

Improvement of phosphorus release from sludge by combined electrochemical-EDTA treatment

Yunfeng Xu, Fanglu Hu, Xin Wang, Yangwei Qu, Lu Xu and Qiang Liu

ABSTRACT

In this paper, combined with the addition of ethylenediaminetetraacetic acid (EDTA), the electrochemical treatment of waste activated sludge (WAS) was investigated to explore its effect on the release of phosphorus (P) from WAS. The results showed that during the electrochemical treatment, the addition of EDTA could significantly promote the release of P from the WAS to the supernatant, the optimal amount of EDTA was 0.4 g/g total suspended solids (TSS), when the release of total dissolved phosphorus (TDP), organic phosphorus (OP) and molybdate reactive phosphorus ($\text{PO}_4^{3-}\text{-P}$) were 187.30, 173.84 and 13.46 mg/L, respectively. OP was the most likely form of P to be released during this process. Moreover, combined electrochemical-EDTA treatment could promote the release of P and metal ions from extracellular polymeric substances (EPSs) to the supernatant, and increase the solubility and disintegration of sludge. EDTA chelated the metal ions of sludge flocs and phosphate precipitates to cause sludge floc decomposition, thereby promoting the release of P from WAS.

Key words | electrochemical-EDTA treatment, extracellular polymeric substances, metal ions, organophosphorus, phosphorus release, waste activated sludge

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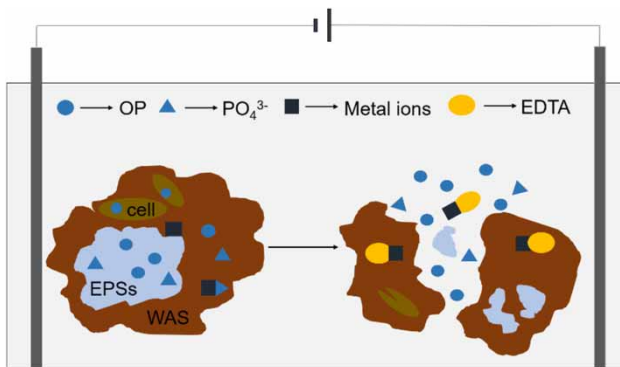
HIGHLIGHTS

- Successfully developed combined electrochemical-EDTA treatment technology to promote the release of P in WAS.
- OP was the most likely phosphorus form to be released.
- Changes of P and metal elements content in EPSs had been investigated.
- EDTA destroys sludge flocs and promotes P release by chelating heavy metals.

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GRAPHICAL ABSTRACT



INTRODUCTION

Phosphorus (P) is an essential element for all living organisms. According to statistics, there are about 70 billion tons of phosphate rock available for mining worldwide. However, phosphate rock is a non-renewable resource; phosphate rock in the world is only enough for humans to continue mining for 50–100 years, currently (Cordell *et al.* 2009; Rittmann *et al.* 2011). Therefore, many scholars are devoted to researching efficient and sustainable phosphorus recovery technologies. Waste activated sludge (WAS) is a by-product of a wastewater treatment plant (WWTP), 90% of P in municipal sewage enters WAS after treatment (Bloecher *et al.* 2012). According to statistics, China produces more than 40 million tons of WAS per year (Tou *et al.* 2017), with the development of industrialization and urbanization, its output continues to increase. Thus, WAS is considered as a new P source material that can replace phosphate rock. In addition, the loss of a large amount of organic substances, nitrogen, phosphorus and other pollutants of WAS will cause water environment pollution (Wang *et al.* 2016; Zhao *et al.* 2018). Therefore, the recycle of P from WAS cannot only reduce the pollution of the receiving waters, but also realize the reuse of P.

