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Alum sludge conditioning with ferrous iron/peroxymonosulfate oxidation: Characterization and mechanism

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Abstract−Alum sludge produced by drinking water plants needs to be conditioned and dewatered before final disposal. In this study, a novel ferrous iron/peroxymonosulfate (PMS) oxidation process was employed to enhance alum sludge dewaterability. The effect of oxidative sulfate radicals generated by Fe^{2+} activated HSO_5^- on alum sludge was studied. The results showed that the optimal conditioning conditions for addition of Fe^{2+} and PMS were 0.5 g/g and 0.1 g/g TSS, respectively. Meanwhile, the capillary suction time (CST) and specific resistance to filtration (SRF) of alum sludge was reduced by 66% and 88%. Also found was that the absolute value of the zeta potential increased and the particle size decreased in alum sludge after Fe^{2+}-PMS conditioning, which indicated that oxidative sulfate radicals destroyed the floc structure of alum sludge and smaller particles were generated. At the same time, the water contained in sludge flocs was released and enhanced sludge dewaterability, while leaching of aluminum ions also characterized decomposition of alum sludge.

Keywords: Water Purification Sludge, Alum Sludge, Dewaterability, Oxidative Conditioning, Ferrous-peroxymonosulfate

INTRODUCTION

Drinking water treatment plants usually use surface water (e.g., river water, lake water) as the water source, which contains suspended solids and colloidal solids and other impurities. The drinking water treatment process includes coagulation, precipitation, filtration and disinfection. Aluminum salts, e.g. aluminum sulphate and polyaluminum chloride, are widely used as coagulant during the treatment process, while large amount of alum sludge is produced. Before the final disposal, the water purification sludge needs to be dewatered to reduce volume by mechanical process [1]. However, due to the difficulty of separating water from sludge, conditioning is a key step to enhance sludge dewaterability. Main conditioning methods include heat treatment [2,3], freezing/thawing [4,5], flocculant [6,7], acid-base process [8-11] and advanced oxidation process [12-15]. Among them, advanced oxidation conditioning is considered to be a significant treatment method that utilizes oxidative free radicals such as hydroxyl radical (·OH) or sulfate radical ($SO_4^{\bullet-}$) to oxidize sludge and enhance sludge dewaterability [13].

The Fenton system uses ferrous ions and hydrogen peroxide to generate hydroxyl radicals (·OH) under acidic conditions, which rapidly oxidize sludge flocs and improve sludge dewaterability. However, ·OH is non-selective, has a short life span, and is susceptible to interference by background materials from solution. In comparison, sulfate radicals ($SO_4^{\bullet-}$) have higher redox potential (2.5-3.1 V) [16], and the application range in solution is much wider than ·OH. Furthermore, negative effects on oxidation process by ·OH quenchers and background materials from water can be avoided [17,18]. Therefore, sulfate radicals can be applied as effective oxidants in sludge conditioning.

$SO_4^{\bullet-}$ is generated by activation of persulfate (peroxydisulfate or peroxymonosulfate) with extra energy (e.g., heat, ultraviolet) or transition metal [19-23]. Compared to peroxydisulfate, peroxymonosulfate is more easily activated by transition metal ions or their oxides at normal temperature and pressure [24,25]. The activation ability of different transition metal ions to peroxymonosulfate is: Co(II)>Ru(III)>Fe(II)>Ce(III)>V(III)>Mn(II)>Fe(III)>Ni(II) [26]. The above activation ability of metal ions was ranked by the significant difference in reactivity with conversion mode substrate 2,4-dichlorophenol. The transient substances formed by the Mn^+/HSO_5^- interaction were determined by quenching of t-butanol (TBA) and ethanol (EthOH). Studies have shown that reactions involving Co(II), Ru(III), Fe(II) [27-29] only produce SO_4^-. The other metal ions can react with HSO_5^- to form other substances that affect the activation ability negatively [30-33]. Among them, Fe(II) is inexpensive, safe and non-toxic, requires lower activation energy and is more suitable as an activator for peroxymonosulfate. The production of $SO_4^{\bullet-}$ by Fe^{2+} activated HSO_5^- can be shown as follows:

