

Improvement of sludge dewaterability and disintegration efficiency using electrolytic zero-valent iron activated peroxymonosulfate

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ABSTRACT

Electrolysis zero-valent iron activated peroxymonosulfate (EZVI-PMS) was applied to enhance sludge dewaterability and disintegration performance. Sludge dewaterability was characterized by capillary suction time (CST), specific resistance to filtration (SRF), and disintegration performance was explored by measuring sludge DNA content, ammonia nitrogen, chemical oxygen demand (COD), extracellular polymeric substances (EPS) and dissolved organic carbon (DOC). EPS, including soluble EPS (SB-EPS), loosely bound EPS (LB-EPS), and tightly bound EPS (TB-EPS) were analyzed by three dimensional fluorescence excitation-emission spectrum (3D-EEM) to confirm the proteins' transformation tendency. DOC, protein and polysaccharide in EPSs were quantified to investigate the conditioning mechanism. The results showed that sludge CST and SRF were reduced significantly when the current was 0.2 A and PMS dosage was 130 mg/gDS with the reductions of 43.8% and 74.1%, respectively, and DNA was released from sludge cells to the liquid phase. Mechanically, sludge TB-EPS converted to SB-EPS with DOC in TB-EPS decreasing from 367.0 mg/L to 210 mg/L, while DOC in SB-EPS increased from 44 mg/L to 167.4 mg/L. Besides, the changes of protein and polysaccharide contents in SB-EPS and TB-EPS were similar to DOC, and protein in TB-EPS transformed to other protein-like or organic substances obviously.

Key words: electrolysis zero-valent iron, extracellular polymeric substance (EPS), peroxymonosulfate (PMS), sludge disintegration, sludge filterability

HIGHLIGHTS

- Electrochemically activated peroxymonosulfate (PMS) was useful for conditioning.
- An electric current of 0.2 A was enough to produce Fe(II) to activate PMS using iron bar as the cathodes.
- Electrochemically activated PMS could oxidize extracellular polymeric substances in sludge to enhance sludge dewaterability.

1. INTRODUCTION

With the increase in the amount of sewage treatment, the waste activated sludge produced by waste water treatment plants (WWTPs) is huge in China (Wang *et al.* 2018; Gao *et al.* 2021), the moisture content of which reaches more than 99% (Liang *et al.* 2020), and is still about 96% even after the thickening process. In order to reduce the moisture content of waste activated sludge, sludge conditioning combined with mechanical dewatering is widely applied at present. However, sludge moisture content in most WWTPs can only be reduced to about 75% after most mechanical dewatering processes (Lin *et al.* 2020; Bian *et al.* 2021), which will undoubtedly produce huge costs for sludge transportation and final disposal (Ye *et al.* 2012).

Extracellular polymeric substances (EPS), as the main component of sludge flocs, account for 80% of sludge solids (Yang *et al.* 2017), and are mainly microbial metabolites, such as proteins, polysaccharides and humic substances (More *et al.* 2014). These substances are highly hydrophilic (Yuan *et al.* 2017), making EPS able to wrap up a large amount of water and impede the separation of sludge particles and water. In actuality, mechanical dewatering pressure is limited, but the EPS and bound water (including interstitial water, capillary water and intracellular water) are difficult to remove from the sludge even under high pressure (Wei *et al.* 2018). Therefore, it is necessary to destroy sludge EPS structure during sludge conditioning through chemical, physical and biological methods to improve the efficiency of sludge filtration, disintegration, and mechanical dewatering. However, simply destroying EPS structure does not necessarily improve the dewatering performance: if the properties

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of proteins and other substances in EPS do not change, water will still be wrapped in them. Therefore, EPS should be oxidized and its inherent properties should be changed in order to improve sludge dewatering performance effectively. At present, many researches are committed to dividing EPS into different layers, mainly soluble EPS (S-EPS), loosely bound EPS (LB-EPS) and tightly bound EPS (TB-EPS), and investigating the influence of each component on sludge dewaterability. Some researchers have found that LB-EPS content was positively correlated with dewaterability (Zhen *et al.* 2018), while others suggested that the TB-EPS content was negatively correlated with sludge dewaterability (Fan *et al.* 2020). As for the influence of protein, humic substances and polysaccharide, the main components in EPS, some researchers have found that a high protein amount in sludge is not a benefit for sludge dewaterability (You *et al.* 2017), while others believed that the substances played different roles in affecting sludge dewatering performance (Wu *et al.* 2016; Ge *et al.* 2019).

The advanced oxidation process, as a promising approach to sludge conditioning, has received extensive concern, because the oxidizers can disintegrate the sludge structure, organic matter and sludge cells (Feng *et al.* 2014), such as ozone (Zhang *et al.* 2016), peroxide (Kim *et al.* 2009), chlorine (Wang 2014), persulfate (PS) (Xiao *et al.* 2017; Maqbool *et al.* 2019), and Fenton reagent (Xiao *et al.* 2018; Cai *et al.* 2019) and so on, which are usually used to degrade macromolecular organic matter and reduce sludge volume, and some of them could also improve sludge dewatering performance. The oxidants listed above are almost all based on the production of highly active reaction free radicals, such as superoxide radicals, hydroxyl radicals and sulfate radicals. Due to producing sulfate radicals, the advanced oxidation process based on activated persulfate has been paid more and more attention in sludge conditioning due to the significant advantages in treatment effect and relatively loose use conditions, such as high REDOX potential, long continuous oxidation time and wide applicable pH range (Zhen *et al.* 2012a, 2012b). For example, some researchers have found that activated persulfate could degrade the digested sludge and enhance the sludge dewatering effect (Waclawek *et al.* 2016). Meanwhile, activated persulfate coupled with other sludge conditioning methods has also achieving excellent synergic effects, such as microwave and persulfate/Fe(II), which could reduce 94.6% CST (Zhen *et al.* 2019), and persulfate/Fe(II) with polymerized material could reduce 94.9% specific resistance to filtration (SRF) reduction (Wang *et al.* 2017).

