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Potassium ferrate addition as an alternative pre-treatment to enhance short-chain fatty acids production from waste activated sludge

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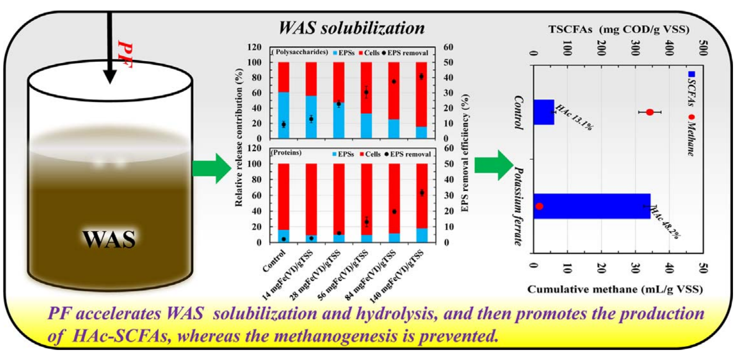
GRAPHICAL ABSTRACT



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ABSTRACT

A potentially practical technology based on ferrate (VI), i.e. potassium ferrate (PF), pretreatment integrated into waste activated sludge (WAS) anaerobic fermentation has been presented to greatly enhance short-chain fatty acids (SCFAs) production with a shortened fermentation time. The maximum production of SCFAs, 343 mg chemical oxygen demand/g volatile suspended solid with acetic acid proportion of 48.2%, was obtained with PF dosage of 56 mg Fe(VI)/g total suspended solid within 5 days, which was increased to 5.72 times compared to that of control. The mechanism study showed that PF accelerated the release rate of both intracellular and extracellular constituents. And the activities of key hydrolytic enzymes were much improved with PF addition. Moreover, PF positively enriched the abundance of microorganisms responsible for WAS hydrolysis and SCFAs production, especially acetic acid-forming characteristic genera such as *Petrimonas*, *Fusibacter* and *Acetoanaerobium*. Besides, the incubation time of acidogenesis and methanogenesis were separated by PF.

1. Introduction

The activated sludge process has been worldwide applied for wastewater treatment, and the by-product, waste activated sludge (WAS), are greatly generated (He et al., 2016a,b; Liu et al., 2016a). Sludge treatment has already been a serious problem because of its potentially environmental risk and high cost (Liu et al., 2016a; Feng et al., 2014; He et al., 2016a). Considering that WAS has an abundance of organic

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matters, anaerobic digestion is generally served as the most economic and efficient technology to simultaneously stabilize sludge and recover resources (e.g., short-chain fatty acids (SCFAs), methane and hydrogen, etc.) (Liu et al., 2016a; Luo et al., 2015; Zhang et al., 2016; Yang et al., 2017). Among the above recoverable resources, SCFAs, particularly acetic and propionic acids, present a potentially practical alternative to be used as carbon source for polyhydroxyalkanoates synthesis or biological nutrients removal system (Liu et al., 2016b; He et al., 2016b; Guo et al., 2015), as well as to reduce recovery cost compared to either methane or hydrogen (Lee et al., 2014; Luo et al., 2015). Therefore, to produce SCFAs from WAS has gained more and more attention.

In general, anaerobic digestion process of WAS includes three steps: solubilization and hydrolysis, acidogenesis and methanogenesis (He et al., 2016a; Li et al., 2015). On basis of SCFAs production just involved solubilization and hydrolysis, and acidogenesis (Lee et al., 2014), to maximize SCFAs accumulation, on one hand, the first two steps should be enhanced (Lee et al., 2014; Li et al., 2015; Liu et al., 2016b), and on the other hand, the methanogenesis should be prevented (Li et al., 2015; Luo et al., 2015). So far, many researches have been conducted to accelerate sludge solubilization and hydrolysis, such as physical (Lee et al., 2014; Liu et al., 2016c; Kavitha et al., 2016a), chemical (He et al., 2016c; Jie et al., 2014; Liu et al., 2012; Li et al., 2015; Su et al., 2013), thermal (Yang et al., 2015) and combination treatments (Kavitha et al., 2016b; Yang et al., 2015; Zhao et al., 2015). However, high energy demand has been a limited factor for their practical application (Lee et al., 2014; Luo et al., 2015). For decades, advanced oxidation processes (AOPs) have been given much attention for sludge dewatering (Ye et al., 2012b; Zhang et al., 2012), in view of low-cost and high-efficiency potential (Zhang et al., 2012; Zhou et al., 2014). AOPs not only have the potential to degrade extracellular polymeric substance (EPS), but also can destruct bacterial cell membranes (Ye et al., 2012a; Zhou et al., 2014). Until now, however, little studies have diverted to the combination of AOPs with anaerobic digestion for sludge treatment.