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$$Fe^{2+}+HSO_5^- \rightarrow Fe^{3+}+SO_4^{\bullet-}+OH^- \quad (1)$$

$$SO_4^{\bullet-}+Fe^{2+} \rightarrow Fe^{3+}+SO_4^{2-} \quad (2)$$

$$SO_4^{\bullet-}+HSO_5^- \rightarrow SO_5^{\bullet-}+SO_4^{2-}+H^+ \quad (3)$$

$$SO_4^{\bullet-}+SO_4^{\bullet-} \rightarrow S_2O_8^{2-} \quad (4)$$

More recently, some researchers successfully applied $SO_4^{\bullet-}$ to organic sludge conditioning. Liu et al. [34] studied the improvement of waste activated sludge dewaterability by Fe^{2+} activated HSO_5^-, the concentration of HSO_5^- and Fe^{2+} was 0.9 mmol/g and 0.81 mmol/g VSS. The reduction rates of CST and SRF were up to 90% and 97%, respectively. Zhen et al. [35] studied the mechanisms of waste activated sludge conditioning by Fe^{2+} activated persulfate oxidization. It was found that a higher CST reduction rate (88.8%) attained when the addition of persulfate and Fe^{2+} were 1.2 mmol/g and 1.5 mmol/g VSS.

Different from the organic sludge, alum sludge contains aluminum from coagulants which could be destructive to plants without pretreatment [36,37]. At present, the commonly used aluminum removal methods were acidification and vacuum electrokinetic technology [38,39]. However, addition of extra reagents increased the cost of secondary treatment.

Although advanced oxidization process can significantly enhance the dewaterability of biological sludge, such as activated sludge [40] and digestion sludge [41,42], very few studies have been conducted to investigate its effects on inorganic sludge. In this study, Ferrous-PMS was primarily adopted to experimentally evaluate its impact on the dewaterability of alum sludge. The conditioning process was optimized to obtain the optimum efficiency. Meanwhile, characterizations of the sludge flocs during conditioning were analyzed and the mechanisms of the reaction process were investigated.

MATERIAL AND METHODS

1. Sludge Source

The alum sludge used in the study was from a drinking water treatment plants located in Shenzhen, China. The main characteristics of the alum sludge are shown in Table 1.

2. Batch Experiment

The alum sludge produced by coagulation/sedimentation processes from the waterworks was studied. The reduction rates of CST and SRF were used as the main indicators of sludge dewaterability to evaluate the effects of different Fe^{2+} and HSO_5^- addition concentrations and initial pH values on alum sludge dewaterability, The changes of sludge characteristics after conditioning and the mechanisms of enhancement of dewaterability were also investigated. As shown in Table 2, a series of batch tests were operated to study the effect of PMS and Fe^{2+} addition on dewaterability of alum sludge (Table 2). In group (I), concentrations of PMS ranging between 0.05-0.50 g/g TSS was used while the addition of Fe^{2+} was maintained as 0.1 g/g TSS. In contrast, group (II) studied the effect of Fe^{2+} addition ranged between 0.05-0.50 g/g TSS, and the PMS concentration was sat as 0.25 g/g TSS. The addition amount of PMS and Fe^{2+} additions used in this study was based on our previous studies [43,44]. Effects of pH adjustments were also investigated during the tests. Different pH values (original pH=6.4, adjusted pH=4.0, 5.0) were used in this study.

Table 1. Main characteristics of alum sludge

Parameter	Primary sludge
Moisture content (%)	99.23±0.01
Solid content (%)	0.77±0.01
TS (g/L)	7.95±0.07
TSS (g/L)	7.76±0.02
VS (g/L)	5.57±0.06
VSS (g/L)	5.40±0.03
pH	6.40±0.02
COD (mg/L)	3 600±13

Table 2. Experimental design of the batch tests

Group	No.	PMS concentration (g/g TSS)	Fe^{2+} addition (g/g TSS)
I. Effect of PMS concentration	1	0.00	0.10
	2	0.05	0.10
	3	0.10	0.10
	4	0.25	0.10
	5	0.40	0.10
	6	0.50	0.10
II. Effect of Fe^{2+} addition	7	0.25	0.00
	8	0.25	0.05
	9	0.25	0.10
	10	0.25	0.25
	11	0.25	0.40
	12	0.25	0.50

In each batch, 100 mL of alum sludge was transferred into a 250 mL screw bottle. Then different dosages of potassium hydrogen persulfate and ferrous sulfate heptahydrate were added into the bottles. The samples in the bottles were then mixed with a shaker at 150 rpm or 30 min. After that, the treated samples were prepared for the following measurements of CST and SRF (as described in Section 2.3).

