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Treatment of landscape water (LSW) by electrocoagulation process

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ABSTRACT

The aim of this study was to utilize an aluminum electrocoagulation for the removal of algae and dissolved organic matter from landscape water, which was taken from an artificial lake and mixed with NaCl stock solutions to make its final concentration in the range of 0.5–3 g/l. The removal efficiency of chlorophyll-*a*, UV_{254} and turbidity was investigated under different current densities, charge loadings, conductivities (689–4684 $\mu s\ cm^{-1}$) and pH values (3–11). The comparative removal performance together with sludge production by chemical coagulation and electrocoagulation were studied. With electrocoagulation, the optimal removal efficiencies of chlorophyll-*a* and UV_{254} were 81% and 56%, respectively, and the residual turbidity and sludge production were less than 2.6 NTU and 5.1% of the total solution (after 10 min sedimentation), respectively. In comparison, for chemical coagulation the optimum removals of chlorophyll-*a* and UV_{254} were 75% and 46%, respectively, and the residual turbidity and the sludge production were 3.6 NTU and 9.3% of the total solution (after 10 min sedimentation), respectively. The results demonstrated that electrocoagulation was an effective process for the removal of algae and dissolved organic matter from landscape water and exhibited advantages to chemical coagulation.

Keywords: Algae; Aluminum electrode; Chlorophyll-*a*; Chemical coagulation (CC); Dissolved organic matter (DOM); Electrocoagulation (EC); Landscape water

1. Introduction

Landscape water is one of the main functional water bodies in people's life. It possesses many properties including flood buffer, entertainment and water supply in emergencies, swimming, fishing, and landscape view. However, algae and organic pollutants would cause the loss of these functions. The character of landscape water may vary with the season change. Normally, algae and dissolved organic matter (DOM) are the typical pollutants in landscape water.

The existence of algae in water not only causes uncomfortable taste and odour, but also affects the normal eco-system due to the decay of algae [1,2] and ultimately, eutrophication. With increasing living standards, more attentions have been paid to the landscape water pollution; removing algae and DOM is one of major tasks to maintain the quality of landscape water.

Due to the high water content in algae [3], algae sludge is normally suspending in water phase with poor sedimentation, and chemical coagulation cannot effectively separate the algae sludge from water. Filter systems are often clogged due to low separation efficiency of the algae sludge. Alternative technologies are

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sought to achieve high treatment efficiency and convenient operations for the removal of algae and DOM from landscape water.

Water treatment with electrocoagulation (EC) process has been studied since 1980s [4,5] and been used in some cases in pilot-and full-scale treatment [6,7]. Until now, EC technologies have been widely studied on the removal of heavy metals [8], oil [9], boron [10], fluoride [11] and phosphate [12], and in the treatment of printing and dyeing wastewater [13,14], landfill leachate [15], ground water [16], urban wastewater [17], washing water [18], and tanning wastewater [19]. Nevertheless, the use of EC to remove low concentrations of algae and DOM from landscape water has not been investigated.

The aim of this paper is to investigate the use of EC to remove algae and DOM from landscape water. The operating parameters of EC which affect the removal efficiencies of algae, DOM and turbidity are to be optimized in terms of the charge loading, current density, initial pH and conductivity. The treatment performance by EC and chemical coagulation were compared for the same dosage of aluminum. In addition, the sludge volume production by EC and chemical coagulation was studied.

2. Materials and methods

2.1. EC reactor and operating conditions

The schematic diagram of the EC reactor can be seen in Fig. 1. The reactor used was a cylinder with a diameter of 100 mm and volume of 1.4 l. Aluminum electrodes were shaped in the size of 70 mm × 20 mm × 0.75 mm. The effective anode area is 1.4×10^{-3} m^2. Magnetic mixing equipment and a magnetic stirrer, which was laid on the bottom of the reactor, were used to mix solutions in the electrolysis. The stirring speed was 100 rpm. The reactor was filled with 1l of landscape water for each run. The applied current ranged between 2.45×10^{-2} A and 9.8×10^{-2} A, which gives the current density of 17.5 to 70 A m^{-2}. The electrolysis time was between 2 min to 10 min for the various charge loadings. 10 min after the electrolysis stopped, the samples were taken from the reactor outlet and centrifuged to separate sludge from the test solution. Various water quality parameters were then analyzed (See 2.4).