The recovery technology of P from sludge mainly produce a phosphorus-rich supernatant (Xu *et al.* 2015), and the generation of phosphorus precipitation is considered to be the best way to recover P from the phosphorus-rich solution (Zou *et al.* 2017). There are some reported literatures using physical (e.g. freezing/thawing, ultrasonic), chemical (e.g. surfactant addition, acid/alkali treatment) or biological (e.g. thermophilic *Geobacillus sp G1*) method to promote the release of P in sludge (Pilli *et al.* 2015; He

et al. 2016b; Wang *et al.* 2017; Xu *et al.* 2018; Zhu *et al.* 2018). In the common methods, P was recovered from WAS by adjusting pH, the results showed that the total phosphorus (TP) release percentages were 36.2% and 12.4% at pH = 2.0 and pH = 11.0, respectively (Xu *et al.* 2015). Fang indicated that P was released, as OH⁻ replaced PO₄³⁻ in Fe-P under an alkaline condition (Fang *et al.* 2018). However, these methods will lead to high cost and secondary pollution. In addition, highly hydrophilic extracellular polymeric substance (EPSs) in WAS usually accumulate in the form of highly stable and viscous colloidal flocs with negative charge (Zhang *et al.* 2016), it is difficult to release P directly from WAS. Very recently, electrochemical pretreatment (EP) has been widely used for its advantages such as high efficiency, cleanliness, flexibility, and environmental friendliness (Chiavola *et al.* 2015; Hasan *et al.* 2016; Ye *et al.* 2016). The principle of EP depends on ohmic heating, electrophoresis and electroosmosis (Zhen *et al.* 2014). It was reported that EP had a significant effect on sludge solubilization, the removal rates of soluble chemical oxygen demand (SCOD), total suspended solids (TSS) and volatile suspended solids (VSS) in sludge supernatant after EP were significant increased (Song *et al.* 2010; Yu *et al.* 2014; Ye *et al.* 2016). During EP process, the high polymers in WAS were converted to low-molecular-weight products and then easily degraded, and the microbial cells were completely ruptured and decomposed (Yu *et al.* 2014; Dereszewska *et al.* 2015). And under the optimal condition, the highest concentrations of PO₄³⁻-P and OP released by WAS reached 5.020 mg/L and 1.888 mg/L, respectively, which were 2.86 and 4.93 times higher than those of raw sludge (Yang *et al.* 2020). In

addition, EP significantly increased the release of P from WAS during anaerobic fermentation (AF) (Xu *et al.* 2020).

However, due to the presence of metal ions such as Ca^{2+} , Mg^{2+} , Fe^{3+} and Al^{3+} in the supernatant, it is possible to form a precipitate with the free phosphate (Zou *et al.* 2017), so that the P content in the supernatant would not continuously increase. FePs and AlPs are the main components of inorganic phosphorus (IP), and the release of P in these two insoluble precipitates is difficult. Studies have shown that adding surfactants which can complex with metal ions was beneficial to reduce the precipitation of P and metal ions in the supernatant (Mulligan 2009). Surfactants could also effectively destroy the polymer matrix of sludge, and promoted the release of extracellular polymers in WAS flocs (Sheng *et al.* 2010; Zhou *et al.* 2013; He *et al.* 2016b). Ethylenediaminetetraacetic acid (EDTA) is a typical metal ion chelating agent that can stably bind to metal ions (Liu & Lin 2013). Studies have reported that EDTA could significantly improve the release of polyphosphates from sewage sludge during thermal pretreatment (Zhang & Kuba 2014). And EDTA could bind to metal cations in the extracellular polymer of sludge and on the surface of cell membranes (Kavitha *et al.* 2013), resulting in sludge decomposition and intracellular material release. At the same time, adding EDTA was an effective method to release the soluble phosphorus in FePs and AlPs (Zhang *et al.* 2010; Ping *et al.* 2020). It can be seen that the full release of P in sludge is facilitated by the addition of EDTA.

This article used combined electrochemical-EDTA treatment, in order to promote the release of P in WAS. Under the optimal conditions of electrochemical treatment (voltage of 4.5 V and time of 60 min) (Yang *et al.* 2020), different doses of EDTA were added. The amount of P released in different forms was investigated to determine the optimal amount of EDTA added. And the solubility and disintegration of WAS were determined by SCOD, TSS and VSS. EPSs is the main component of the WAS flocculation matrix, and P has significant accumulation in EPSs (Cloete & Oosthuizen 2001; Zhang *et al.* 2013). Thus we also discussed the content changes of P and metals contents in EPSs.