3. Analytical Procedure and Data Analysis

In this study, CST and SRF were used as the main indicators of alum sludge dewaterability. The measurement procedure of CST was as follows. Before the measurement, the sludge to be tested was shaken in the bottle thoroughly. A 10 mL pipette with the removal of 1 cm head allowed the sludge sample to be collected. In a pipette about 6 mL of the sludge to be tested was placed in the funnel configured by the CST timer to record the time required for the moisture in the sludge to diffuse from the inner ring probe to the outer ring probe through the filter paper, which was the CST of the sludge sample [45,46]. The specific measurement steps of SRF are as follows: (1) Determine the moisture content of the sludge; (2) Place a wet filter paper with a diameter larger than the funnel in the Buchner funnel. Adjust the vacuum pressure of vacuum pump to about 1/3 less than the experimental pressure, so that the filter paper is close to the funnel wall, and suction filtration for 30 s to ensure that there are no water droplets in the filter paper; (3) Take 50 mL sludge and pour it into the funnel slowly. Keep the height of the poured sludge within the edge of the filter paper. After settling for 1 min, adjust the vacuum pressure to 0.03 Mpa, and record the volume of the filtrate in the cylinder. Then record the corresponding filtrate volume in the metering cylinder every 10 s when starting to filtration. As the filtration speed slows, gradually increase the recording interval for every 1 min until the filter cake is broken, so there is no further filtrate within 1 min; (4) Measure the moisture content of the filter cake after filtration; (5) Plot the straight line and calculate the slope b while using the volume V of the sludge filtrate as the abscissa and t/V as the ordinate [47]. Capillary suction timer (Type 304M, Triton Electronics Ltd.) was used to record the penetration time of the moisture from alum sludge through filter paper. The change of alum sludge dewaterability was assessed via the CST reduction percentage R (%), as shown in Eq. (5):

$$R(\%)=\frac{CST_0-CST_1}{CST_0}\times 100\% \tag{5}$$

where CST_0 represents capillary suction time (s) before sludge conditioning; CST_1 represents capillary suction time (s) after sludge conditioning.

SRF was measured by sludge specific resistance test device (TG-250, Shanghai Tongguang Science and Education Instrument Co., Ltd.). The moisture content of the alum sludge before and after filtration was recorded. Sludge specific resistance was calculated according to Eq. (6):

$$SRF=\frac{2PA^2b}{\mu\omega} \tag{6}$$

where SRF - sludge specific resistance (m/kg); P - experimental pressure (N/m^2); A - filtration area (m^2); b - filtration time ratio (s/m^6), the slope of the straight line obtained by V (volume of the sludge filtrate) as the abscissa, t/V is the ordinate; μ - the viscosity of the filtrate (N·s/m^2); ω - the weight of the dry sludge (kg/m^3) produced per unit volume of the filtrate.

Reduction rate of sludge specific resistance was calculated according to Eq. (7):

$$R'(\%)=\frac{SRF_0-SRF_1}{SRF_0}\times 100\% \tag{7}$$

where R′ - reduction rate of sludge specific resistance (%); SRF_0 - the specific resistance before sludge conditioning (m/kg); SRF_1 - the specific resistance after sludge conditioning (m/kg).

Sludge particle size was measured using laser particle size analyzer (Mastersizer 2000, Malvern). Zeta potential was measured by zeta potential analyzer (Nano ZS). Scanning electron microscopy (HITACHI SU8010) was used to observe the microscopic morphology of the sludge. The concentration of dissolved aluminum in sludge filtrate was measured with an inductively coupled plasma