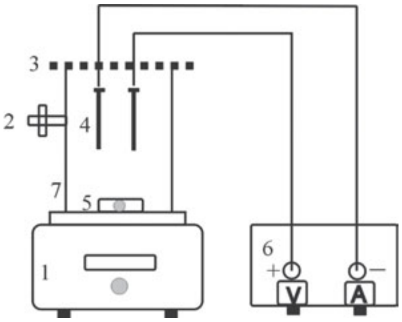

Fig. 1. Schematic diagram of electrocoagulation-flotation reactor. (1) magnetic mixing equipment (2) sample outlet (3) plates gap controller (4) electrode plates (5) magnetic stirrer (6) D.C. power supply (7) reactor.

Aluminum dissolution concentration from EC was calculated by Eq. (1).

$$C_{Al} = I \times t \times M_{Al} / (Z \times F \times V) \tag{1}$$

where, C_{Al}, the Al concentration in the reactor (g m^{-3}); I, the current applied (A); t, electrolysis time (s); M_{Al}, atom mass of aluminum (26.98 g/mol); Z, electron loses (Z = 3); F, Faraday constant (96500 C/mol); V, volume of EC reactor (m^3).

2.2. Coagulant

Aluminum sulfate (AS, analytical grade) and poly aluminum chloride (PAC, purchased from National Group, Al_2O_3 = 30%, percent basicity = 90%) were chosen as coagulants in chemical coagulation process.

2.3. Raw water properties

The landscape water sample was taken from an artificial lake flowing around the Tongji University campus. The volume of the artificial lake is 100,000 cubic meters with average depth of two meters. The parameters of the landscape water are shown in Table 1. Before electrocoagulation studies, the landscape water samples were mixed with a NaCl stock solution to make the final concentrations to be 0.5–3 g/l. The same test solutions with NaCl concentrations of 0.5–3 g/l were used for chemical coagulation studies.

2.4. Analytical methods and equipment

The pH and conductivity were measured by Metrohm Ion Meter (Switzerland). The turbidity was measured by 2100P turbidity meter (HACH). UV-abs at 254 nm was measured by UV-1700 spectrophotometer (Japan) after filtration by a 0.45 μm filter. The centrifuge was TDL80–2B (Pigeon, Shanghai). The power supply was DC 9100 (Xinjian, Shanghai, 0~36V). The chlorophyll–*a* and all other parameters were measured following the standard methods [20]. The chemicals used in this study were in analytical grade. All test were

Table 1
Characteristics of the landscape water

pH	Chlorophyll-*a* (μg/l)	UV_{254}-abs. (cm^{-1})	Conductivity (μs cm^{-1})	Turbidity (NTU)	Temperature (°C)	Color	Total P (mg/l)	Total N (mg/l)
7.63	45.05	0.154	689	11.34	20–25	light green	0.22	11.2

performed in triplicates with a standard deviation less than 4%. Statistical calculations were based on confidence level equal or higher than 95%.

3. Results and discussion

3.1. Mechanism of EC with aluminum electrode

EC technology uses sacrificial electrode such as Al and Fe to produce metal ions which can be dissolved in the solution and served as coagulants.

The main reactions occurred in anodes and cathodes are shown in the following equations:

$$\text{Anode: Al} \leftrightarrow Al^{3+} + 3e \tag{2}$$

$$\text{Cathode: } 3H_2O + 3e \leftrightarrow 3/2H_2 + 3OH^- \tag{3}$$

$$\text{Total reaction: } Al + 3H_2O \leftrightarrow Al^{3+} + 3OH^- + 3/2H_2 \tag{4}$$

If Cl^- exists in solution, side reaction will happen:

$$2Cl^- \leftrightarrow Cl_2 + 2e \tag{5}$$

$$2H_2O \leftrightarrow O_2 + 4H^+ + 4e \tag{6}$$

When pH value is between 3 and 4, the following reactions also occur:

$$Cl_2 + H_2O \leftrightarrow HOCl + H^+ + Cl^- \tag{7}$$

$$HOCl \leftrightarrow OCl^- + H^+ \tag{8}$$

Since the aluminum can be dissolved in solution at different pH, a series of hydrolyzing species could be formed, which include polymeric species such as $Al_{13}O_4(OH)_{24}^{7+}$ [21].

3.2. Current densities and charge loadings

The effect of the current density (*i*) and charge loadings on the removal efficiency of chlorophyll-*a* and UV_{254} -abs. are shown in Figs. 2 and 3.