MATERIALS AND METHODS

Sludge sample and characterization

The WAS used in this experiment was taken from the secondary sedimentation tank of a sewage treatment plant in Shanghai. Before the test, the sludge was filtered by a

Table 1 | Basic properties of the sludge

Parameter	Mean	Parameter	Mean
pH	6.86 ± 0.21	TP	490.82 ± 20.68 mg/L
TCOD	13.25 ± 0.20 g/L	SCOD	1.51 ± 0.11 g/L
TSS	17.03 ± 0.38 g/L	VSS	10.91 ± 0.30 g/L
Al^{3+}	89.38 ± 4.35 mg/L	Ca^{2+}	72.08 ± 3.40 mg/L
Fe^{3+}	14.01 ± 2.90 mg/L	Mg^{2+}	26.84 ± 1.40 mg/L

1 × 1 mm screen to remove large particles such as sand and gravel in WAS. Then the sludge was left to stand for 24 h, after the supernatant was discarded, the remaining activated sludge was stored in a refrigerator at 4 °C for later use. The main characteristics of the sludge are shown in Table 1.

Experimental setup and procedures

The electrolytic cell device used in this experiment was 10 × 5.5 × 11 cm (L × W × H), and the electrode plates used for the cathode and anode were 5 × 13 cm Ti/RuO₂ mesh electrodes. In order to prevent the sludge from sinking during the experiment and causing the reaction to be incomplete, this experiment used a magnetic stirring method, and placed the electrolytic cell on a magnetic stirrer. 300 mL of WAS was taken into the electrolytic cell, set up five experimental groups, added 0.0, 0.2, 0.4, 0.6, 0.8 g/g TSS EDTA, and set the voltage to 4.5 V, the electrode spacing was 3 cm, and processed for 60 min (Yang *et al.* 2020). All the experiments were performed in triplicate.

Analytical methods

The treated sludge was centrifuged (3,000 r/min, 10 min), the supernatant was taken through a 0.45 μm membrane, and various indicators in the supernatant and solid phase were measured. The measurement of pH, TSS, VSS, SCOD and TCOD were the same as previous publication (Xu *et al.* 2015; He *et al.* 2016a). The amounts of total dissolved phosphorus (TDP) and molybdate reactive phosphorus (free $\text{PO}_4^{3-}\text{-P}$) in the liquid were monitored following the standard method of ammonium molybdate spectrophotometry (GB11893-89, China), and organophosphorus (OP) was calculated from the difference between TDP and $\text{PO}_4^{3-}\text{-P}$ (Juston & DeBusk 2011). The metal ions (Al^{3+} , Fe^{3+} , Mg^{2+} and Ca^{2+}) in the sample were extracted with concentrated HNO_3 , and the metal ion contents were determined by inductively coupled plasma emission spectroscopy (Prodigy, Liman Instrument Manufacturing Co., Ltd, USA). In this work, the EPSs were

extracted by an improved thermal extraction method, the specific method can be found in a previous publication (Li & Yang 2007). And data was analyzed by Origin.

The contributions of OP and PO_4^{3-} released from solid to liquid were determined by Equations (1) and (2), respectively (Xu et al. 2015).

$$\begin{aligned} &\text{Released OP contribution (\%)} \\ &= \frac{OP(c, \text{liquid}) - OP(0, \text{liquid})}{OP(c, \text{liquid})} * 100\% \end{aligned} \quad (1)$$

$$\begin{aligned} &\text{Released } PO_4^{3-} \text{ contribution (\%)} \\ &= \frac{PO_4^{3-}(c, \text{liquid}) - PO_4^{3-}(0, \text{liquid})}{PO_4^{3-}(c, \text{liquid})} * 100 \end{aligned} \quad (2)$$

where OP (c, liquid) (mg/L) and PO_4^{3-} (c, liquid) (mg/L) were the contents of OP and PO_4^{3-} in the supernatant after treatment, respectively. OP (0, liquid) (mg/L) and PO_4^{3-} (0, liquid) (mg/L) were the OP and PO_4^{3-} contents of the supernatant in the raw sludge without treatment.

RESULTS AND DISCUSSION

Effect of combined electrochemical-EDTA treatment on phosphorus in sludge

Effect of EDTA dosages on phosphorus release from sludge

In order to verify that the combined electrochemical-EDTA treatment can promote the release of P from sludge, the sludge was treated with electrochemical treatment and combined electrochemical-EDTA treatment, the indicators of the raw sludge and treated sludges were compared. At this time, the reaction conditions were as follows: voltage was 5 V, time was 90 min, EDTA dosage was 0.4 g/g TSS. The results are shown in Table 2.