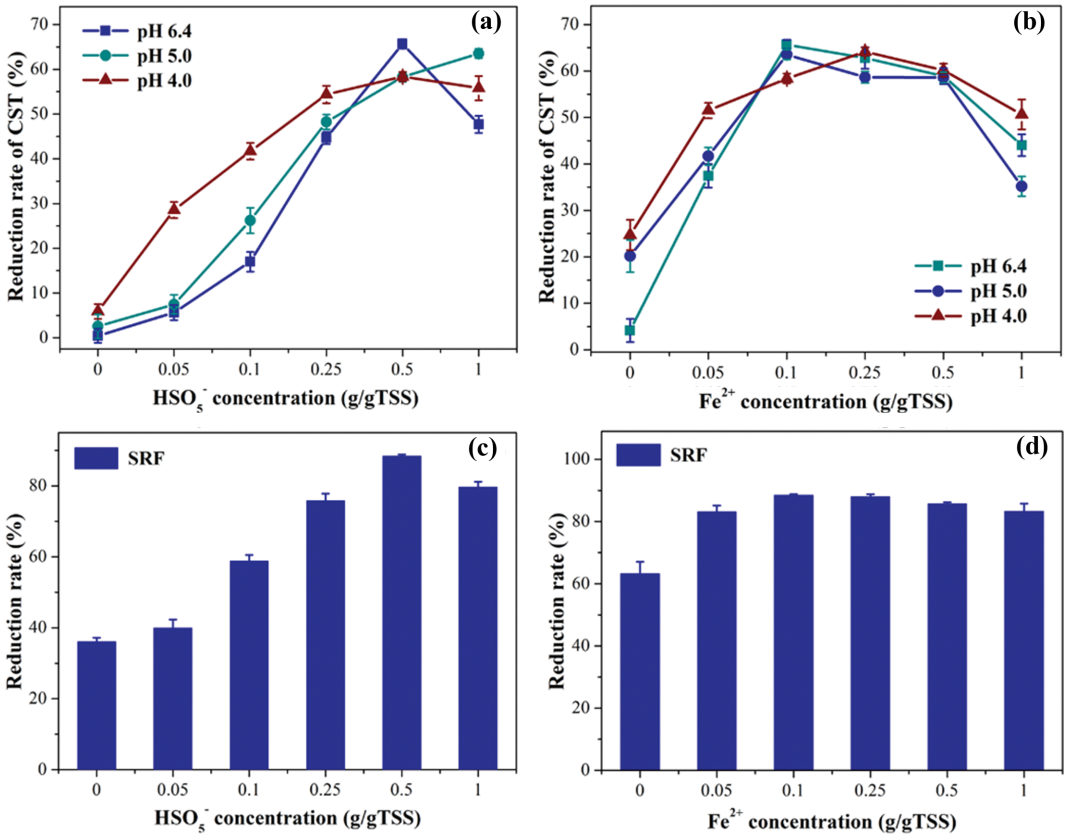

Fig. 1. (a) Effect of different HSO_5^- concentrations on reduction in CST (%). (b) Effect of different Fe^{2+} additions on reduction in CST (%). (c) Effect of different HSO_5^- concentrations on reduction in SRF (%). (d) Effect of different Fe^{2+} additions on reduction in SRF (%). In (a/c), the Fe^{2+} addition was 0.1 g/g TSS. In (b/d), the HSO_5^- concentration was 0.5 g/g TSS.

optical emission spectrometer (Optima 2100DV, PerkinElmer, USA).

RESULTS AND DISCUSSION

1. Impact of HSO_5^- Concentrations on CST Reduction Rates of Alum Sludge

As shown in Fig. 1(a), when the concentration of Fe^{2+} was 0.1 g/g TSS, with the increase of HSO_5^- concentration, the reduction rate of CST increased first and then decreased. The CST reduction rate reached a maximum of 65.7%, while the concentration of HSO_5^- was 0.5 g/g TSS with unadjusted pH=6.4.

In addition, when the HSO_5^- concentration was lower than 0.5 g/g TSS, lower pH value further promoted the enhancement of CST reduction rate with the same dosages of Fe^{2+} and HSO_5^-. However, when the concentration of HSO_5^- was higher than 0.5 g/g TSS, lower pH value presented no effect on the enhancement of CST reduction rate and inhibitory effect might exist.

2. Impact of Fe^{2+} Addition on CST Reduction Rates of Alum Sludge

According to Fig. 1(b), when the concentration of HSO_5^- was 0.5 g/g TSS, the reduction rate of CST increased with the increase of Fe^{2+} addition. When Fe^{2+} addition was 0.1 g/g TSS, the maximum reduction rate of CST reached 65.7%. Therefore, the optimal condition of HSO_5^- and Fe^{2+} addition was 0.5 g/g and 0.1 g/g TSS, respectively.