Ferrate (VI), the oxyanion FeO_4^{2-} containing iron in +6 oxidation state, particularly potassium ferrate (PF), has been considered as an environmentally-friendly oxidant in both water and wastewater treatment (Gombos et al., 2013; Yang et al., 2012; Zheng and Deng, 2016). It has also been applied for sludge dewatering due to its dual functions of an oxidant and a subsequent coagulant/precipitant by forming ferric hydroxide (Yang et al., 2012; Ye et al., 2012a; Zhang et al., 2012). It can be expected that on one hand, organic matters in EPS and cells will be released because of destruction of EPS and disintegration of bacterial cell membranes caused by PF, which contributes to the acceleration of WAS solubilization and hydrolysis, and then acidification. On the other hand, as one reason for the low production of SCFAs from WAS is attributed to rapid consumption by methanogens (Zhao et al., 2015), if PF pretreatment can prevent methanogenesis at some level, the accumulation of SCFAs will be enhanced. To date, however, little has been reported the effects of PF pretreatment integrated into WAS anaerobic fermentation on SCFAs production, in addition, whether the incubation time of acidogenesis and methanogenesis can be separated by PF also remains unknown.

This study, therefore, aimed to develop a highly-efficient and environmentally-friendly technology by adding PF into WAS anaerobic fermentation system to improve solubilization, hydrolysis and acidification of particulate organic matters (POMs), but prevent the methanogenesis for remarkably promoting both quantity and speed of SCFAs. The effects of PF dosages on production and composition of SCFAs were investigated. Moreover, the mechanisms of enhanced SCFAs production were explored by analyzing the role of PF on disintegration of both EPS and microbial cell, the activities of the key enzymes and microorganisms, and the relevant microbial community structure. Based on the findings of this work, it is expected to propose a potentially practical technology for producing carbon source from WAS.

2. Materials and methods

2.1. Reagents and WAS properties

The PF (K_2FeO_4, with a purity of above 90% as Fe(VI) (w/w)) was purchased from Shanghai Macklin Biochemical Co., Ltd., China. The WAS was collected from the secondary sedimentation tank of a municipal WWTP, Harbin, China, and stored as the previous studies described (He et al., 2016a,b). The main characteristics of applied WAS were: total suspended solid (TSS) 15,000 ± 362 mg/L, volatile suspended solid (VSS) 8,740 ± 157 mg/L, total chemical oxygen demand (TCOD) 12,158 ± 222 mg/L, soluble COD (SCOD) 280 ± 10 mg/L, soluble proteins 137 ± 3 mg COD/L, soluble polysaccharides 9 ± 0 mg COD/L, total SCFAs (TSCFAs) 66 ± 9 mg COD/L, NH_4^+ 54.55 ± 1.88 mg/L, PO_4^{3-} 73.02 ± 5.87 mg/L, and pH 6.89 ± 0.01.

2.2. Experimental set-up and operation conditions

2.2.1. PF integrated into WAS anaerobic fermentation for SCFA production

The effects of PF on WAS anaerobic fermentation system for SCFAs production were conducted in batch laboratory-scale reactors by using serum bottles (V = 500 mL) filled with 350 mL of WAS each. The addition levels of PF were 0, 14, 28, 56, 84, and 140 mg Fe(VI)/g TSS, respectively. There were two stages involved in this integration technology. At the first stage, the reactors filled with sludge and PF were stirred at 400 rpm for 30 min with magnetic stirring apparatus at ambient temperature to mix sludge and PF rapidly and completely. And at the second stage, nitrogen gas was introduced to the reactors for 10 min to remove oxygen, then the reactors were capped, sealed, and stirred in an air-bath shaker (100 rpm) at 35 ± 1 °C for 12 days. All the fermentation experiments were carried out in triplicate.

2.2.2. SCFA consumption and methane production with PF addition

Herein, the other 6 reactors were set up to investigate the effects of PF integrated into anaerobic digestion for methane production. One group was operated without PF, serving as the control, and the other was operated with PF at dosage of 56 mg Fe(VI)/g TSS, serving as the experiment group. The operation condition was the same as Section 2.2.1. And the difference was that no samples were taken during the 30-day anaerobic digestion period.

2.3. Mechanism exploration for SCFA production by PF addition

Microbial EPS is believed to act as the glue that binds cells together to form sludge flocs (He et al., 2016c; Li and Yang, 2007), and recognized as the key factor to maintain the structure and function of microbial aggregate (He et al., 2016c). EPS are recorded as a dynamic double-layered structure of loosely bound EPS (LB-EPS) diffused from the tightly bound EPS (TB-EPS), which surrounds the cells (Li and Yang, 2007), this work has divided EPS into two parts (i.e., LB and TB-EPS). And the methods for extraction and determination of EPS were the same as the previous publications (He et al., 2016c).

To investigate the influence of PF addition on WAS solubilization and hydrolysis, concentrations of polysaccharides, proteins, total organic carbon (TOC), and total nitrogen (TN) in both suspension and EPS at 1-day fermentation time were analyzed. Also, the activity of the key hydrolytic enzymes (i.e., α-glucosidase and protease) were measured. And to explore the effect of PF on shift of microbial community structure related to WAS hydrolysis and acidification, three sludge samples (raw sludge, control, and PF (56 mg Fe(VI)/g TSS) treatments) were analyzed at 4^{th} day fermentation time.

2.4. Analytical methods

Sludge samples collected from the reactors were centrifuged at