Bioresource Technology 271 (2019) 182–189

Contents lists available at ScienceDirect

Bioresource Technology

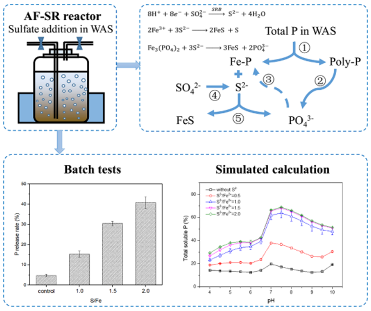
journal homepage: www.elsevier.com/locate/biortech

Simultaneous release of polyphosphate and iron-phosphate from waste activated sludge by anaerobic fermentation combined with sulfate reduction

Peishan Hu[a], Jianyong Liu[a,*], Liang Wu[a], Lianpei Zou[a], Yu-You Li[a,b], Zhi Ping Xu[a,c]

[a] School of Environmental and Chemical Engineering, Shanghai University, 333 Nanchen Road, Shanghai 200444, China
[b] Department of Civil and Environmental Engineering, Graduate School of Engineering, Tohoku University, 6-6-06 Aza, Aramaki, Aoba-ku, Sendai, Miyagi 980-8579, Japan
[c] ARC Centre of Excellence for Functional Nanomaterials, Australian Institute for Bioengineering and Nanotechnology, The University of Queensland, Brisbane, QLD 4072, Australia

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Polyphosphate (poly-P)
Iron-phosphate (Fe-P)
Waste activated sludge
Anaerobic fermentation
Sulfate reduction

ABSTRACT

Iron is widely used in sewage treatment systems and enriched into waste activated sludge (WAS), which is difficult and challenging to phosphorus (P) release and recovery. This study investigated simultaneous release performance of polyphosphate and iron-phosphate from iron-rich sludge via anaerobic fermentation combined with sulfate reduction (AF-SR) system. Batch tests were performed, with results showing that AF-SR system conducted a positive effect due to the relatively low solubility of ferrous sulfide in comparison with ferric phosphate precipitates. Simulation study was performed to investigate the total P release potential from actual waste activated sludge, finding that about 70% of the total P could release with the optimized pH of 7.0–8.0 and the theoretical S^{2-}/Fe^{2+} molar ratio of 1.0. A potential new blueprint of a wastewater treatment plant based on AF-SR system, towards P, N recovery and Fe, S, C recycle, was finally proposed.

1. Introduction

Phosphorus (P) is an indispensable nutrient for the normal growth and maintenance of all forms of life (Westheimer, 1987). However, it is estimated that phosphorus rock will encounter the shortage within 50–100 years due to the one-way flow of P from land to oceans (Elser and Bennett, 2011; Force, 2009). Therefore, P recovery from P-containing wastes is being paid much more attention in recent years (Cooper et al., 2011). Waste activated sludge (WAS), generated from wastewater treatment plants (WWTPs) with as high as about 2–10% of P content (Metcalf and Eddy, 2003; Oehmen et al., 2007), is a kind of P-containing waste for P recovery (García et al., 2006; Zhou et al., 2017).

---

* Corresponding author.
E-mail address: liujianyong@shu.edu.cn (J. Liu).

https://doi.org/10.1016/j.biortech.2018.09.117
Received 21 June 2018; Received in revised form 18 September 2018; Accepted 21 September 2018

It is well known that P in WAS originates from biological combined chemical P removal process by phosphorus accumulating organisms (PAOs) and chemical agents to meet the effluent requirement of WWTPs in general. Thus, polyphosphate (poly-P) and inorganic precipitated P were the two main fractions in WAS (Oehmen et al., 2007). Xu et al. proposed that approximately 40% of P in WAS is poly-P, while around 50% is inorganic P in precipitates (Xu et al., 2015). The specific composition of inorganic P depends directly on P removal agents, with aluminum and ferric salts being the most widely used (Gifford et al., 2015). It has been noted that the existence of virulence of microbial activity and public health risk for human determines the limitations of aluminum salts (Dorea and Clarke, 2008; Rosseland et al., 1990). Thus, iron salts are becoming the mainstream chemical additive for phosphorus removal in WWTP. In other words, it is essential to find an appropriate and efficient way for P recovery from poly-P and iron-phosphate (Fe-P).

As a traditional, convenient and economical process, anaerobic fermentation (AF) is considered to be one of the most desirable WAS treatment and poly-P release process (He et al., 2016). Various pretreatment methods, such as mechanical, low-temperature thermal, microwave were studied to improve the P release effect (Yang et al., 2015; Zou and Li, 2016). However, Fe-P in WAS would not release during AF due to its low solubility (Munch and Ottow, 1983). To make matters worse, some P released from poly-P is always recaptured by an overdose of phosphate removal agent with the increasing effluent quality (Xu et al., 2015). How to release Fe-P is a pending problem to be solved to improve the total P recovery rate from WAS.