Further addition of Fe^{2+} led to the decrease of CST reduction rate. As shown in Eq. (1)-(4), since Fe^{2+} was easily oxidized to Fe^{3+}, excessive addition of Fe^{2+} could consume sulfate radicals; thus, the alum sludge was not sufficiently oxidized, which deteriorated sludge dewaterability.

On the other hand, from Fig. 1(b), when the Fe^{2+} addition was lower than 0.1 g/g TSS, lower pH value enhanced CST reduction rate. By contrast, when the addition of Fe^{2+} was over 0.1 g/g TSS, a contrary tendency was observed. The phenomenon was similar to the effects of HSO_5^- dosing under different pH values in section 3.1. Comparing the CST reduction rates after conditioning at different pH values (6.4, 5.0, 4.0), it was found that the optimum conditioning effect was obtained at pH=6.4. This indicated that different from the Fenton or Fenton-like systems, oxidative effect of Fe^{2+}-HSO_5^- system was not enhanced under acid condition.

3. Impact of Fe^{2+}-HSO_5^- Concentrations on SRF Reduction Rates of Alum Sludge

As shown in Fig. 1(c), when the concentration of Fe^{2+} was 0.1 g/g TSS, as the concentration of HSO_5^- increased, the reduction rate of SRF increased and the maximum value was obtained at 0.5 g/g TSS Fe^{2+}. When the concentration of HSO_5^- was maintained as 0.5 g/g TSS, the reduction rate of SRF increased first and then slightly decreased along with the increase of Fe^{2+} addition. The reduction rate of SRF reached a maximum of 88.4% while the Fe^{2+} addition was 0.1 g/g TSS. Therefore, the optimum concentration of Fe^{2+} and HSO_5^- was 0.1 g/g and 0.5 g/g TSS, respectively. The results were consistent with the changes of CST reduction rates described in section 3.1 and 3.2.

4. Changes of Al Concentrations in the Sludge Filtrate

Aluminum sulfate and polyaluminum chloride are widely used as coagulants during the drinking water treatment process. Therefore, the water purification sludge contains a large amount of Al [48]. The sludge produced by using alum is gelatinous and has a feathery, fluffy nature [49-51]. Alum recycling is simple in chemical mechanisms. However, this process is difficult due to the removal of some raw water impurities during the water treatment and mixing with the aluminum hydroxide [52]. Therefore, it is an effective resource recovery process to dissolve the sludge and release aluminum by oxidation conditioning. Normally, aluminum sulfate is very soluble in water and the aqueous solution is weakly acidic. When the mixture becomes alkaline, the aluminum ions in the solution combine with the hydroxide to form flocs. According to the literature, the Fe^{2+}-PMS system is more effective in acidic conditions [53]. In this study, all the pH was acidic, so the precipitate of aluminum hydroxide could not be produced in the mixture. Moreover, regardless of the pH value, most of the residual Al existed in the form of dissolved Al [54]. It can be seen from Fig. 2 that when the Fe^{2+} addition maintained 0.1 g/g TSS, the Al concentration in the sludge filtrate increased with the increase of HSO_5^- concentration; when the HSO_5^- concentration was set as 0.5 g/g TSS, the Al concentration in the filtrate increased with the increase of Fe^{2+} addition, indicating that $SO_4^{\bullet-}$ destroyed the structure of sludge flocs; thus, Al was dissolved from the coagulation sediment,

