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Optimization of low-concentration phosphorus removal from raw lake water

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ABSTRACT

Many of our lakes are suffering from algal blooms and depleted dissolved oxygen, which are caused by nutrients such as phosphorus deposited in the lakes by large rainstorms. Because the issues of eutrophication have not been solved due to the difficulties in treating low-concentration phosphorus samples, advanced treatment of raw lake water is needed. In this study, electrocoagulation (EC) and microfiltration (MF) were used to efficiently remove the total phosphorus from raw water samples from a lake. Based on the optimal conditions presented in a previous study—the type of electrode: aluminum, iron, and steel use stainless, spacing of the electrodes: 3 mm, reaction time: 1 min, temperature: 15°C, and pH: 7.0—we applied a molar ratio according to the percentage of average concentration of total phosphorus. As the current increased, the removal efficiency of the phosphorus increased. The validity of the method presented in this paper was examined by the molar ratio (5:1 for aluminum vs. phosphorus) using Faraday's law. It is noteworthy that (i) the aluminum electrodes exhibited scaling, whereas the TiO_2-coated aluminum electrodes did not, and (ii) that chloride in the electrolyte inhibited the removal of phosphorus. The raw water after EC was precipitated for 30 min and filtered using Amicon® (made of cellulose acetate with a 0.22-μm pore size). As a result, the water quality from this study achieved level III (≤0.05 mg/L for total phosphorus and ≤0.6 mg/L for total nitrogen). This suggests that a process combining EC with MF reduced eutrophication, resulting in high-quality water.

Keywords: Electrocoagulation; Electrochemical; Membrane; Molar ratio; Scale prevention

1. Introduction

Since nutrients cause algal blooms in contaminated lakes, clean water can be obtained, if the nutrients are controlled. In this study, we attempted to address this problem by removing phosphorus, the most influential nutrient, among others. Although phosphorus is mostly removed by biological treatments, these procedures have a number of disadvantages such as an extensive building area required and high maintenance costs. To overcome these, electrocoagulation (EC) is recommended, which simply needs a small-scale building sites, and keeps the water quality stable even after coagulation with the easy maintenance.

However, an additional membrane (microfiltration [MF]) process is also required, because it separates or removes the particles such as organic, inorganic, and ionic matter which cannot be removed solely through EC [1,2]. In the EC process, the generated metal cations are converted and diffused owing to the power of the electric field and the concentration of the contaminants in the water. Consequently, they electrically combine with colloids particle, which eventually coagulate and precipitate out.

(Anode reaction): Elution of metal ions:

$$\begin{aligned} &Al \rightarrow Al^{3+} + 3e^- \\ &4OH^- \rightarrow 2H_2O + O_2 + 4e^- \end{aligned} \quad (1)$$

(Cathode reaction): The electrolysis reaction of water, formation reaction of hydrates of aluminum, and hydrogen generation:

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$$\begin{aligned}&H_2O \rightarrow H^+ + OH^-\\&Al^{3+} + 3OH^- \rightarrow Al(OH)_3\downarrow\\&2H^+ + 2e^- \rightarrow H_2\uparrow\end{aligned} \tag{2}$$

The aluminum anode elutes Al^{3+} ions, when Al metal is oxidized in the electrolytic reaction. And then the eluted Al^{3+} ions are chemically removed, combining with phosphoric acid under the water. The Al^{3+} ions combine with OH^- ions generated by electrolysis at the cathode, creating $Al(OH)_3$, which functions as a coagulant. Phosphoric acid is then removed by physical absorption. The generated $Al_n(OH)_{3n}$ as a high-polymer substance seems to mutually combine the contaminants. Contaminants such as inorganics and organics combine with $Al_n(\text{total organic carbon})_{3n}$, which coagulates and precipitates. The simultaneous chemical reactions at the anode and cathode can be represented as in Eqs. (3) and (4), respectively.