The release of TDP from the sludge treated with separate electrochemical treatment was 6.91 mg/L, the release of PO_4^{3-} -P was 5.02 mg/L, and the release of OP was

1.89 mg/L, which were 2.34, 2.00 and 4.30 times than those of the raw sludge, it showed that the electrochemical treatment can indeed increase the P released from sludge. In addition, a large amount of metal ions were released into the liquid phase after electrochemical treatment, which also confirmed the release of P. Furthermore, by combined Electrochemical-EDTA treatment, the amounts of TDP, OP and PO_4^{3-} -P released into the supernatant were 194.40 mg/L, 26.73 mg/L and 167.67 mg/L, which were 65.90, 10.65 and 381.07 times those of the raw sludge, respectively. The TDP release rate of WAS reached 39.6%, which was slightly higher than that of TDP released by acid method (36.2%) (Xu et al. 2015). This meant that during the combined electrochemical-EDTA treatment, the release of P could be significantly increased. And the SCOD value after combined electrochemical-EDTA treatment was the highest, followed by the electrochemical treatment, and the least was the SCOD value of the raw sludge, which indicated that the dissolution of sludge after combined electrochemical-EDTA treatment was the largest. Moreover, the values of TSS and VSS also reached the minimum after combined electrochemical-EDTA treatment indicating that the degree of sludge disintegration has reached the maximum at this time.

After confirming that combined electrochemical-EDTA treatment can promote the release of P from sludge, it was necessary to study the release of different forms of P in different EDTA additions to determine the optimum amount of EDTA. The results are shown in Figure 1.

After adding 0.2, 0.4, 0.6, 0.8 g EDTA/g TSS, respectively, the TDP, PO_4^{3-} -P and OP in the sludge supernatant increased with the addition of EDTA after electrochemical treatment. When the added EDTA reached 0.4 g/g TSS, the released P reached the maximum; since then, with the increase of EDTA, the released P has remained basically unchanged. When 0.4 g/g TSS of EDTA was added, the release of TDP, OP and PO_4^{3-} -P were 187.30, 173.84 and 13.46 mg/L, respectively. Different from the electrochemical treatment alone, the concentration of OP in the sludge supernatant was much higher than the concentration of PO_4^{3-} -P. After adding EDTA, more OP can be released

Table 2 | Various indicators of sludge under different conditions

Processing group	TDP	PO_4^{3-} -P	OP	Ca^{2+}	Mg^{2+}	Fe^{3+}	Al^{3+}	SCOD	TSS	VSS
	(mg/L)							(g/L)		
Raw	2.95	2.51	0.44	40.10	28.30	3.10	25.00	1.58	15.50	8.40
Electrochemical	6.91	5.02	1.89	64.50	30.20	10.70	64.60	2.78	16.18	10.67
Electrochemical-EDTA	194.40	26.73	167.67	65.70	30.60	155.80	29.60	2.88	10.60	4.50

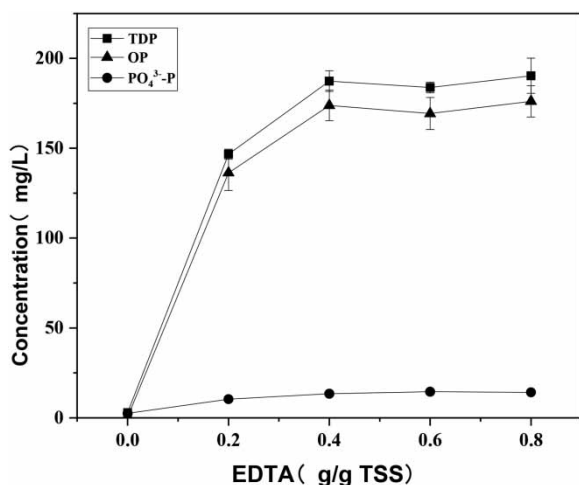


Figure 1 | Concentration of different forms of P in supernatant treated by electrochemical treatment combined with different doses of EDTA.

into the liquid phase. This was because that EDTA can combine with most metal ions to release macromolecular organic substances that were originally linked by metal bridging, which promoted the decomposition of sludge and increased the release of P (Kavitha et al. 2013; He et al. 2016b). In addition, EDTA could kill cells and promoted the release of OP on cell surface (Zou et al. 2017).