However, as a kind of strong oxidant, persulfate produces sulfate radical only under certain activation conditions (such as transition metal, ultrasound and heat) (Li *et al.* 2018; Wang *et al.* 2018; Bian *et al.* 2021). At present, there are many researches on ferrous ion (Fe(II)) and thermally activated persulfate to improve the sludge filtration performance (Kim *et al.* 2016; Guo *et al.* 2019; Ge *et al.* 2020). Due to the easy operating condition and simple equipment, Fe(II) was the most widely used reagent to activate persulfate in sludge conditioning. It should be noted that Fe(II) readily converts to ferric iron, while the conversion rate of ferric iron (Fe(III)) converting to Fe(II) is very slow, which makes it necessary to add large amounts of ferrous salts to activate persulfate. When zero-valent iron (ZVI) is used as an electrode, sequential conversion between ZVI, Fe(II) and Fe(III) could be achieved easily, and the activation efficiency of persulfate could be improved by certain electrolytic conditions (Lin *et al.* 2014).

Therefore, the object of this study is to: investigate the improvement of sludge dewatering and disintegration performance using peroxymonosulfate (PMS) activated by electrolysis zero-valent iron (EZVI-PMS); evaluate the effects of the oxidizing condition on sludge dewaterability; examine the effects of EZVI-PMS oxidation on solubilization and physico-chemical properties of organic substances; and get more comprehensive insights into the mechanism of EZVI-PMS in conditioning sludge.

2. METHODS AND MATERIALS

2.1. Raw sludge

The raw sludge was taken from the reflux pipe from the secondary sedimentation tank to the biochemical pool in a WWTP using the improved oxidation ditch process in Wuhan city. The daily treatment capacity of the sewage plant is 50,000 t/d. The collected sludge was filtered using a 30 mesh sifter to get rid of large particulate matter then condensed to a moisture content of about 97.8%. The sludge was stored at 4 °C in a plastic bucket before use, and all the relevant indicators of sludge samples were measured integrally within 3 days. Raw sludge has a pH of about 7.4, suspended solids (SS) concentration of about 21.7 g/L, and volatile suspended solids (VSS) content of about 12.3 g/L.

2.2. Experimental method

The device for conditioning sludge with EZVI-PMS is shown in Figure 1. The anode and cathode both use iron rods, of which the diameter is 12 mm. The center distance between the two electrodes is 50 mm, and the depth of immersion in the sludge is 60 mm. During the experiment process, 600 g sludge was pour into a 1 L beaker, and PMS was dosed when the power supply

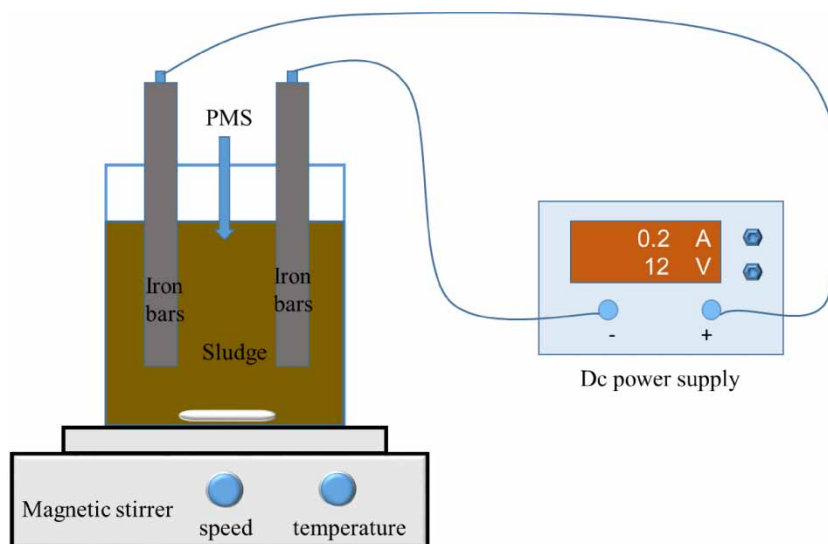


Figure 1 | Schematic diagram of the experimental apparatus.

was switched on. PMS dosage was controlled at 130 mg/gDS, which was determined by previous laboratory tests. The current levels were controlled at 0.2 A and 0.4 A, respectively. The experimental group with PMS was defined as the electric oxidation group, while the group without PMS was defined as the electric flocculation group. CST and other indicators were sampled at 15, 30 and 60 min in each group.