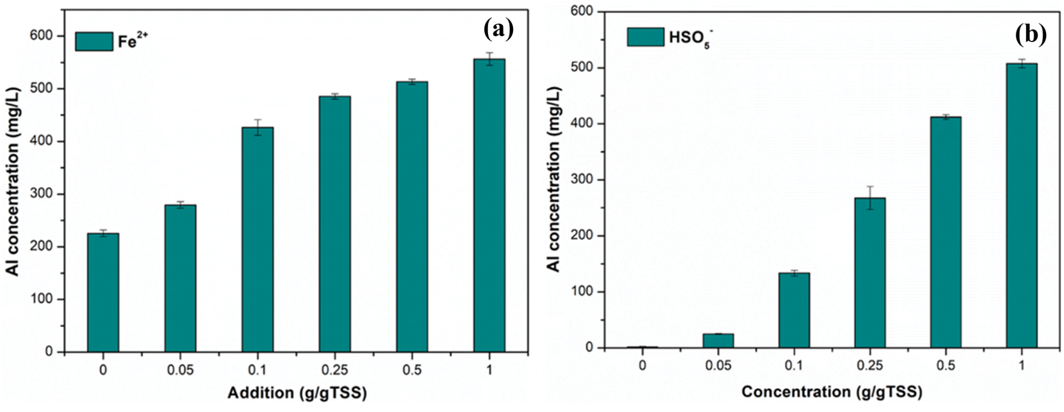

Fig. 2. Effect of different Fe^{2+} addition (a) and HSO_5^- concentrations (b) on dissolved aluminium from alum sludge. In (a), the HSO_5^- concentration was 0.5 g/g TSS. In (b), the Fe^{2+} addition was 0.1 g/g TSS, pH=6.4.

which facilitated the subsequent sludge disposal. On the other hand, Al in the filtrate can also be further recycled [55,56]; thus oxidative conditioning provided an alternative way for extracting and recovering aluminum from water purification sludge.

5. Changes of Alum Sludge Characterizations

According to Fig. 3, the particle sizes of the alum sludge at different pH values were significantly reduced after the dosage of Fe^{2+}-HSO$_5^-$. At pH 6.4, the particle size of alum sludge before con-

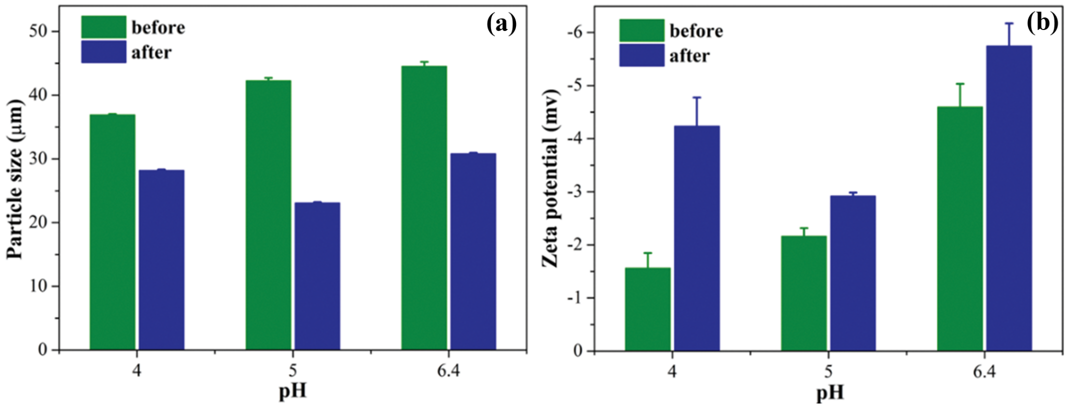

Fig. 3. Particle size and zeta potential of alum sludge before (a) and after oxidative conditioning (b). Experimental conditions: Fe^{2+} addition: 0.1 g/g TSS, and HSO$_5^-$ concentration: 0.25 g/g TSS. pH: 6.4, 5.0, 4.0.