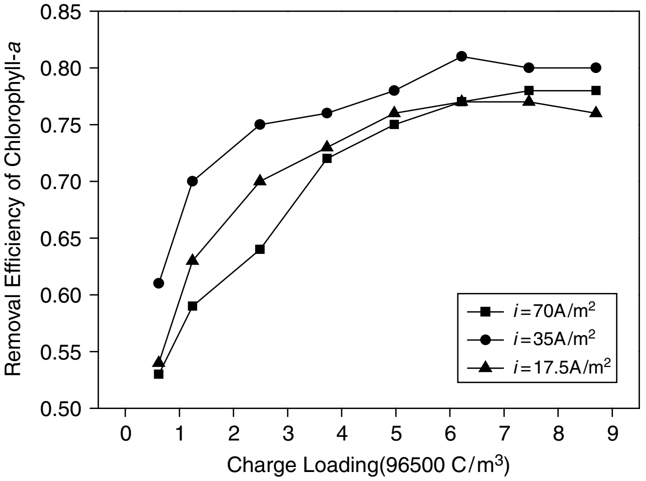

Fig. 2. Removing efficiency of chlorophyll-*a* with various current densities and charge loadings (pH = 7.63, NaCl = 1g/l).

Fig. 2 shows that the removal efficiency of chlorophyll-*a* increased with the enhancement of charge loading. When the charge loading was greater than 4.9 (unit is 96500 C/m^{-3}) chlorophyll-*a* removal efficiency was more than 70% in three different current densities and the residual concentration of chlorophyll-*a* in the effluent was less than 13.5 μg/l. When the charge loading continued to grow and current density was 35 A m^{-2}, the removal efficiency of chlorophyll-*a* reached 81% and the residual chlorophyll-*a* concentration in the effluent was 8.43 μg/l. According to Fig. 2, the optimal current density to achieve the highest removal efficiency of chlorophyll-*a* was 35 A m^{-2}. The reasonable explanation could be that for the optimal current density, the aluminum flocs formed had optimized specific surface areas, which could be beneficial for sedimentation, adsorption and trapping process [21].

Fig. 3 shows that the removal efficiency of UV_{254}-abs. was positively related to current density. When the charge loading was 7.59 and current density was 70 A m^{-2}, the removal efficiency of UV_{254} was over 56%. The high UV_{254}-abs. removal efficiency occurred in high current density. The possible reasons included two aspects.

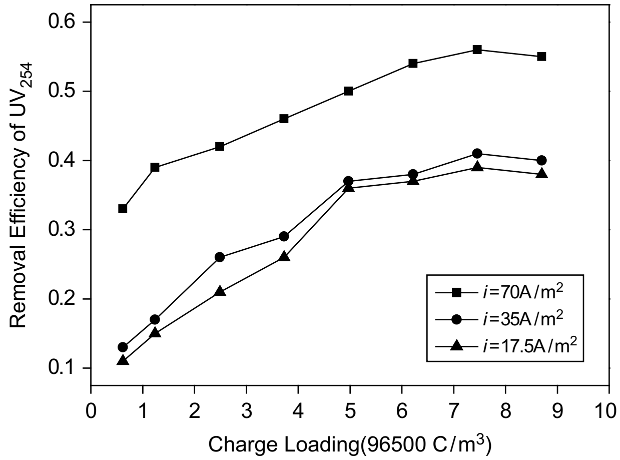

Fig. 3. Removing efficiency of UV_{254} with various current densities and charge loadings (pH = 7.63, NaCl = 1g/l).

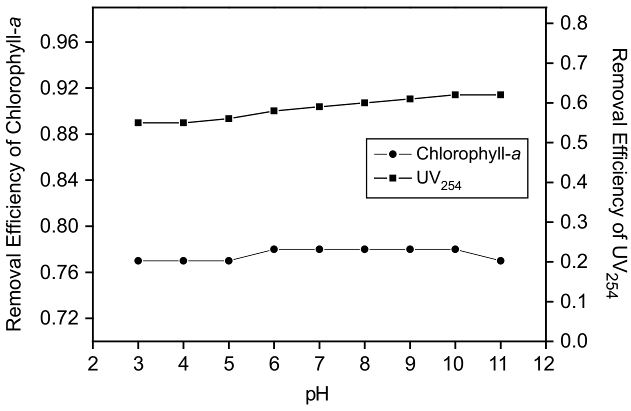

Fig. 4. Removing efficiency of chlorophyll-*a* and UV_{254} with varied pH (i = 70 A m^{-2}, electrolysis time = 10 min).