$$\begin{aligned}&Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3e^-\\&Al^{3+} + TOC \rightarrow Al(TOC)\\&Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+\\&Al(OH)_3 \rightarrow Al_n(OH)_{3n}\\&Al^{3+} + PO_4^{3-} \rightarrow AlPO_4\\&Al(OH)_3 + PO_4^{3-} \rightarrow AlPO_4(OH)_3\end{aligned} \tag{3}$$

$$\begin{aligned}&2OH^- + 1/2O_2 \rightarrow H_2 + 2e^-\\&2H_2O + 2e^- \rightarrow H_2 + 2OH^-\\&HCO_3 + OH^- \rightarrow CO_3^{2-} + H_2O\\&CO_3^{2-} + Ca^{2+} \rightarrow CaCO_3\end{aligned} \tag{4}$$

The metal hydroxide is generated by hydrolyzing the dissolved metal ions at the anode. Because this metal hydroxide activates more strongly than those generated using chemical methods and has a low zeta potential, it results in high coagulation, absorption, and precipitation.

Precision filtration removes bacteria as well as floating solid matter, resulting in high-purity water. The membrane separation process separates or removes particles that cannot be removed through normal filtration. Precision filtration explains permeate flux in Lmh units through Darcy's law as shown in Eq. (5).

$$Q = k\frac{P_b - P_a}{pgL}A \tag{5}$$

As presented in Eq. (5), the permeate flux is inversely proportional to the resistance. Thus, when resistance increases, we are unable to obtain useful water flow. Resistance increases when the deposition of colloidal substances on the membrane surface blocks the pores. This blocked membrane can be returned to its original state by replacing the contaminated membrane. We tried to remove fouling through backwashing using a physicochemical method and to increase the restoration rate using a clean-in-place (CIP) chemical cleansing process. In this study, we investigated the optimum operating conditions of EC and MF, focusing particularly on phosphorus removal in a lake. We also presented a comparative analysis of phosphorus removal rates through molar ratios in an attempt to compensate for the demands of EC process, which requires considerable electrical power and regular electrode changes. A pretreatment process is considered to avoid the formation of scales on the electrodes, which decrease processing efficiency. The effect of chloride ions is also considered, which can influence the phosphorus removal.

2. Materials and methods

The experimental methods are given [3]. The EC and MF experiments were then conducted through a batch process.

2.1. Materials

This study used raw water at the lake at Hanulmos in Seoul, Korea. In raw water which has the low concentration total phosphorus (TP), a 0.1-N KH_2PO_4 solution was added to clearly confirm the removal efficiency of phosphorus. The sample pH is an important factor in coagulation; within the neutral range of pH, insoluble $Al(OH)_3$ is generated as a result of the combination of the hydroxyl ions (OH^-) with aluminum in the water. Therefore, in this study, we used an aqueous solution of 0.1 N HCl and 0.1 N NaOH in order to maintain a pH of 7. The pH was measured using the 238-180 model (ISTEK, Korea) of a PCE-6000 (Eutech, Korea) system. In the experiment, conductivity was measured as a function of the concentration of chloride from 0.2 to 8 mM. Table 1 shows the average values of the pollutants in the Hanulmos lake water.

2.2. Experimental methods

The raw water was analyzed using the following methods and equipment. Two types of electrodes were used: one was aluminum and the other was an aluminum electrode coated in sequence with 0.3% and 2% TiO_2. The TiO_2 used was a liquid product patented by ENB Korea Corporation, Korea. The coating method was simple: first, we removed impurities from the electrode using a brush with alcohol. Then, we applied 0.3% TiO_2 to the electrode surface. The electrode was allowed to dry for 3 h at room temperature. We then coated the electrode with 2% TiO_2 and again left it dry. The presence and volume of scaling was investigated using a scanning electron microscope (SEM) and an energy dispersive X-ray spectrometer (EDS). The SEM used an a-3400N (Hitachi) system and EDS used a JSM-7001F (JEOL) system.

Table 1
Raw water characteristics[a]

Parameters	Average
pH	7.04
Conductivity (μs/cm)	337.5
BOD (mg/L)	24
COD (mg/L)	34
TN (mg/L)	1.35
TP (mg/L)	0.2
SS (mg/L)	42

[a] BOD, biochemical oxygen demand; COD, chemical oxygen demand; and SS, suspended solid.