2.3. EPS extraction and detection

EPS was extracted using the thermal extraction method (Guo *et al.* 2019). In brief, 25 mL of sludge was centrifuged at 4,000 r/min. After the centrifugal liquid was filtered through a 0.45 μm membrane, the organic substance in the filtrate was defined as SB-EPS. The residue solid after centrifugation was diluted to the original volume with deionized water and centrifuged at 10,000 r/min. The organic matter in the filtrate was defined as LB-EPS after the centrifugal liquid was filtered through a 0.45 μm membrane. The residue solid was diluted to the original volume with deionized water once again. Then the sample was placed into a water bath at 80 $^{\circ}\text{C}$ for 10 min, followed by centrifuging at 4,000 r/min and collection of the centrifugate. The organic matter in the filtrate of the centrifugate filtering through a 0.45 μm membrane was defined as TB-EPS. The concentrations of dissolved organic carbon (DOC), polysaccharide (PS) and protein (PN) in SB-EPS, LB-EPS and TB-EPS were detected. The contribution of each EPS layer to sludge dewaterability was determined by a three-dimensional fluorescence excitation – emission (3D-EEM) spectrometer (F7000, Hitachi, Japan). The polysaccharide and protein content was detected by anthrone – concentrated sulfuric acid method and fast Lowry method, respectively. All indexes were tested three times and the average was taken.

2.4. Other detection methods

All the reagents used in the experiment were analytically pure, among which persulfate (PMS) was Aladdin's reagent. CST was monitored by a CST instrument (Triton, Modle 304M, Essex, UK). Specific resistance to filtration (SRF) was measured using the Buchner funnel method. According to the requirements of the PowerSoil[®] DNA Isolation Kit, DNA was extracted from 0.25 g of sludge for quantitative detection by UV-Vis spectrophotometer (NanoDrop ND-2000, Thermo Fisher Scientific, US). Sludge particle size could reflect the sludge accumulation or diffuse state and was analyzed by a laser particle analyzer (Mastersizer 2000, Pennsylvania-based firm, UK). SS, VSS and moisture contents were conducted by gravimetric method. DOC was analyzed by organic carbon analyzer (multi N/C 2100, Jena, Germany). Bound water was detected by the expansion method using dilatometer with xylene as the indicator (Huo *et al.* 2014). Chemical oxygen demand (COD) and ammonia nitrogen ($\text{NH}_4^+\text{-N}$) were measured using international standard methods. All indexes were tested three times and the average was taken.

3. RESULTS AND DISCUSSION

3.1. Effect of electro-oxidation on sludge filterability

The effects of EZVI-PMS on sludge dewaterability in terms of CST and SRF under different current intensities are shown in Figures 2(a) and 1(b). With the PMS dosage fixed at 130 mg/gDS and currents fixed at 0.2 and 0.4 A, respectively, sludge CST and SRF decreased rapidly and then tended to stabilize as the conditioning time increased from 15 to 60 min. The maximum reductions of CST and SRF were 49.6% and 76.9%, respectively. The results indicated that EZVI-PMS has a remarkable influence on sludge dewaterability. When sludge was conditioned by electro-flocculation only; that is, no PMS was added, CST and SRF changed slightly, no matter whether the current was 0.2 or 0.4 A. It suggested that the flocculation effect of Fe (II) and Fe (III) generated by electrolysis on sludge was negligible, so the sludge filtration performance could not be improved significantly. CST was signally reduced when both electrification and PMS were added to condition sludge. Sludge CST and SRF decreased from 195.5 s to 109.9 s and $1.47 \times 10^9 \text{ s}^2/\text{g}$ to $0.38 \times 10^9 \text{ s}^2/\text{g}$ with the reductions of 43.8% and 74.1%, respectively, as the current was 0.2 A and treatment was for 15 min. However, the CST just declined to 104.4 s and 100.5 s, with the treatment time increasing to 30 and 60 min, respectively, which indicated that the reduction of CST was not obvious when the treatment time was extended by a certain value. When the current was increased to 0.4 A, sludge CST decreased to only 105.3 s after 15 min conditioning. The changing regulation of SRF was similar to CST. According to the above analysis, when the current was higher than 0.2 A or the conditioning time exceeded 15 min, the improvement effect of sludge dewaterability tended to be stable. Therefore, in the subsequent experiment, the current intensity and treatment time were fixed at 0.2 A and 15 min, respectively.

3.2. The disintegration effect of electro-oxidation sludge

Sludge disintegration was mainly analysed from the following characterization parameters, namely sludge DNA, ammonia nitrogen and COD contents in the supernatant. As shown in Figure 3(a), the DNA content extracted from raw sludge and sludge conditioned by electro-flocculation was 29.0 mg/L and 27.3 mg/L respectively. Nevertheless, the DNA content in the electro-oxidized sludge was only 2.7 mg/L. The results showed that the cell structure of electro-oxidation treated sludge was broken and lysed, and resulted in DNA releasing into the liquid phase. For the sludge treated by electro-flocculation, there was no obvious release of DNA, indicating that the disintegration effect of electro-flocculation on sludge cells was negligible.