In general, with the electrochemical treatment, EDTA could effectively and greatly promote the release of P from the sludge into the liquid phase, and it was less affected by the supernatant metal ions. As the added EDTA increased, the released P increased; when it reached 0.4 g/g TSS, the released P reached a maximum. Different from the separate electrochemical treatment, the amount

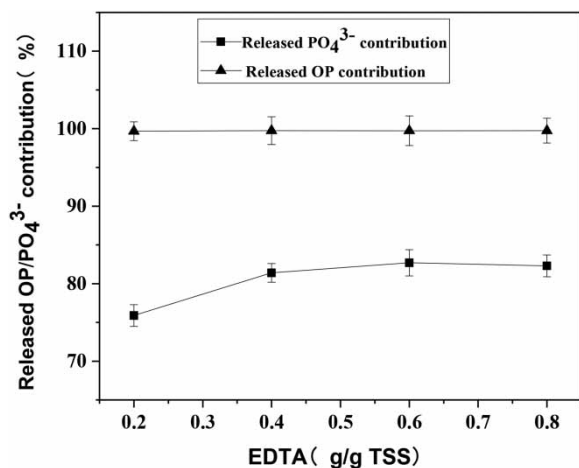


Figure 2 | Effect of the treatment of electrochemical combination of EDTA with different additions on the release contributions of PO₄³⁻ and OP.

of OP released from the sludge by adding EDTA was much larger than that of PO₄³⁻-P.

Effect on release contributions of OP and PO₄³⁻-P

Phosphate precipitation is considered to be the best way to recover P from phosphorus-rich solutions (Zou et al. 2017). Previous studies have shown that OP in solution can be converted to PO₄³⁻-P under the action of microorganisms (He et al. 2016a, 2016b). Therefore, it is necessary to study the soluble PO₄³⁻-P and OP of the sludge, and explore their contributions to the release during the electrochemical process. The release contributions of PO₄³⁻-P and OP from solid to liquid are shown in Figure 2.

As can be seen from Figure 2, with the amount of EDTA added increased, the release contribution of PO₄³⁻ and OP increased, and the released contribution of OP reached 99.68% when the additional amount was 0.2 g/g TSS. And it would keep this level as the amount of EDTA increased. The released PO₄³⁻ was 75.9% at 0.2 g/g TSS, and when the additional amount was 0.4 g/g TSS, it reached 81.4%, and then remained at about 82%. The addition of EDTA could make the PO₄³⁻ and OP in the sludge get better releasing, and the release contributions reached a higher level than that of electrochemical treatment. This was mainly because EDTA would chelate heavy metals on EPSs and cell membrane, thus destroy sludge flocs and cell membrane, and promote the release of OP (Ping et al. 2020). Moreover, EDTA had strong complexation ability and would compete for heavy metals in insoluble metal phosphates, such as Fe and Al (Zou et al. 2017; Ping et al. 2020).

Effect on phosphorus components in sludge solid phase

The results of different forms of phosphorus in the sludge solid phase after different dosages of EDTA combined with electrochemical treatment are shown in Figure 3.

It can be seen from Figure 3 that the component of non-apatite inorganic phosphorus (NAIP) and OP in the solid phase accounted for more than 80% of TP, so there was more releasable P in the sludge solid phase. The proportion of NAIP was the largest of the four different phosphorus forms, which remained around 65%, indicating that the amount of released NAIP was less in this process. The proportion of OP in the solid phase of sludge without EDTA was 15.85%. With the increase of EDTA, the proportion of OP in the solid phase of sludge decreased gradually, which reduced to 12%. It indicated that OP was released into the supernatant after combined electrochemical-EDTA