The effect of chloride was measured using ion chromatography (IC, ICS-900, Dionex, USA). The nonuniform pore size of MF accelerated fouling. Therefore, before the experiment, the film was soaked in an aqueous solution of 20% methanol for 24 h and then rinsed in the dark. Last, an Amicon® Filtration cell (Model 8200, USA) was applied to the membrane filter (a mixed cellulose ester manufactured by GSWP09000 Millipore, USA). The arrangement of the experimental equipment is illustrated in Fig. 1.

3. Results and discussion

3.1. Electrocoagulation

The optimal conditions for EC include the electrode type, spacing of the electrodes, reaction time, temperature, current and voltage, and conductivity [2]. Although the iron electrode showed the highest removal efficiency for phosphorus, additional costs needs to remove its chromaticity [4]. Thus, aluminum electrodes were selected. The space between each electrode was 3 mm and the reaction time was 1 min because 99% of the phosphorus was removed under these conditions.

3.1.1. Optimal conditions: current and voltage using molar ratio

EC is the major electrochemical reaction that releases metals at the anode, producing metal hydroxides by hydrolysis [4,5]. According to Faraday's law, these metal cations are released in proportion to the electrical power, which is represented by Eq. (6).

$$W = \frac{ItM}{zF} \tag{6}$$

In Eq. (6), W is the mass of the dissolved metal (g), I is the applied current (A), t is the reaction time of EC process (s), M is the molecular weight of the anode metal (g/mol), F is the Faraday constant (96,485 C/mol), and z is the valance number of ions of the substance. The optimal current, voltage, and electrical power were determined by examining the molar ratios given by Eq. (6). Experiments were conducted at different voltages and currents as functions of the molar ratios, which indicated the concentration of metal ions vs. phosphorus in solution. In this study, based on the Faraday equation, the optimum molar ratio that satisfied the concentration of TP of the raw water of 0.05 mg/L (i.e., water quality standard from 0.1 to 1 mg/L) was selected. The following equation (Eq. (7)) with the optimal reaction time fixed as 1 min was used to determine the optimum molar ratio. First, the average concentration of phosphorus in the lake (0.2 mg/L) was calculated.

$$P(\text{mol/L}) = \frac{0.002\,\text{g}}{\text{L}} \times \frac{\text{mol}}{30.974\,\text{g}} = 6.457 \times 10^{-6}\,\text{mol/L} \tag{7}$$

Then, the current was calculated by Faraday's law. The results are given in Table 2.

Based on the molar ratio applied, the phosphorus concentration of the target water quality was satisfied under the conditions in which Al:P was 5:1. The optimal current was 0.16 A and the voltage 11.61 V. Under these conditions the phosphorus removal efficiency was approximately 75%. Table 3 shows the optimal molar ratio, current, TP concentration, and removal efficiency.

The greatest advantage of using the molar ratio is to satisfy the amount of eluted aluminum and the target water quality depending on raw water concentration over adjusting current and voltage. Table 4 shows a comparison of the amount of electrical power used. If the molar ratio is applied according to the raw water concentration after setting the target water quality, the current will be controlled, thus reducing costs.

3.1.2. Effect of chloride by conductivity

Numerous studies demonstrated that electrolyte increases the conductivity, minimize the energy consumption and thus improves the removal efficiency of the phosphorus during EC [6,7]. Their principles are as follows: adding NaCl in EC can improve the amount of the released Al^{3+} by increasing the amount of combinable metal coagulant in the solution thus helping removal efficiency of phosphorus. However, I need a thorough investigation on the correlation between Cl^- ions and Al^{3+} in phosphorus removal, because I used NaCl as an electrolyte and the electrodes made of Al^{3+} in this study. The result of my investigation is shown in Fig. 2.