In one hand, the high current density could lead to high Al dissolution and more formation of $Al(OH)_3$ which could lead to the co-precipitation of $Al(OH)_3(s)$ with UV_{254} substances (Eq. (8)). On the other hand, side reactions around electrodes would occur according to Eqs. (4) and (6). The product of active HClO would degrade the UV_{254} substances and reduce the concentration of UV_{254} substances [7]. Nevertheless, a suitable current density must be determined based not only on the maximum removal of the UV_{254} matter but also on the highest removal of chlorophyll-*a.* Consequently, a current density between 35 and 70 A m^{-2} could be considered as the operating current density for the treatment of landscape water using EC.

$$n\left(\mathrm{Al(OH)_3}\right)(s) + m\mathrm{UV}_{254} \leftrightarrow \left[n\left(\mathrm{Al(OH)_3}\right)(s)\cdot m\mathrm{UV}_{254}\right](s) \qquad (9)$$

3.3. Initial pH

It was widely accepted that initial pH was an important parameter which could affect the EC efficiency. In this study, NaOH (1 M) and HCl (1 M) were used to adjust the pH value from 3 to 11. The removal efficiencies of UV_{254} and chlorophyll-*a* were investigated.

Fig. 4 shows that removal efficiency of chlorophyll-*a* was not significantly influenced by pH. The reasonable explanation was that aluminum dissolution can occur for both acid and alkaline conditions [11]. In low pH value (<5), aluminum dissolved as Al^{3+} and promoted the removal efficiency of chlorophyll-*a*. When the pH was high (9~10), a lot of hydroxyl groups were absorbed to surface of chlorophyll-*a*. The aluminum dissolved in anode electrode could form complexity precipitants with hydroxyl groups and chlorophyll-*a*. When the pH was over 10, the removal of chlorophyll-*a* decreased because the produced aluminum ions were mainly $Al(OH_4)^-$.

Fig. 4 also shows that the removal efficiency of UV_{254}-abs. increased slightly with pH value (4 to 10); from 55% (pH = 3) to 62% (pH = 10). The reason was probably that in high pH value, the morphology of aluminum was in plenty and the adsorption function of aluminum complexity was greatly enhanced.

The pH effect on the removal efficiencies of chlorophyll-*a* and UV_{254}-abs. was different. Chlorophyll-*a* represents algal particles with diameter over 0.45 μm whilst UV_{254} substances were in the size of less than 0.45 μm (soluble matter). The surface tension of chlorophyll-*a* was not as sensitive as that of soluble UV_{254} substances when solution pH changed, which could lead to the different removal efficiency in responding to the variations of pH.

3.4. Conductivity

Conductivity was the parameter showing the ion concentrations in solutions, which could affect the actual operating voltage of the EC. By adding given amount of NaCl, into the raw water, conductivity of the water increases. For a constant current density applied, a high conductivity in the electrolyte reduces the voltage required, which decreases the energy consumption and then the operating cost. A number of previous studies have approved this [22].

Fig. 5 shows that removal efficiency of chlorophyll-*a* was not affected by the conductivity. When the conductivity of solutions increased (corresponding to increasing in NaCl concentration from 0.5 g/l to 3 g/l), the fluctuation of removing efficiency of chlorophyll-*a* was just 1%. With increasing in the concentration of NaCl,

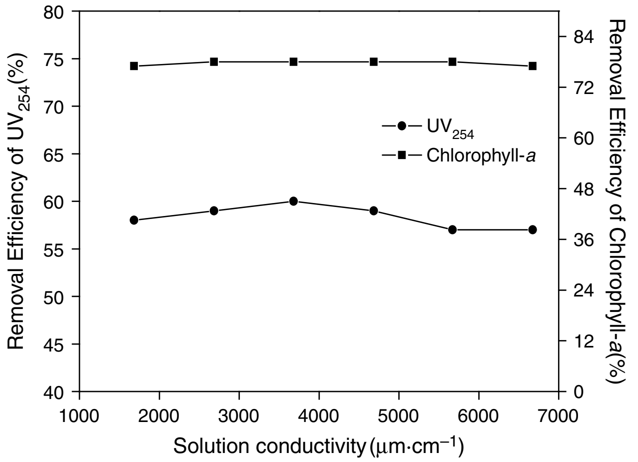

Fig. 5. Removing efficiency of chlorophyll-*a* and UV_{254} vs. solution conductivity (i = 70 A m^{-2}, Electrolysis time = 10 min).