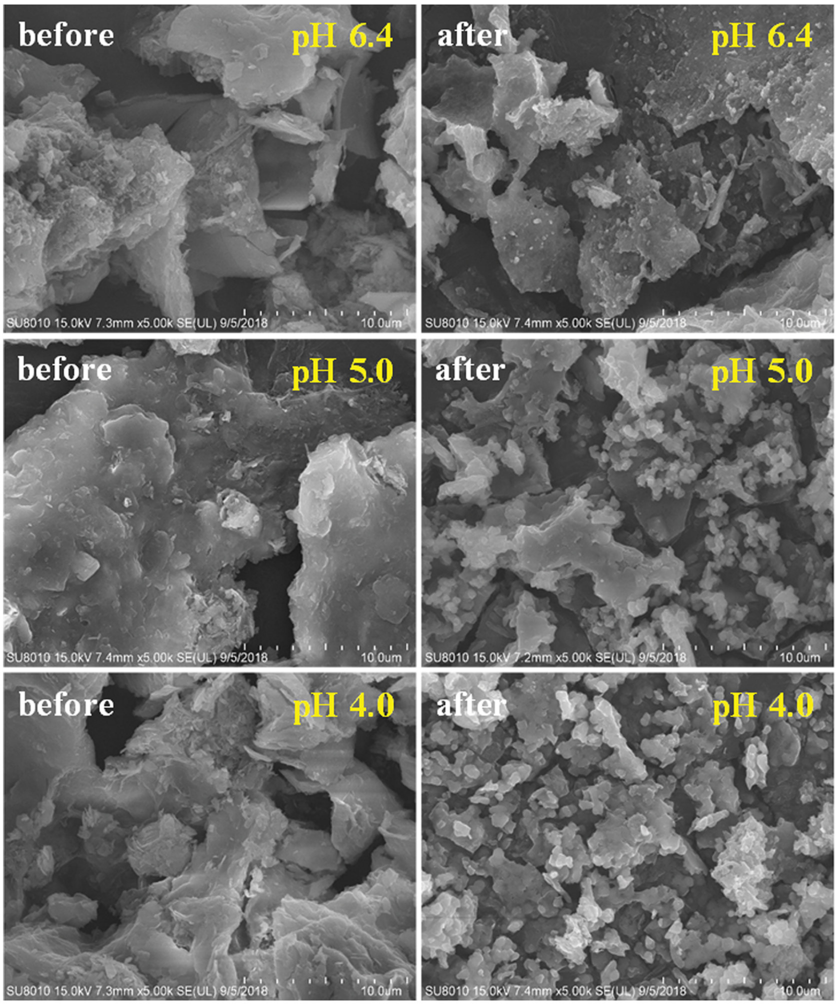

Fig. 4. SEM results of alum sludge before and after oxidative conditioning. Experimental conditions: Fe^{2+} addition: 0.1 g/g TSS, and HSO$_5^-$ concentration: 0.5 g/g TSS.

ditioning was 44.5 μm. After conditioning, the particle size was reduced to 30.8 μm, indicating that $SO_4^{\bullet-}$ generated by Fe^{2+} activated HSO_5^- oxidized and destroyed sludge flocs. Along with the decrease of sludge particle sizes, the bound water was released, and the sludge dewaterability was thus improved [53].

It has been reported that when the absolute value of the zeta potential is larger, the electrostatic repulsion between the colloidal particles is greater, and the system is more stable, which inhibits the aggregation of the sludge particles [57]. Zeta potential increases after oxidation, which may be related to changes in the concentration of Al ions. The higher the absolute value of Zeta potential achieved, the greater the electrostatic repulsion between colloidal particles that exists. The system is therefore more stable and the sludge particles are less likely to aggregate, which is more favorable for dewatering [58]. Before conditioning, the absolute value of the zeta potential decreased with the pH value decreased. This was due to the addition of acid, and the introduced H^+ neutralized the negative charge carried by the sludge particles [34]. Compared with the changes of zeta potential before and after sludge conditioning (Fig. 3(b)), the absolute value of the zeta potential of the alum sludge increased after conditioning, which might have been due to the increase of aluminum concentration in the sludge filtrate. $SO_4^{\bullet-}$ oxidation destroyed the original coagulation state of the alum sludge, dissolved the Al from it, and increased the absolute value of the Zeta potential of the alum sludge [59].

For further investigating the changes of the alum sludge structure and analyzing the reasons for the improvement of alum sludge dewaterability, scanning electron microscopy (SEM) was used to observe the microscopic characteristics of the alum sludge before and after oxidative conditioning under different pH values. As shown in Fig. 4, the alum sludge was in block shape before oxidation and the surface was relatively flat and smooth with compact texture. After oxidation, the blocky structure diminished and fine particles were generated on the surface of alum sludge. It was because the large sludge floc particles were cracked into small particles by oxidative effects, while destroying the originally smooth and flat surface. With the tight texture of the alum sludge destroyed, the bound water in sludge flocs was released. The coarse small sized particles were not easily combined with water again; thus the dewaterability of alum sludge was improved [60].

CONCLUSION

A novel Fe^{2+}-activated peroxymonosulfate oxidative process was applied to enhance dewaterability of alum sludge from water purification facilities. Tests were conducted to explore the optimal conditions for oxidative conditioning of alum sludge. The results showed that the alum sludge dewaterability was enhanced significantly, with the reduction rates of CST and SRF up to 66% and 88%. The characteristics of alum sludge (particle size, zeta potential, microscopic morphology) during conditioning process were also observed and analyzed. It was found that the oxidative sulfate radicals produced by Fe^{2+}-PMS system could effectively decompose the alum sludge flocs, while releasing trapped water and aluminum ions, thus further enhancing alum sludge dewaterability.

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