The change of ammonia nitrogen content in sludge supernate could show the degree of sludge disintegration on another aspect. The ratio of ammonia nitrogen in conditioned sludge supernate to that in raw sludge supernate is shown in Figure 3(b) under different treatment conditions, which were 1.02 and 1.93 respectively for electro-flocculation and electro-oxidation treated sludge, respectively. The results suggested that nitrogenous organic matter such as protein-like substance were

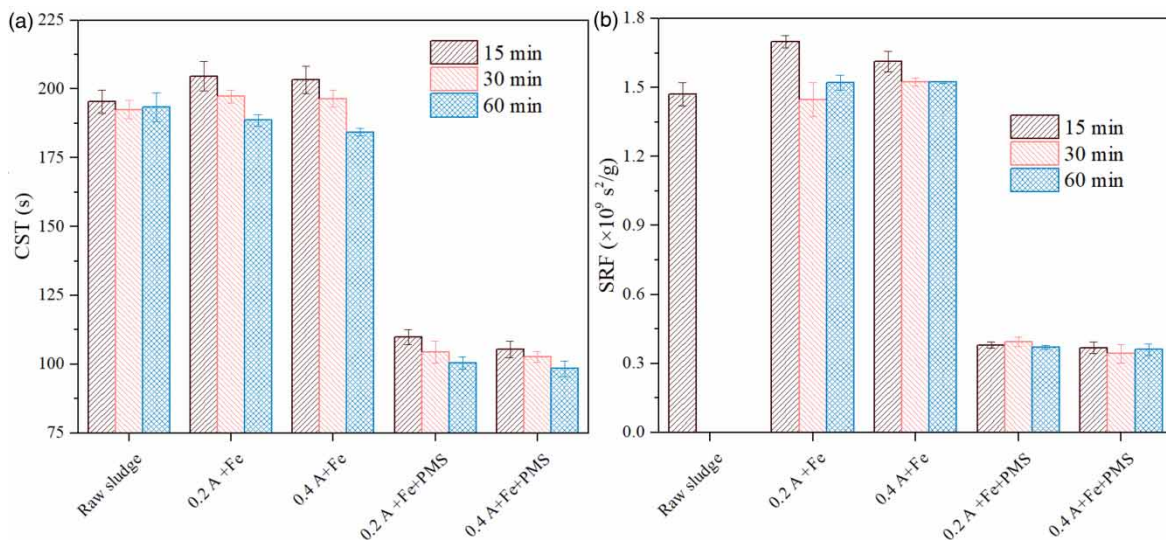


Figure 2 | Sludge CST change under different condition methods.

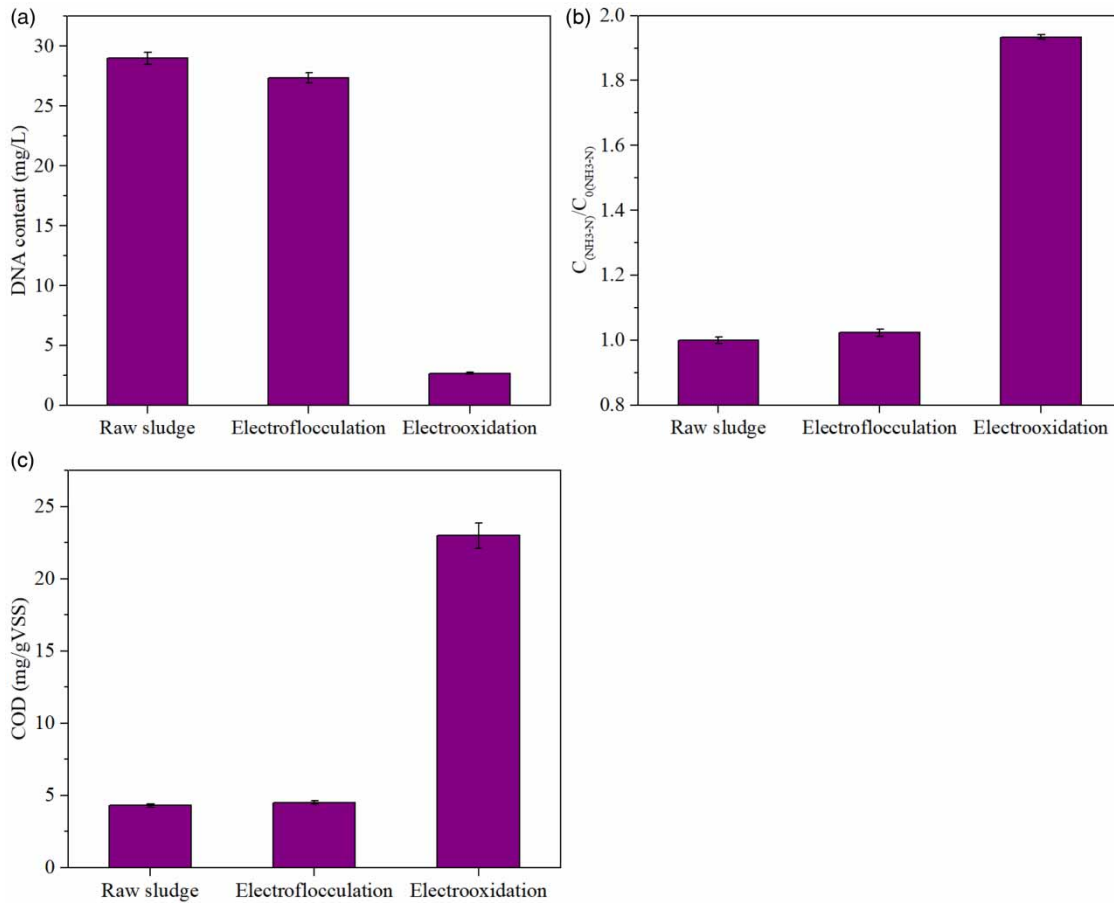


Figure 3 | The change of DNA in sludge (a), C_{NH_3-N}/C_{O-NH_3-N} (b) and COD (c) in supernate of sludge treated under 0.2 A current and 15 min condition time with 130 mg/gDS PMS.

partially hydrolyzed under the oxidation of EZVI-PMS, and ammonia nitrogen was produced and released into the liquid phase. However, electro-flocculation alone had no promoting effect on the release of ammonia nitrogen, and organic substances could not be oxidized in the sludge, so the ammonia nitrogen content was basically unchanged.