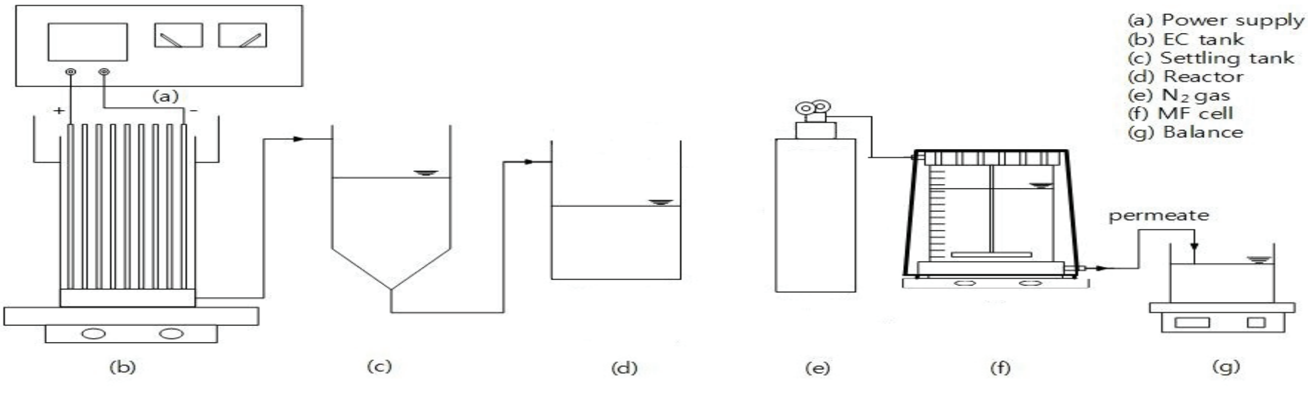


Fig. 1. Experimental equipment.

Table 2
Current calculations through molar ratios

Al:P = 1:1

$$\frac{xA \times 60\,\mathrm{s} \times 27\,\mathrm{g/mol}}{3 \times 96,487\,\mathrm{C/mol} \times 1\,\mathrm{L}} = 6.457 \times 10^{-6}\,\mathrm{mol/L} \times 27\,\mathrm{g/mol} \times 1$$

$X = 0.03$ A

Al:P = 1.5:1

$$\frac{xA \times 60\,\mathrm{s} \times 27\,\mathrm{g/mol}}{3 \times 96,487\,\mathrm{C/mol} \times 1\,\mathrm{L}} = 6.457 \times 10^{-6}\,\mathrm{mol/L} \times 27\,\mathrm{g/mol} \times 1.5$$

$X = 0.05$ A

Al:P = 2:1

$$\frac{xA \times 60\,\mathrm{s} \times 27\,\mathrm{g/mol}}{3 \times 96,487\,\mathrm{C/mol} \times 1\,\mathrm{L}} = 6.457 \times 10^{-6}\,\mathrm{mol/L} \times 27\,\mathrm{g/mol} \times 2$$

$X = 0.06$ A

Al:P = 2.5:1

$$\frac{xA \times 60\,\mathrm{s} \times 27\,\mathrm{g/mol}}{3 \times 96,487\,\mathrm{C/mol} \times 1\,\mathrm{L}} = 6.457 \times 10^{-6}\,\mathrm{mol/L} \times 27\,\mathrm{g/mol} \times 2.5$$

$X = 0.08$ A

Al:P = 3:1

$$\frac{xA \times 60\,\mathrm{s} \times 27\,\mathrm{g/mol}}{3 \times 96,487\,\mathrm{C/mol} \times 1\,\mathrm{L}} = 6.457 \times 10^{-6}\,\mathrm{mol/L} \times 27\,\mathrm{g/mol} \times 3$$

Al:P=3.5:1

$X = 0.09$ A

$$\frac{xA \times 60\,\mathrm{s} \times 27\,\mathrm{g/mol}}{3 \times 96,487\,\mathrm{C/mol} \times 1\,\mathrm{L}} = 6.457 \times 10^{-6}\,\mathrm{mol/L} \times 27\,\mathrm{g/mol} \times 3.5$$

$X = 0.11$ A

Al:P = 4:1

$$\frac{xA \times 60\,\mathrm{s} \times 27\,\mathrm{g/mol}}{3 \times 96,487\,\mathrm{C/mol} \times 1\,\mathrm{L}} = 6.457 \times 10^{-6}\,\mathrm{mol/L} \times 27\,\mathrm{g/mol} \times 4$$