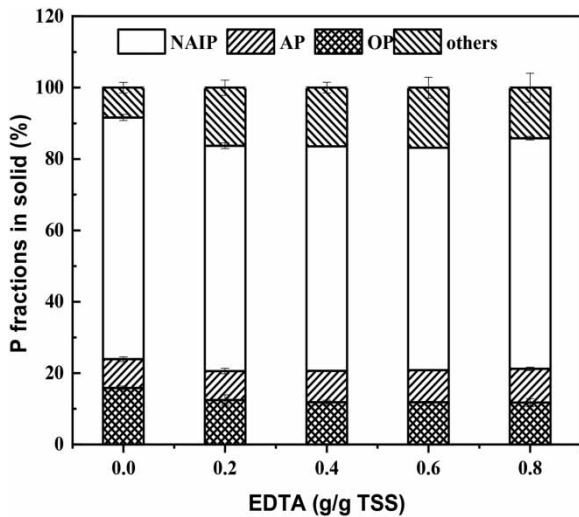


Figure 3 | Proportion of different forms of P in solid phase of sludge after different additions of EDTA combined with electrochemical treatment.

treatment, thereby reducing the content of OP in the solid phase. In addition, the presence of microorganisms in the activated sludge might also convert the OP into $\text{PO}_4^{3-}\text{-P}$ (He *et al.* 2016a, 2016b), and $\text{PO}_4^{3-}\text{-P}$ would be released to the supernatant. The flocculation structure of WAS after combined electrochemical-EDTA treatment would be destroyed by strongly chelating heavy metals (Zou *et al.* 2017; Ping *et al.* 2020).

Generally, four different forms of phosphorus in the solid phase exhibited different releasability during combined electrochemical-EDTA treatment. OP was most likely to be released into the liquid phase as a kind of phosphorus, which was consistent with the value of OP in the liquid phase above. On the contrary to the EP, the amount of OP released by combined electrochemical-EDTA treatment was much larger than that of $\text{PO}_4^{3-}\text{-P}$ (Yang *et al.* 2020).

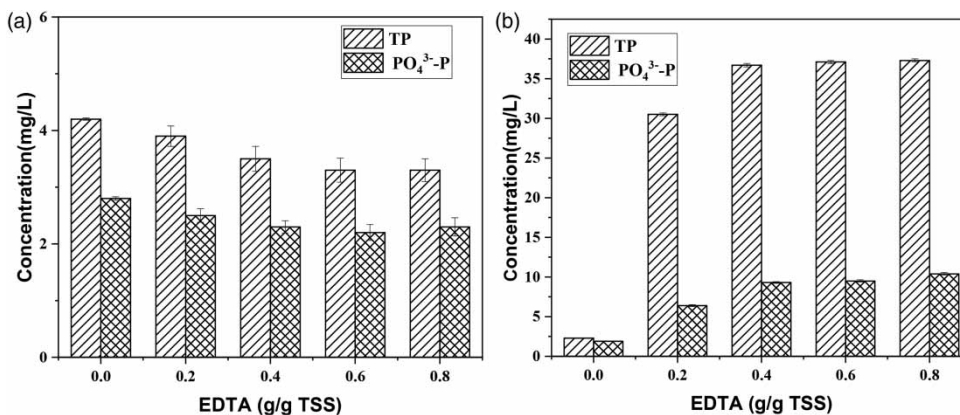


Figure 4 | Changes of P fractions and species in TB-EPSS and LB-EPSS.

Effect of combined electrochemical-EDTA treatment on EPSS

Changes of phosphorus components in EPSS

In order to investigate the effect of EDTA treatment on the release of phosphorus from the EPSSs of sludge, this paper studied the content of different forms of phosphorus in the EPSSs of sludge treated by electrochemical treatment combined with different amounts of EDTA, as shown in Figure 4.

It can be seen from Figure 4 that in the tightly bound extracellular polymeric substances (TB-EPSSs) and loosely bound extracellular polymeric substances (LB-EPSSs), the contents of different forms of P present different trends. The content of P in TB-EPSSs decreased with the increasing EDTA addition. This was because under the combined treatment of electrochemistry and EDTA, the floc structure of the sludge was destroyed and some EPSSs were also destroyed (Zhou *et al.* 2013; Yang *et al.* 2015), so the P in the TB-EPSSs was released.

Unlike the case of TB-EPSSs, the P content in LB-EPSSs showed an increasing trend with the increase of EDTA, which was the same as previous study which used rhamnolipid to promote the release of P from WAS (He *et al.* 2016b). This was due to the loose structure and good rheological property of LB-EPSSs outside the extracellular polymer structure, which received the P released from TB-EPSSs, and thus the concentration of P increased (Zhou *et al.* 2013; Yang *et al.* 2015).