The COD contents in supernate of raw sludge, electro-flocculation and electro-oxidation treated sludge are showed in Figure 3(c). COD contents in electro-oxidized sludge was highest at about 5 times higher than the other two. The result showed that electro-oxidation could make sludge release COD into the liquid phase. Which was highly consistent with the change of ammonia nitrogen in supernate and contrary to DNA content in sludge. It is main attribution to the sludge disintegration promoted by EZVI-PMS, which destroyed the structure of the sludge flocs and the organic matter inside and outside the sludge cells.

3.3. Effects of electro-oxidation on sludge EPS

The DOC, protein and polysaccharide contents of S-EPS, LB-EPS and TB-EPS in each sludge sample are shown in Figure 4., The DOC content of S-EPS (as shown in Figure 4(a)) in electro-oxidation treatment sludge is the highest, at a value of 167.4 mg/L, while that of raw sludge and electro-flocculation treated sludge were relatively close, at about 45 mg/L. On the contrary, the DOC concentration in TB-EPS of electro-oxidized sludge was the lowest, at 210.4 mg/L, and the difference between the other two sludge samples was roughly equal with DOC content of about 375.0 mg/L. The difference in DOC content in LB-EPS extracted from the three sludge samples was relatively small. It can be seen from the above results that high DOC in TB-EPS would hamper sludge dewatering, and electro-oxidation can transform DOC in sludge TB-EPS, into DOC in SB-EPS, which was a benefit for sludge dewatering. It might be owing to that the S-EPS can be easily removed

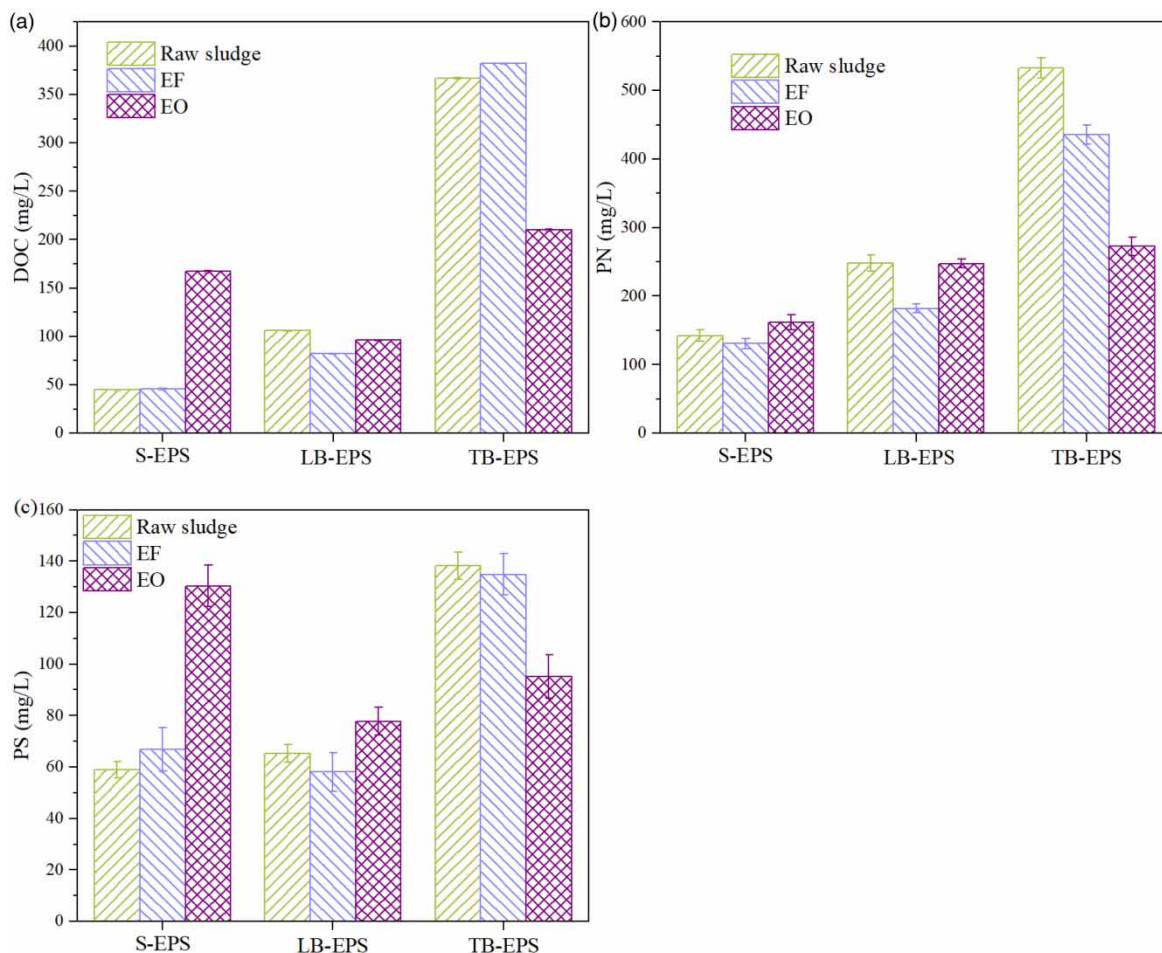


Figure 4 | Changes of (a) TOC; (b) polysaccharides; (c) proteins in the three EPSs of sludge treated under 0.2 A current and 15 min condition time with 130 mg/gDS PMS.

along with free water. Hence, it has an unobvious influence on limiting sludge dewatering (Bian *et al.* 2021). However, the compact structure of TB-EPS can prevent the separation of sludge and water.