$X = 0.12$ A

Al:P = 4.5:1

$$\frac{xA \times 60\,\mathrm{s} \times 27\,\mathrm{g/mol}}{3 \times 96,487\,\mathrm{C/mol} \times 1\,\mathrm{L}} = 6.457 \times 10^{-6}\,\mathrm{mol/L} \times 27\,\mathrm{g/mol} \times 4.5$$

$X = 0.14$ A

Al:P = 5:1

$$\frac{xA \times 60\,\mathrm{s} \times 27\,\mathrm{g/mol}}{3 \times 96,487\,\mathrm{C/mol} \times 1\,\mathrm{L}} = 6.457 \times 10^{-6}\,\mathrm{mol/L} \times 27\,\mathrm{g/mol} \times 5$$

$X = 0.16$ A

Table 3
Removal efficiency by application of molar ratio

Molar ratio (Al:P)	Current (A)	TP concentration (mg/L)			TP concentration average (mg/L)	Removal efficiency (%)
	Raw water	0.191	0.188	0.188	0.189	
1.5:1	0.05	0.165	0.167	0.164	0.165	12.70
2.5:1	0.08	0.053	0.056	0.051	0.053	71.96
5:1	0.16	0.053	0.045	0.047	0.048	74.60
7.5:1	0.23	0.021	0.021	0.023	0.022	88.36
10:1	0.31	0.016	0.012	0.010	0.013	93.12
20:1	0.62	0.003	0.003	0.005	0.003	98.41

Table 4
The electrical energy comparison according to the molar ratio applied

Molar ratio (Al:P)	Current (A)	Voltage (V)	Quantity of electricity (W)	Electric energy (Wh)	Removal efficiency (%)
5:1	0.16	11.61	1.86	0.03	75
20:1	0.62	20	12.40	0.21	98

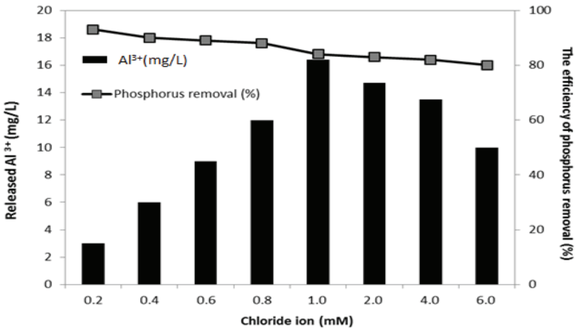


Fig. 2. Concentration of the released Al^{3+} and the efficiency of phosphorus removal according to the concentration of chloride ions.

Fig. 2 shows that (i) the efficiency of phosphorus removal was highest at 0.2 mM Cl^-, (ii) as the concentration of Cl^- ions is over 0.2 mM, the efficiency of phosphorus removal decreased, and (iii) the concentration of Al^{3+} increased and decreased proportional to the concentration of Cl^- ions by the standard of 1.0 mM of Cl^- ions. This result suggests that high concentration of Cl^- ions does not necessarily improve phosphorus removal. Rather, it enhances removing phosphorus to a certain extent (i.e., 1.0 mM of its concentration), but over this stage, it may corrode the electrodes, thus decreasing phosphorus removal.

3.1.3. Pretreatment method for scale minimization of electrode

Kim et al. [3] showed that aluminum electrodes were the most efficient for removing phosphorus as compared with iron and steel used stainless electrodes. However, concentrated contaminants in water attach to the surface of the electrode, resulting in the formation of scales. This causes economic disadvantages such as significant consumption of electrical power and the cost of replacing electrodes. This paper therefore presents a pretreatment method to minimize scaling of the electrode. Pretreatment to prevent scaling on the electrodes should be conducted so as to prevent an increase in resistance and electrical power; it should also address the corrosion issue. The use of TiO_2 prevents scaling and corrosion of the electrode, but it is not perfectly coated on metal electrodes [8]. We assessed this characteristic of TiO_2 using aluminum electrodes coated with 0.3% and 2% TiO_2 and aluminum electrodes with no titanium. The aluminum ions released from the uncoated part of the electrode helped remove phosphorus in the solution. On the other hand, the coated part prevented the formation of scales as a result of organics and inorganics attaching to the electrode. SEM images were used to analyze scaling on the surfaces of the electrodes.

