cultivated during the treatment of fermentation liquor, P recovery from this huge amount of wastewater would not only greatly ameliorate environmental contamination but also ease the burden of rapid consumption of phosphate rock which is estimated to be depleted in 50–100 years (Cordell et al., 2009). Up to now, little information is available with respect to this aspect.

Restated, acetate and propionate are the two dominant VFA products which generally amount to 60–80% of the total VFAs, about 350–1330 mg/L in fermentation liquor regardless of solids retention time (SRT) varied from 5 to 10 days during anaerobic digestion of WAS (Yuan et al., 2009). According to previous studies, acetate- or propionate- dominant VFAs fermentation liquor can be achieved after adjustment or optimization of the AD operation conditions. In addition, how to get acetate- or propionate-dominant fermentation liquor for multi-functional utilizations is one of the research focuses of AD during recent years. Research works show that these two VFA species have different impact on phosphate accumulating organisms (PAOs) responsible for P uptake and accumulation from wastewater by using conventional enhanced biological P removal (EBPR) process (Chen et al., 2004) and on the structure of AGS (Lin et al., 2003; Wu et al., 2010). Currently no information can be found on the impact of these two major VFAs on P bioavailability of P-rich granules, the most important aspect of P recovery from fermentation liquor by using AGS process for agricultural purpose.

This study aimed to investigate the feasibility of cultivation of P-rich AGS through 6 months' operation of sequencing batch reactors (SBRs) to treat synthetic fermentation liquor after ammonia being recovered by stripping process (Huang et al., 2016). In addition to glucose, the two VFAs, namely acetate and propionate were

added as additional carbon source in the synthetic fermentation liquor. P species and its bioavailability in seed sludge and AGS were evaluated and compared. Finally, changes in microbial biodiversity in the granules cultivated with acetate and propionate were analyzed to shed light on the mechanisms involved in this complex granulation process.

2. Materials and methods

2.1. Experimental setup and operation conditions

Two identical laboratory scale SBRs made of acrylic plastic were used in this study. Their individual dimension was 6 cm × 6 cm × 60 cm (L × W × H) with working volume of 1.40 L. Seed sludge was sampled from the secondary sedimentation tank of the Shimodate Sewage Treatment Plant, Ibaraki Prefecture, Japan. Conventional activated sludge process is applied in this plant to treat domestic wastewater, mainly including primary sedimentation tank, aeration tank and secondary sedimentation tank. The initial concentrations of mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) were 4830 mg/L and 3570 mg/L (MLVSS/MLSS = 0.74), respectively in both reactors. Based on our previous works (Lin et al., 2013; Huang et al., 2015a; Huang et al., 2016), the synthetic wastewater used in these experiments consisted of 500 mg COD/L (glucose), 50 mg $\text{PO}_4^{3-}\text{-P/L}$ (KH_2PO_4), 100 mg $\text{NH}_4^+\text{-N/L}$ (NH_4Cl), 10 mg $\text{Ca}^{2+}\text{/L}$ (CaCl_2), 5 mg $\text{Mg}^{2+}\text{/L}$ ($\text{MgSO}_4\cdot 7\text{H}_2\text{O}$), 5 mg $\text{Fe}^{2+}\text{/L}$ ($\text{FeSO}_4\cdot 7\text{H}_2\text{O}$), and 1 ml/L of trace element solution to mimic the fermentation liquor after ammonia recovery by stripping. The composition of the trace elements solution was the same as Huang et al. (2015c). In addition to 500 mg COD/L of glucose, sodium acetate (~500 mg COD/L) and sodium propionate (~500 mg COD/L) were also used as carbon source and added into the influents of the two reactors, i.e. Ra and Rp, to simulate the acetate- and propionate- dominant fermentation liquor after mesophilic anaerobic digestion of WAS and livestock manure (Yuan et al., 2009; Huang et al., 2015a).

The two SBRs were operated automatically at room temperature (25 ± 2 °C) under alternative anaerobic and aerobic conditions with each cycle of 6 h. The initial one cycle consisted of 2 min feeding, 60 min non-aeration, 270 min aeration, 15 min settling, 2 min decanting and 11 min idling. After 10 days operation, due to improved settleability of the sludge the settling time was reduced to 2 min to wash out the sludge with poor settleability and to accelerate the granulation process, and the residual 13 min was used for idling. During aeration, air was provided by an air pump (AK-30, KOSHIN, Japan) from the bottom of SBRs through air bubble diffusers at an air flow rate of 0.6 cm/s. For each cycle, 0.76 L of supernatant was discharged right after the settling period (HRT ~ 11 h), and sludge retention time (SRT) of the two SBRs was controlled around 22 days. Dissolved oxygen (DO) concentration was 5–8 mg/L during aeration period. The granules in Ra and Rp were labelled as Ra-granules and Rp-granules. On day 130, the previous Rp-granules were evacuated and replaced by half of the Ra-granules in order to further clarify the influence of propionate on mature AGS functioned as P removal and P accumulation. The two SBRs were operated as same as day 130 before, and thereafter the granules in Ra and Rp were referred as Ra'-granules and Rp'-granules, respectively.

2.2. Analytical methods

2.2.1. Physicochemical characteristics of granules and wastewater

MLSS and MLVSS were used to quantify biomass growth in accordance with standard methods (APHA, 2012). DO concentra-

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