Changes of metal content in EPSSs

Ca^{2+} , Mg^{2+} , Al^{3+} and Fe^{3+} are the main metal ions in the sludge that affect P release, the changes of these metal

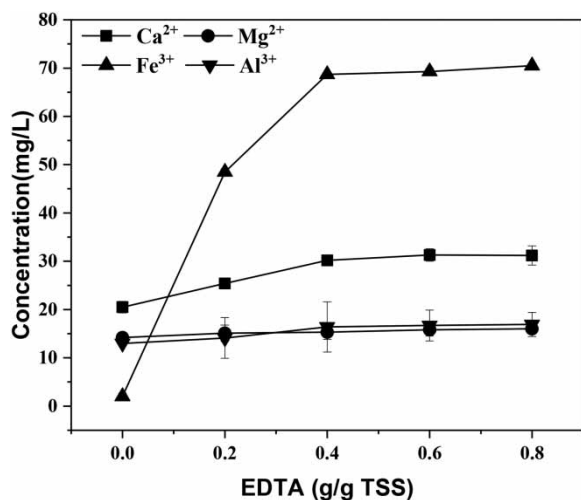


Figure 5 | Changes of metal content in EPSs when electrochemical treatment combined with different amounts of EDTA.

content in EPSs and sludge supernatant at different EDTA additions were studied. The results are shown in Figures 5 and 6.

The concentration of metals in the sludge supernatant and EPSs after combined electrochemical-EDTA treatment increased greatly, and the optimal amount of EDTA added was 0.4 g/g TSS. Among them, the release of Fe³⁺ was much more than that of Ca²⁺, Mg²⁺ and Al³⁺. This was because Fe-EDTA had a greater stability constant than Al-EDTA, Ca-EDTA, and Mg-EDTA, so Fe³⁺ was preferentially complexed by EDTA (Zou et al. 2017). Studies have shown that metal ions such as Ca²⁺ and Mg²⁺ could promote the stability of sludge and EPSs matrix structure (Sheng et al.

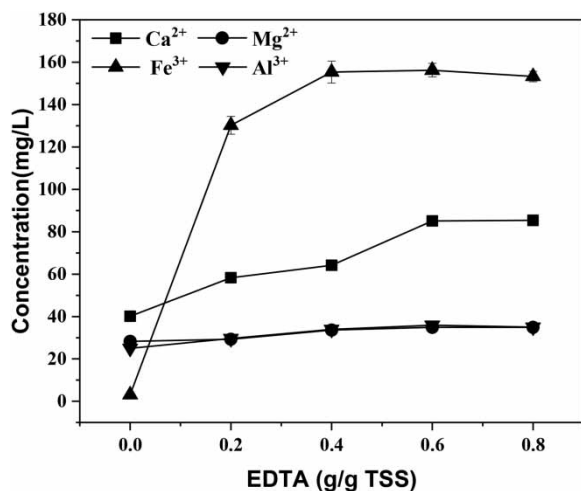


Figure 6 | Changes of metal content in sludge supernatants when electrochemical treatment combined with different amounts of EDTA.

2010; Yan et al. 2015). Therefore, the release of metal might destroy the stability of the matrix structure of the sludge and extracellular polymer, which also contributed to the solubilization of sludge.

Figures 5 and 6 show the similar trends, and the metal contents in the sludge supernatant were higher than that in EPSs. With the increase of EDTA, more microbial cells and extracellular polymers in sludge were destroyed, and metal ions such as Ca²⁺, Mg²⁺, Al³⁺ and Fe³⁺ were released. At the same time, as a chelating agent, EDTA could also interact with the metal ions in the precipitate combine to form a metal chelate (Mulligan 2009). The chelates were mainly distributed in the liquid phase, and part of them were adsorbed in EPSs, thus the metal contents in both the supernatant and EPSs increased.