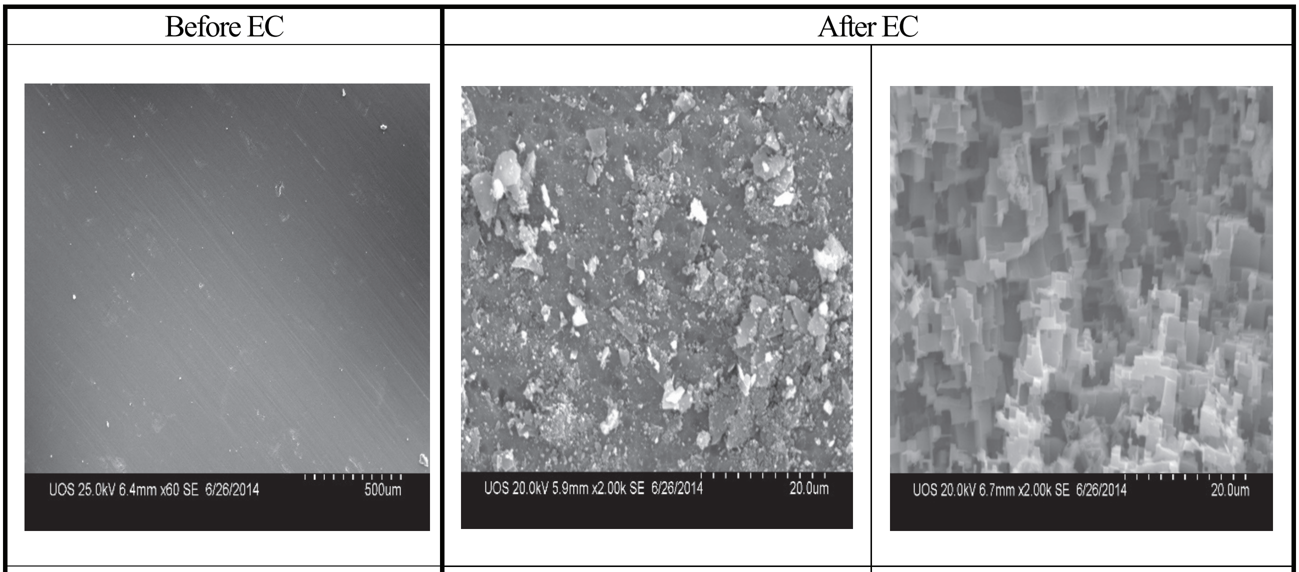
According to the SEM images, the TiO_2 coating before EC helped prevent scaling on the surfaces of the electrodes as shown in Fig. 3. Figs. 3(a) and (d) show the aluminum electrode and the coated electrode, respectively, before EC. On the cathode, larger amounts of organic and inorganic materials were attached on the aluminum electrode (Fig. 3(b)), compared with the coated aluminum electrode (Fig. 3(e)). In addition, on the anode, more serious corrosion occurred on the aluminum electrode (Fig. 3(c)) than the coated aluminum electrode (Fig. 3(f)). EDS chemically analyzes the element of electrodes. It shows the percentages of the remaining number of elements on the film by emitting the X-ray spectrum into the film. Therefore, EDS was used to analyze the scaling and corrosion of the electrodes during EC. The aluminum electrode and the TiO_2-coated aluminum electrodes were analyzed to determine the scale patterns generated on their surfaces.

Fig. 4 shows the surfaces of the two types of electrodes before EC. According to EDS, the aluminum electrode in Fig. 4(a) mainly consisted of aluminum, carbon, a relatively low amount of oxygen compared with the coated aluminum electrode in Fig. 4(b). Because of the TiO_2 coating, on the other hand, the coated aluminum electrode showed higher amounts of titanium and oxygen.

