Chemical Engineering Journal 303 (2016) 636–645

Contents lists available at ScienceDirect

Chemical Engineering Journal

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

Enhancing the sludge dewaterability by electrolysis/electrocoagulation combined with zero-valent iron activated persulfate process

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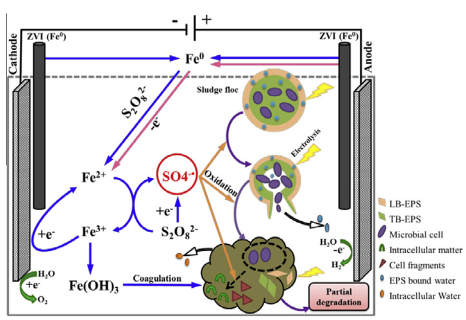
HIGHLIGHTS

- EL/EC and ZVI activated persulfate had synergetic effect on sludge dewatering.
- EZP conditioning effectively enhanced different types of sludge dewaterability.
- EZP oxidation destroyed the EPS and cells, releasing bound water in sludge flocs.
- Degradation of protein-like substances contributed to sludge dewatering.
- Cost of EZP conditioning saving up to 41.9% in comparison with Fenton method.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 28 March 2016
Received in revised form 7 June 2016
Accepted 8 June 2016
Available online 11 June 2016

Keywords:
Wasted activated sludge (WAS)
Dewaterability
Electrocoagulation
Persulfate
Extracellular polymeric substance (EPS)

ABSTRACT

Waste activated sludge (WAS) is difficult to be dewatered due to the highly water bounded in sludge flocs, and the extracellular polymeric substances (EPS) was the major factor affecting sludge dewatering performance. In this study, the hybrid process of electrolysis/electrocoagulation and zero-valent iron activated persulfate oxidation (EZP) showed a significant synergetic effect in enhancing municipal sludge dewaterability, and has the potential for enhancing industrial sludge dewaterability. The optimal dewatering conditions for municipal sludge were voltage 40 V and 4.15 g/L $Na_2S_2O_8$ when zero-valent iron induced electrodes were applied. After EZP pretreatment, the municipal sludge specific resistance to filtration (SRF) and capillary suction time (CST) decreased by 87.4% and 49.1% respectively. The effects of EZP pretreatment on zeta potential, EPS property, viscosity and dewaterability of different sludge were analyzed to unravel the underlying mechanism of sludge conditioning. Results showed that the EZP oxidation was capable to effectively disrupt the EPS, crack the entrapped cells, and degrade the protein-like substances, reducing the viscosity and negative zeta potential, releasing bound water inside EPS and cells and thus improving sludge dewaterability. According to the analysis of Three-dimensional excitation emission matrix (3D-EEM), the EZP technology greatly decomposed tryptophan and aromatic protein-like substances in EPS. Scanning electron microscope (SEM) analysis further revealed that the disrupted EPS and cells were coagulated after EZP conditioning and reinforced sludge dewatering. The preliminary economic analysis showed that the optimized EZP was economically favorable.

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http://dx.doi.org/10.1016/j.cej.2016.06.041
1385-8947/

1. Introduction

Waste activated sludge (WAS) is continuously generated from municipal and industrial waste water treatment plants (WWTP) by activated sludge process, which must be properly treated and disposed [1]. Nowadays, the treatment and disposal of excess WAS accounts for 50–60% of total operating expense wastewater treatment process [2]. The disposal of wastewater sludge still remains crucial technical challenges due to the high moisture content of different types of sludge [3]. Dewatering is an important step for reducing water content and sludge volume, which is an efficient method to reduce transportation and disposal costs [3–5]. However, the complex colloidal nature and strong hydrophilicity of the WAS substantially hamper the dewatering process without pretreatment, failing to meet the requirements of subsequent sludge disposal [3,6].

To improve the dewaterability of sludge, different pretreatment processes have been widely studied and developed including thermal, chemical, magnetic mechanical, acoustic, electric, electro-acoustic and their hybrid treatment processes [7]. Among them, electrolysis is considered as an efficient way because of its potential to remove interstitial and vicinal water from sludge flocs [7]. Moreover, it has less adverse effects to environment and is cost-effective [8]. The simultaneous application of electrolysis and electrocoagulation enhances the performance of sludge dewaterability and disintegration [1]. Unfortunately, electrocoagulation is only able to control the extracellular polymeric substances (EPS) concentration through charge neutralization and bridging but with limited impact for degrading EPS and simultaneously destroying cells [6].

As is known to all, EPS are a crucial constituent of the floc matrix, which contributes to the formation of floc, surface properties of floc and floc strength [9]. Furthermore, EPS is the major organic fraction in activated sludge which has been found to account for 80% of the mass of activated sludge [3]. The distribution and chemical composition of EPS had significant effect on sludge dewatering performance [10]. EPS was found to have double layers including the loosely bound EPS (LB-EPS) which is diffused from the tightly bound EPS (TB-EPS) that surrounds the cells [11]. Li and Yang [11] found that sludge settleability and dewaterability were much more strongly related with the concentration of LB-EPS than TB-EPS, and excessive LB-EPS always worsened sludge dewaterability [11,12]. Moreover, the water retained in the EPS-structure is bound mainly by the polysaccharides (PS) and proteins (PN) of the EPS in the activated sludge. The molecular mechanisms of water binding are of crucial importance for a rational basis of the improvement of dewatering technology [13].