The main substances in sludge EPS are proteins and polysaccharides. As shown in Figure 4(b), the protein in S-EPS of electro-oxidation treated sludge was highest, followed by that of raw sludge, and electro-flocculation treated sludge was the lowest. Nevertheless, the difference among the three samples was unobvious. The protein content in LB-EPS of raw sludge and electro-oxidation treatment sludge were close to each other, while that of the electro-flocculation treated sludge was the lowest, with a value of 181.9 mg/L. It should be noted that the protein content in TB-EPS was significantly different, with values of 532.9 mg/L and 272.9 mg/L corresponding to raw sludge and electro-oxidized sludge, which decreased by nearly 48.8% compared to raw sludge. The above results showed that under the oxidation of EZVI-PMS, the proteins in sludge TB-EPS were not only transformed into soluble proteins, but also converted into non-protein substances. In the presence of the polysaccharide in S-, LB- and TB- EPS of different sludge samples (as shown in Figure 4(c)), it presented a similar changing tendency to protein, but there is a big difference in the value of concentrations. The highest polysaccharide content in S-EPS appearance in electro-oxidation conditioned sludge, with a value of 130.3 mg/L, was more than 2.0 times that of raw sludge and electric flocculation sludge. The polysaccharide contents in LB-EPS were pretty close, with values of 65.2, 58.1 and 77.8 mg/L corresponding to raw sludge, electro-flocculation sludge and electro-oxidation sludge. The polysaccharides in TB-EPS of raw sludge reached 138.3 mg/L, which was the maximum and very close to that of the electro-flocculation conditioned sludge, and much higher than 95.1 mg/L for the electro-oxidation treatment sludge. The results showed that electro-oxidation could transform the adhesive polysaccharides into soluble polysaccharides.

The results indicated that protein-like substances might be mineralized by EZVI-PMS, while polysaccharides were not, when comprehensively analyzing proteins and polysaccharides. Besides, polysaccharides and proteins in S-EPS did not affect sludge dewatering performance. It can be speculated that protein substances in TB-EPS are the dominant substances limiting sludge dewatering efficiency.

3.4. Analysis of EPS by 3D-EEM

Figure 5 shows the 3D-EEM spectra of SB-EPS, LB-EPS and TB-EPS extracted from raw sludge, electro-flocculation and electro-oxidized sludge. According to the fluorescence spectra of the three kinds of EPS and the fluorescence substances at different Ex/Em regions (as shown in Table 1), the main fluorescence peaks were A (Ex/Em: 230/306), B (Ex/Em: 275/306), C (Ex/Em: 280/346), and D (Ex/Em: 325–350/438), representing aromatic family proteins (Zhu *et al.* 2015), soluble microbial byproducts (Bourven *et al.* 2012), tryptophan protein substances (Pang *et al.* 2014), and humic acids (Wang *et al.* 2009), respectively. In the 3D-EEM diagram of the raw sludge S-EPS, C peak was not obvious, and the sludge treated with electro-flocculation did not show a peak at this position, while peak B in the EEM diagram of the sludge SB-EPS treated with electro-oxidation was more significant than that of the raw sludge. For LB-EPS and TB-EPS, peak C of raw sludge and electro-flocculation treatment sludge was significant, while peak C of electro-oxidation treatment sludge became relatively insignificant. For SB-EPS and TB-EPS, peak D changed negligibly. It can be inferred from the above results that protein