Dosing sulfide was proved to be an effective method for Fe-P release because sulfide can replace phosphate from Fe-P. About 44.0% of P from pre-coagulated sludge could be extracted to liquid phase with NaHS addition (Kato et al., 2006). Furthermore, it was also proposed that S^{2-} from biological sulfate removal reactor could be collected and reused in P recovery process (Liu et al., 2015). In fact, environmental conditions of AF process are suitable for growth of sulfate reducing bacteria (SRB) (Chen et al., 2014; Muyzer and Stams, 2008; Weng et al., 2015). At the same time, volatile fatty acids (VFAs) produced by AF could also be used as carbon source of SRB, while sulfide produced by SRB could be consumed by iron, preventing SRB inhibition by H_2S accumulation. Presently, some studies have showed that the release tendency of phosphate increased in the high sulfur test (Ge et al., 2013; Takashima, 2018). However, this process of P release was often simplified with only the dissolved total P were analyzed. In fact, the forms of P are diverse in WAS (such as poly-P and Fe-P), and the final release rate of P is greatly related to influent properties and reaction conditions. Hence, it is of great necessity to figure out the specific releasing mechanism of different P species and the practicability of anaerobic fermentation combined with sulfate reduction (AF-SR) system.

The objectives of this study were: 1) to examine the feasibility of anaerobic fermentation combined with sulfate reduction (AF-SR) system to improve total P release rate from WAS; 2) to explore the release pathway of P in AF-SR system; 3) to investigate the suitable condition of AF-SR system for P release; 4) to evaluate if AF-SR system is applicable in reality. It could be expected that the results of this study be meaningful to improve P recovery rate from WAS using AF-SR system.

2. Materials and methods

2.1. Source of sludge

The WAS used in this study was obtained from the secondary sedimentation tank of a WWTP in Shanghai with both biological and Fe-P removal process. The inoculum sludge (IS) was collected from the anaerobic digester of another WWTP in Shanghai, and was domesticated for 2 weeks in laboratory with good ability of sulfate reduction. Both WAS and inoculum sludge were thickened by gravitational settling for 24 h, screened with a 1 mm sieve to get rid of impurities, and then reserved at 4 °C before later experiments. The main characteristics of WAS and inoculum were listed in Table 1.

Table 1
Characteristics of WAS and IS.

Parameters	WAS	IS
pH	6.9 ± 0.1	7.8 ± 0.1
TSS (g/L)	19.4 ± 0.3	33.5 ± 0.2
VSS (g/L)	13.4 ± 0.3	20.7 ± 0.1
TCOD (g/L)	13.7 ± 0.8	22.6 ± 0.6
SCOD (mg/L)	421.2 ± 12.0	334.2 ± 12.6
Total P (mg/g)	28.7 ± 0.5	19.2 ± 0.2
PO_4^{3-}-P (mg/L)	32.9 ± 0.2	151.8 ± 1.3
Total S (mg/g)	14.2 ± 0.2	12.2 ± 0.3
SO_4^{2-}-S (mg/L)	49.1 ± 4.7	180.1 ± 9.5
Total Fe (mg/L)	1048.1 ± 23.1	204.6 ± 42.3
Total Al (mg/L)	189.0 ± 15.4	1510.1 ± 60.5
Total Ca (mg/L)	309.6 ± 12.9	1075.8 ± 49.9
Total Mg (mg/L)	57.0 ± 13.3	329.6 ± 11.6

2.2. Batch tests of AF-SR

Laboratory scale of batch tests were set up in 2.0 L serum bottles containing 1.6 L of mixed sludge (effective volume) with the volumetric ratio of WAS to inoculum sludge of 9:1. The addition of sulfate depended on the original sulfur content. For the four experimental groups of feasibility study, the ratios of total sulfur to iron (S/Fe) was 0.5 (control without extra sulfate addition), 1.0, 1.5 and 2.0 by addition of sodium sulfate. The initial pH of the mixed sludge was adjust to 10.0 by adding 2 M sodium hydroxide (NaOH) for enough carbon source in the fermentation period (Wu et al., 2017; Yuan et al., 2006). Magnetic stirring thermostat water bathers (37 ± 1 °C) were used to maintain the temperature of the batch reactors for 20 days, with samples of day 0, 1, 2, 3, 5, 7, 10, 15 were obtained and analyzed. After solid-liquid separation, phosphorus recovery experiment was performed to investigate the P recovery efficiency from solution of AF-SR system with magnesium sulfate dosing (Mg: P = 2:1) and pH being adjusted to 10.0 by adding NaOH.

2.3. Analysis methods of sludge samples

The pH of the sludge was detected by a METTLER TOLEDO (FE20) pH-meter. The sludge samples were then centrifuged at 12,000 rpm/min for 10 min. The supernatant was filtered by 0.45 μm cellulose membrane for determination of PO_4^{3-}-P, SO_4^{2-}-S, soluble chemical oxygen demand (SCOD) in liquid, soluble chemical oxygen demand (TCOD) and the solid was dried at 105 °C for 24 h for total suspended solids (TSS), volatile suspended solids (VSS) and total sulfur analysis. Standards in Measurements and Testing (SMT) program extraction was used as the sequential extraction procedures for the determination of total P (TP), inorganic P (IP) and organic P (OP) in solid phase. The details of this extraction method were exhibited in previous studies (Xie et al., 2011). The TP is equal to PO_4^{3-}-P in liquid plus IP and OP in solid phase. The PO_4^{3-} in supernatant was determined to analyze the phosphorus fractions in solid phase. S elemental analyses in solid phase were performed by elemental analyzer (Vario Micro cube). The determinations of TSS, VSS, PO_4^{3-}-P, SO_4^{2-}-S and SCOD were tested by Standard Methods of water and wastewater (Clesceri et al., 2012). The samples were digested by nitric acid, perchloric acid and hydrofluoric acid, filtered by 0.45 μm cellulose membrane and tested by inductively coupled plasma atomic emission spectrometer (ICP-AES, Prodidy, Leeman Co.) for metal ions detection.