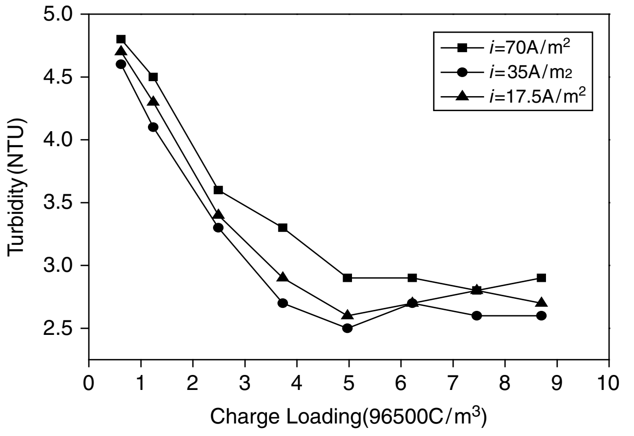

Fig. 6. Residual turbidity varied with charge loading for various current densities (pH = 6.76, NaCl = 1.0 g/l).

HOCl concentration would increase as well via the side electrolytic reactions which reduce the UV_{254} absorbance. However, this was offset by the side reaction of forming HOCl when [Cl^-] was greater than 1.5 g/l, which would reduce the aluminum dissolution rate. Consequently, the increase of conductivity did not promote removal efficiencies of UV_{254} and chlorophyll-*a*, but will reduce the operating voltage between electrodes and then save the operating cost.

3.5. Turbidity removal by EC

Some researchers [23] reported excellent removal of turbidity and ultra-fine particles by EC. This is also the case of this study. Fig. 6 shows when the charge loading was over 5.9 (unit = 96500 C m^{-3}), the turbidity of effluent was stable at 2.5~3 NTU. This is consistent with previous researches [16], where the residual turbidity was less than 5 NTU and not affected by the current densities.

3.6. Comparison between EC and chemical coagulation (CC)

Comparison between EC and CC was conducted in keeping same dosages of aluminum ions for EC and CC processes. CC was assessed using two coagulants as stated previously (AS and PAC) and the same reactor as EC equipment. Rapid mixing was very important to affect the coagulation performance then a preliminary study was conducted to determine the optimal rapid mixing speed, which was 300 rpm, and the efficiencies of removing UV_{254} and chlorophyll-*a* under 300 rpm were in the optimized value for all dosages. So, the CC process was operated at the following conditions: fast mixing at 300 rpm for 1 min, slow mixing at 50 rpm for 10 min, and sedimentation for 10 min. The samples were then centrifuged to separate sludge from the test solution. The velocity gradient of rapid mixing was 161.8 1 s^{-1}. The supernatant was sampled to measure UV_{254} and chlorophyll-*a*.

Figs. 7 and 8 show the comparative removal efficiency of chlorophyll-*a* and UV_{254}-abs. by EC and CC. When the aluminum dose was less than 40 mg/l, which corresponded to an electrolysis time of 2 min, the chlorophyll-*a* removal performance with EC was not as good as that of CC (Fig. 7). The reason was that for 2 min electrolysis, the EC process could be in a lag stage, where the process did not produce enough aluminum ions. Then, the removal efficiency of chlorophyll-*a* and UV_{254}-abs. is relevant to the initial electrolytic time of

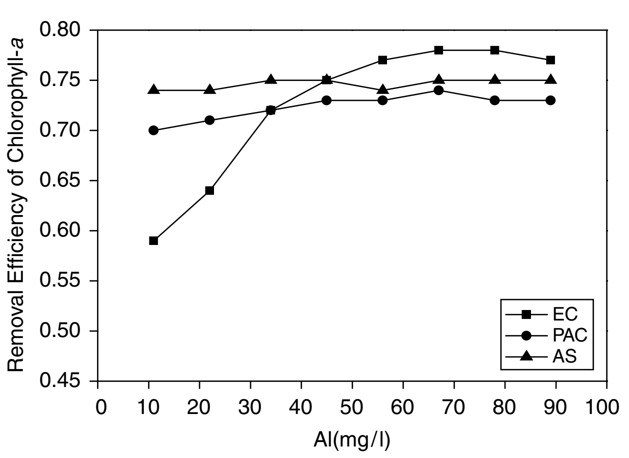

Fig. 7. Comparative chlorophyll-*a* removal performance by EC and CC (pH = 7.63).

the EC process. Fig. 8 shows that when the aluminum dose was less than 40 mg/l, the UV_{254} removal was very similar by EC and CC; whilst the aluminum dose was greater than 40 mg/l, the UV_{254} removal by EC was up to 56% which is greater than that by CC. Partly, this is due to not only the destabilsation of UV_{254} matter but also the electrolytic oxidation, which plays the role in the degradation of the UV_{254} matter by EC. The optimized removal efficiencies of PAC and AS were 50% and 48%, respectively in UV_{254} removing. For chlorophyll-*a*, the optimized removal efficiencies by EC, PAC and AS were 79%, 71%, 74%, respectively.