Effect of combined electrochemical-EDTA treatment on solubility and disintegration of sludge

The SCOD of sludge increased with the increase of EDTA (Figure 7), which indicated that the combination could effectively promote the release of organics of WAS. Electrochemical treatment would decompose sludge flocs and destroy cells (Song et al. 2010; Yu et al. 2014; Ye et al. 2016), electrochemical treatment also could swell the colloidal structure of sludge, and most complex organic compounds were degraded into monomers (Zhen et al. 2014). Meanwhile EDTA could break the bond bridge connection between organics of EPSs and cells by chelating metal ions, thus destroy the floc structure of sludge (Kavitha et al. 2013). Moreover, adding EDTA would increase the

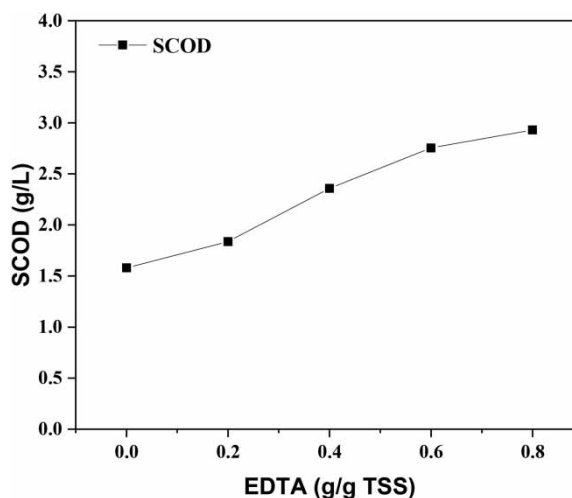


Figure 7 | SCOD of the sludge after electrochemical treatment combine with different EDTA additions.

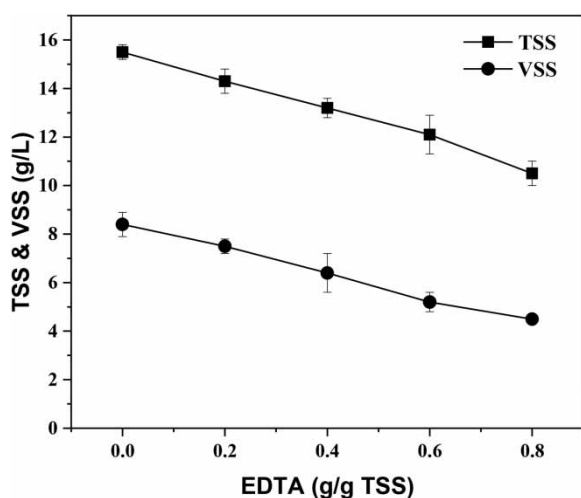


Figure 8 | TSS and VSS of the sludge after electrochemical treatment combine with different EDTA additions.

percentage of dead cells, the main components of cells (proteins, polysaccharides and other organic substances) would be released (Zhen *et al.* 2014; Zou *et al.* 2017). Thus combined electrochemical-EDTA treatment resulted the concentration of SCOD increased.

TSS and VSS are the basic indicators for measuring sludge degradation and decrement. After the combined treatment, the TSS and VSS of the sludge decreased with the increase of EDTA added (Figure 8). EDTA could not only destroy EPSs, but also dissolve the insoluble calcium, magnesium and iron salts (Ping *et al.* 2020). Therefore, the declining trend of TSS after EDTA treatment was obvious. Meanwhile, on the basis of EDTA treatment, the further use of electrochemical treatment effectively promoted the destruction of sludge cells, so that a large amount of substances in the cells were released. Generally, the solubility and disintegration of sludge increased after combined treatment.

CONCLUSIONS

In this paper, the feasibility of combined electrochemical-EDTA treatment to promote the release of P in WAS was studied, the effect of EDTA on the release of P and its mechanism were explored, and the changes of P and metal content in EPSs during this process were also discussed. The results showed that EDTA could significantly promote the release of P and heavy metals of WAS, with the increase of EDTA dosage, the release of P increased. And the optimal addition of EDTA was 0.4 g/g TSS, at this time, the different

forms of P have been well released, and OP was the most likely form of P to be released from WAS. In addition, electrochemical-EDTA treatment can promote the release of P and metal ions from EPSs to the supernatant, and improve the solubility and disintegration of sludge. EDTA can destroy sludge flocs and cells through chelation, and compete for heavy metals with insoluble phosphate, thus promoting the release of P in WAS. Although electrochemical-EDTA treatment has not achieved the lowest cost, it still has good application value to recover P from WAS and remove heavy metals.

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DATA AVAILABILITY STATEMENT

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

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