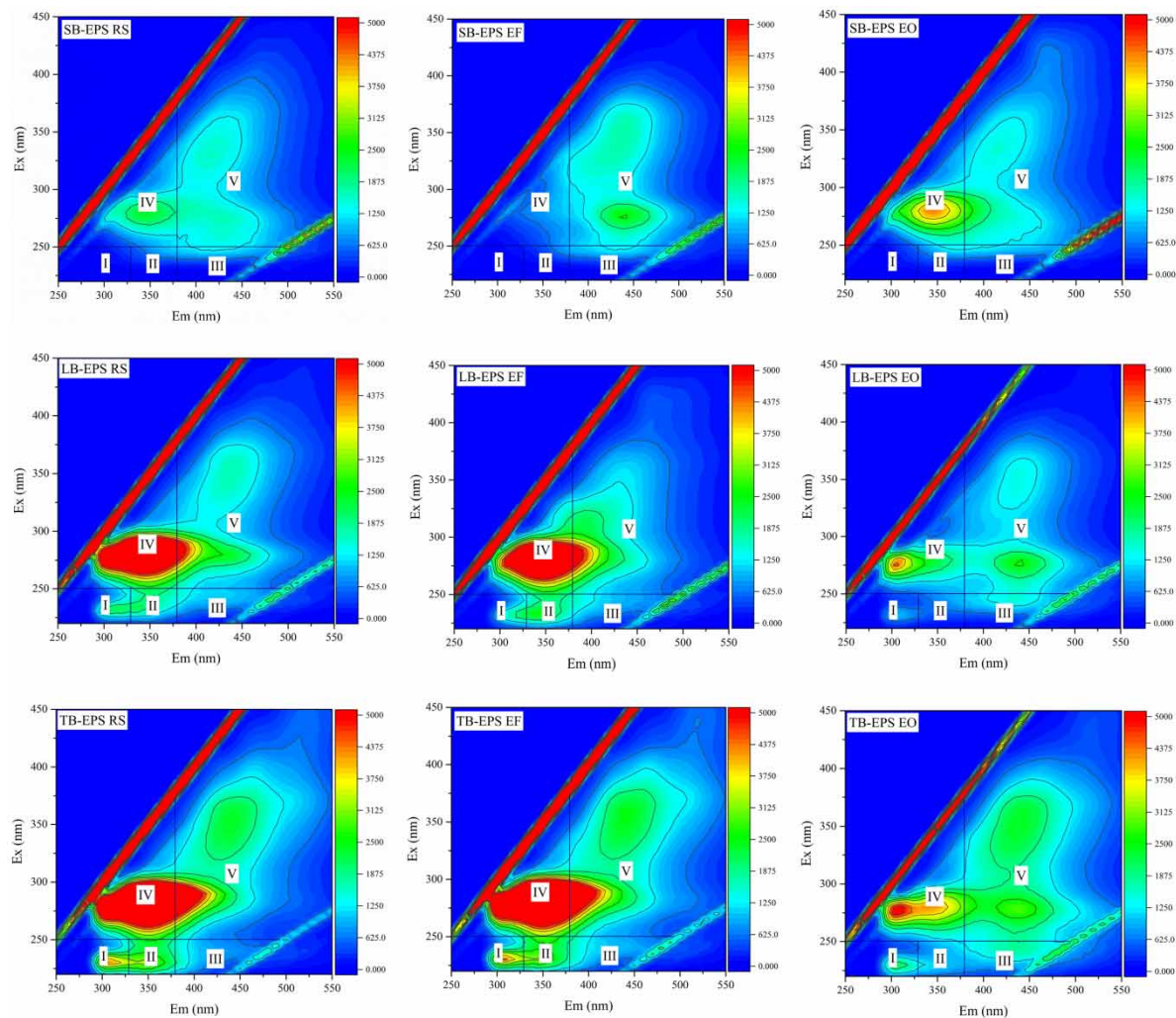


Figure 5 | 3D-EEM spectra of SB-EPS, LB-EPS and TB-EPS of sludge treated under 0.2 A current and 15 min condition time with 130 mg/gDS PMS.

Table 1 | The fluorescence substances of different Ex/Em regions

| Region | Ex/Em | fluorescence substances |
|--------|-----------------|------------------------------------------------------------------------------------------------------------------------------|
| I | 200–250/300–330 | Aromatic protein I (Tyrosine) |
| II | 200–250/330–380 | Aromatic protein II (Tryptophan) |
| III | 200–250/380–500 | Fulvic acid-like |
| IV | 250–400/300–380 | Soluble microbial by-product-like (Protein-like containing tryptophan, tryptophan and protein-like related to biological) |
| V | 250–400/380–500 | Humic acid-like |

substances in sludge TB-EPS was oxidized by EZVI-PMS and transferred to SB-EPS, while humic acid sludge was decomposed negligibly. Moreover, it was found through 3D-EEM spectra that the variation trend of protein contents in SB-EPS, LB-EPS and TB-EPS was consistent with the variation trend of directly detected protein contents. It could be confirmed again that there was a significant correlation between proteins in TB-EPS and sludge dewatering performance.

When comprehensively analysing the contents of TOC, polysaccharide and protein in EPSs, electro-oxidation can disintegrate the proteins and polysaccharides in TB-EPS into dissolved homogeneous substances, or transform them into other kinds of organic substances, especially the proteins into non-protein substances under the oxidation of electro-oxidation. By analyzing the changing tendency of EPS content and substance types in sludge, it was found that higher TB-EPS corresponded to higher CST value, which was similar to other research results. With the cracking of TB-EPS, part of the bound water was released, and the separation effect of sludge and water was improved. The decrease of tryptophan – protein content in sludge TB-EPS was also positively correlated with the decrease of CST. In addition, the increase of ammonia nitrogen in the electro-oxidized sludge supernatant may mainly be derived from the fracture of some protein substances in TB-EPS. Moreover, the protein content was much higher than the polysaccharide content in the three sludge EPS samples, which indicated that protein content had a more significant influence on sludge dewatering performance to a certain extent. In a nutshell, TB-EPS in sludge was the main factor that restricted sludge dewatering.