Sludge volume is an important parameter to show the density of flocs for the same dosage of aluminum ions. The height of the sludge in the reactor would reflect the volume and density of the sludge which affect the sedimentation and dewatering process for the same aluminum dosage.

Fig. 9 shows that for the dose range of aluminum ions applied from EC and CC, the sludge volume from EC was much less than that from dosed coagulants, AS and PAC. The phenomenon matched with previous researches about the sludge advantage of EC [21]. The increased concentration of aluminum in CC didn't improve the removal percentage of UV_{254} and chlorophyll-*a* greatly. Figs. 7–9 show that for a wide dose range by EC and CC, EC can achieve the overall better performance in terms of the removal of chlorophyll-*a* and UV_{254} matter and less sludge production.

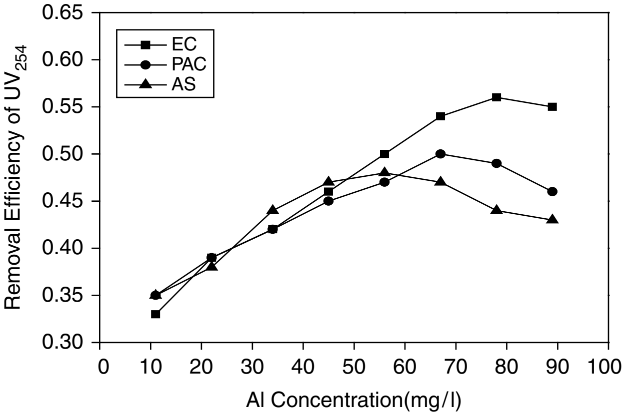

Fig. 8. Comparative UV_{254}–abs. removal performance by EC and CC (pH = 7.63).

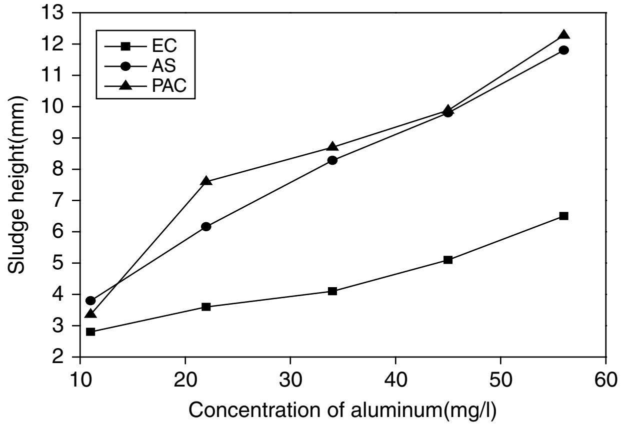

Fig. 9. Comparison of sludge between EC and CC (pH = 7.63).

4. Conclusions

The EC and CC were used to assess the performance of removing algae, turbidity and DOM from landscape test solutions (artificial lake mixed with NaCl solutions (concentrations ranged 0.5–3 g/l)) and the following conclusions can be made from the study:

1. After the EC treatment, the effluent was with low concentration of UV_{254} (DOM), chlorophyll-*a* and turbidity.
2. For different current densities, the EC process removed chlorophyll-*a* of 77%~81% and UV_{254} of 56%~62%, respectively. The residual turbidity was between 2.5 and 2.9 NTU. The residual concentration of chlorophyll-*a* in the effluent was less than 10 μg l^{-1}, which matched the eutrophication control threshold.
3. pH and conductivity hardly affected the performance of the EC process.
4. The EC process performed better than CC process for the same aluminum dosages based on a high removal efficiency of UV_{254} (DOM), chlorophyll-*a* and turbidity and less sludge production, this demonstrates that EC is a promising technology in landscape water treatment, especially for the removal of algae, DOM and turbidity.

Future work is considered to scale up the EC reactor for the full scale trials in the treatment of landscape water with electrocoagulation. The assessment of the performance will be focus to the overall treatment efficiencies as well as economic robust by modifying reactor designs and optimization of the operation.

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