The advanced oxidation processes (AOPs) have turned out to be one of the promising and competitive technologies for improvement of sludge dewatering. The AOPs methods improve the dewaterability of the sludge by affecting the EPS in two ways: (1) they have the potential to degrade the EPS and; (2) they affect the multifunctional groups of the EPS and promote their participation in several interactions [13]. Typically, the release and degradation of EPS is mainly based on the generation of extremely reactive species like hydroxyl radicals ($HO^{\cdot}$) and sulfate radicals ($SO_4^{-\cdot}$) in AOPs [4,5,14]. In Fenton reaction, hydrogen peroxide was catalytically decomposed ferrous ion under acidic condition (normally pH 3) to generate $HO^{\cdot}$ (Eq. (1)).

$$H_2O_2 + Fe^{2+} \rightarrow Fe^{3+} + {}^{\cdot}OH + OH^- \tag{1}$$

Unfortunately, the hydrogen peroxide and Fe^{2+} are unstable. Furthermore, the requirement for the extremely acidic condition (i.e. pH 2.0–3.0) is also the drawback of the traditional Fenton method [5].

In recent years, activated persulfate receives much attention as an alternative oxidant. The Fe^{2+} and zero-valent iron (ZVI i.e. Fe^0) activated persulfate oxidation were applied to condition the WAS [2,4]. Compared with hydrogen peroxide, persulfate salt is easier and safer to transport and store. Moreover, Fe^{2+}/persulfate has a good oxidant effect in a wider range of pH from 2 to 10 [4]. In addition, ZVI is able to catalyze persulfate at neutral condition [15]. In our previous study, Fe^{2+} activated peroxymonosulfate (PMS) oxidation was employed to enhance the WAS dewatering [14]. The rapid disruption of EPS by $SO_4^{-\cdot}$ generated from ZVI or Fe^{2+} activated persulfate and PMS system could strengthen the sludge dewaterability, making it attractive for WAS dewatering. Generally, $SO_4^{-\cdot}$ can be generated by activating persulfate and PMS with heat, UV, or transition metal. The reactions between persulfate and Fe^{2+} or ZVI are shown in the following equations [16]:

$$S_2O_8^{2-} + Fe^0 \rightarrow Fe^{2+} + 2SO_4^{2-} \tag{2}$$

$$S_2O_8^{2-} + Fe^{2+} \rightarrow Fe^{3+} + SO_4^{-\cdot} + SO_4^{2-} \tag{3}$$

$$2Fe^{3+} + Fe^0 \rightarrow 3Fe^{2+} \tag{4}$$

Like the Fenton process (Eq. (1)), a high concentration of Fe^{2+} is required in the Fe^{2+} activated persulfate process due to the slow regeneration of Fe^{2+} after conversion to Fe^{3+} via reaction (Eq. (4)) [17]. Therefore, a great deal of iron sludge is generated during the conditioning stage [18]. When compared with the Fe^{2+} salt, ZVI is more stable and economical [2]. Recently, it has been declared that $SO_4^{-\cdot}$ could be generated via electrolytic activation of persulfate anions [18]. However, few of previous studies investigated the potential of combined electrolysis/electrocoagulation and ZVI activated persulfate (EZP) pretreatment on dewatering performance.

Hence, this study systematically researched dewaterability of variously composed waste activated sludge from three full-scale wastewater treatment plants via a novel EZP pretreatment. The objectives of this study were to: (1) optimize the operating parameters of EZP conditioning, (2) evaluate the potential of EZP system as a novel technic to enhance the different sludge dewaterability, (3) investigate the effect of zeta potential, EPS properties and viscosity on WAS dewaterability, (4) further explore the enhanced dewatering mechanism of sludge with EZP conditioning by three-dimensional excitation emission matrix (3D-EEM) spectra and SEM analysis, (5) assess preliminarily economic potential of EZP system.

2. Methods

2.1. Sludge sources and chemicals

The three types of WAS used in this study were collected from a municipal WWTP in Changsha, Hunan province, China (Sample 1), a caprolactam production WWTP in Yueyang, Hunan province, China (Sample 2), and a mixed WWTP of refining, cyclohexanone, rubber, epoxy resin and chemical fiber production wastewater in Yueyang, Hunan province, China (Sample 3). The collected sample was stored at 4 °C after screened through 4.0 mm sieve to remove grit. Table 1 tabulates the main characteristics of these WAS. All the chemicals used in this study were of analytical grade. Sodium persulfate ($Na_2S_2O_8$), 98% Sulfuric acid (H_2SO_4), Nitric acid (HNO_3) and Ferrous sulfate ($FeSO_4{\cdot}7H_2O$) were purchased from Sinopharm Chemical Reagent of China. All other chemical reagents used were of analytical grade.