3.5. The mechanism of electro-oxidation on sludge

The sludge particle size distribution is shown in Figure 6(a). It can be found that the average particle size increased slightly for electro-flocculation and electro-oxidation sludge, which were bigger than that of raw sludge with a value of 56 μm . The average particle size of conditioned sludge samples was about 64 μm ; however, it should be noted that the sludge conditioned by EZVI-PMS accounted for a larger proportion at the position of average size, while the electro-flocculation sludge had a larger particle size range. The particle size distribution of electro-oxidation treated sludge was consistent with the change trend of

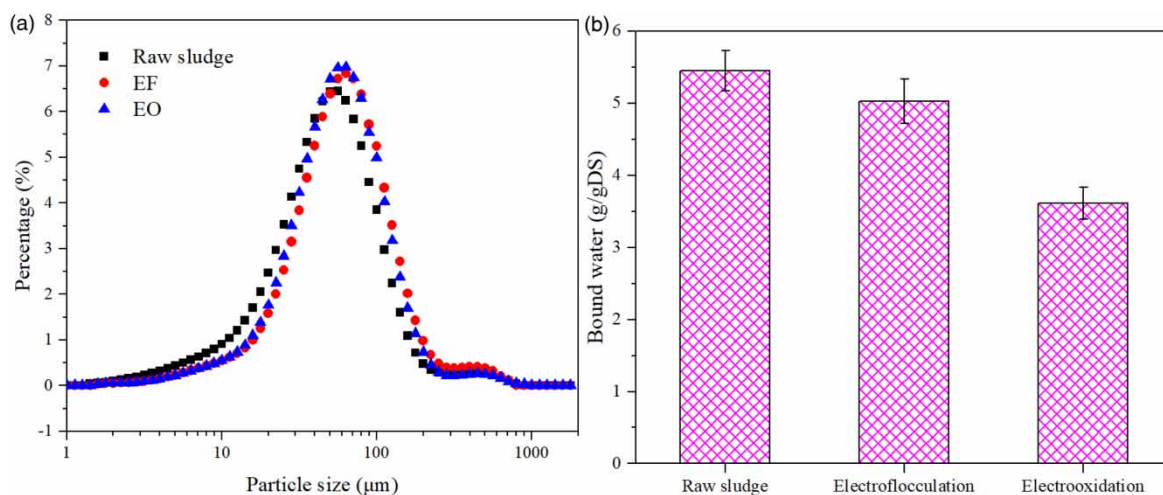
**Figure 6** | Particle size distribution (a) and changes of sludge bound water (b).

Table 2 | Cost analyses of EZVI-PMS

| Methods | CST | | SRF | | Oxidant dosage/consumption | Cost (USD/tDS) |
|------------|---------------|--------------------|-----------------------------------------------|--------------------|--------------------------------------------------------------------------------|----------------|
| | Raw sludge(s) | Reduction rate (%) | Raw sludge ($\times 10^9$ s ² /g) | Reduction rate (%) | | |
| Fe(II)-PMS | 201.8 | 90.1 | 9.52 | 97 | Fe(II) 0.81 mmol/gVSS HSO ₅ ⁻ 0.9 mmol/gVSS | 336.2 |
| EZVI-PMS | 195.5 | 43.8 | 1.47 | 74.1 | KHSO ₅ 0.0611 g/gDS ZVI 0.004 g/gDS Electricity 0.045 W·h/gDS | 213.6 |

sludge treated using activated persulfate by other researchers (Xiao *et al.* 2017). For sludge treated by electro-flocculation, the particle size increased because ferrous ions and iron ions generated in the electrolysis process could produce a flocculation effect. However, for the sludge conditioned by electro-oxidation, although the sludge was disintegrated and the cells were destroyed, the electronegativity of the sludge surface is reduced due to the reduction of TB-EPS, so the sludge can be more easily aggregated, which leads to the increase of sludge particle size (Mikkelsen & Keiding 2002).

The enhancement of sludge dewaterability may be attributed to an oxidation step by SO₄^{·-} through degrading the EPS and sludge cells, and converting the bound water to free water and a subsequent re-coagulation step by the generation of Fe(III). As shown in Figure 6(b), the bound water content of raw sludge, electro-flocculation sludge and electro-oxidation sludge was 5.4 g/gDS, 5.0 g/gDS and 3.6 g/gDS, respectively. The results showed that the bound water of electro-oxidation conditioned sludge decreased significantly, which was mainly due to the cracking of EPS and cell rupture, which played a promoting role in improving sludge dewaterability.

Comprehensive analysis indicated that the improvement of sludge filtration performance and the disintegration effect of sludge was mainly due to the oxidation of EZVI-PMS to destroy extracellular polymers and sludge cells, so that the sludge was disintegrated and released bound water simultaneously. In the process of improving sludge filtration performance, Fe(III) ions in the electrolysis process can also produce a flocculation effect (Gao *et al.* 2021), which made sludge re-aggregate into sludge flocs with larger particle size after being destroyed. Meanwhile, the change of protein types in sludge EPS improved the sludge-water separation effect to a certain extent.

3.6. Economic analyses of EZVI-PMS

The total cost of EZVI-PMS was 213.6 USD/t DS including the consumption of PMS, ZVI and electricity (as shown in Table 2). Though EZVI-PMS exhibits a lower sludge dewatering efficiency than Fe(II)-PMS (Liu *et al.* 2016), the cost is much lower than using Fe(II)-PMS to condition sludge. So ZVI-PMS still has a great potential to be the effective alternative for sludge dewatering.

4. CONCLUSION

The effects and mechanism of electrolytic zero-valent iron activated peroxymonosulfate on enhancing sludge disintegrating and dewatering performance were investigated in this study. Sludge dewaterability was significantly improved in term of CST and SRF, with significant reductions. Besides, COD in the supernate increased substantially, while DNA content declined observably, which were highly correlated with sludge disintegration. Simultaneously, electrolytic zero-valent iron activated peroxymonosulfate could break down the tightly bound extracellular polymers into dissolved extracellular polymers in sludge, change the protein substance types in sludge significantly, and convert quite a few proteins into non-protein substances.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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