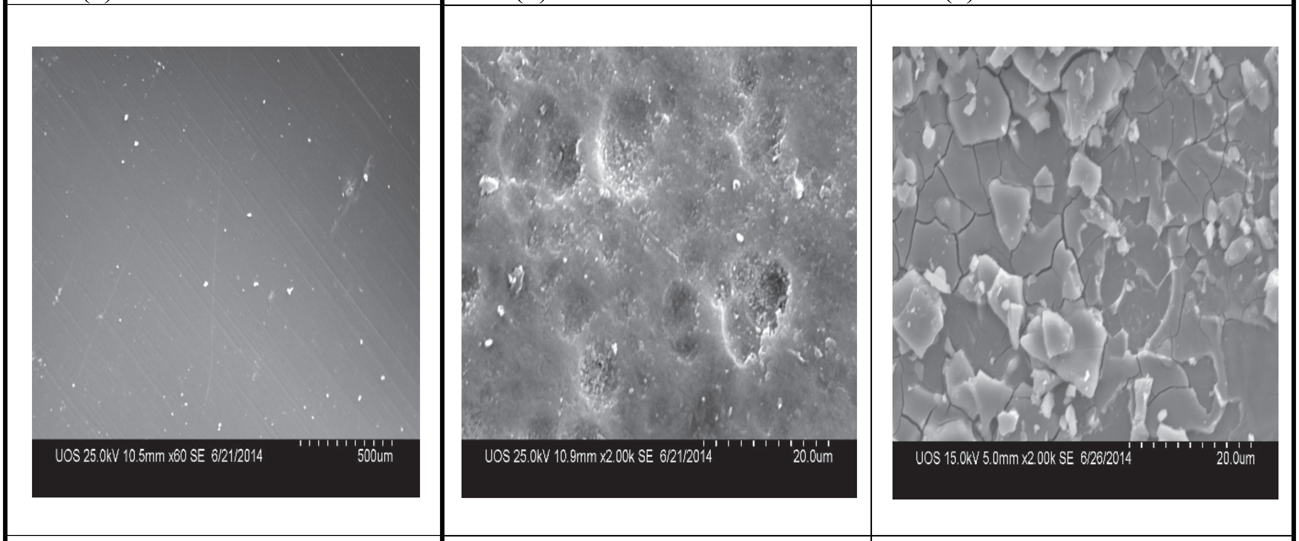
Fig. 5 shows a comparison of the surfaces of the aluminum and coated aluminum anodes after EC. The aluminum anode shows serious corrosion on its surface, particularly in the area marked as Spectrum 5 in Fig. 5(a), which was caused by pitting corrosion. The overall pattern of the corrosion is angular, which suggests that much of the corrosion occurred as a result of loss of the oxide film. The EDS images also show a higher amount of aluminum in the two areas marked as Spectrum 5 and Spectrum 6 than in the original. This shows that little oxide film remains.

After EC, the surfaces of the coated electrodes exhibited serious corrosion, although the degree of corrosion was somewhat less severe for the coated electrode than for the aluminum electrode. The difference between Spectrum 7 and Spectrum 8 in Fig. 5(b) is their compositional ratio of titanium and aluminum: Spectrum 7 had a higher amount of titanium, whereas Spectrum 8 had a higher amount of aluminum. The reason for this lies in the fact that the coating with TiO_2 helped Spectrum 7 elute less aluminum than Spectrum 8. As a result, this study demonstrates that TiO_2 is not perfectly coated on the metal electrodes, as cited by Jones [8], but it can reduce both partial corrosion of the electrode and the elution of aluminum, consequently extending the life of the electrodes.

Fig. 6(a) shows the EDS results for the aluminum cathode after EC. In Spectrum 7, the aluminum element decrease and oxygen element increase on the electrodes compared with



(a)Aluminum electrode (b) Aluminum cathode (c) Aluminum anode



(d)Coated aluminum electrode (e) Coated aluminum cathode (f) Coated aluminum anode

Fig. 3. SEM analysis of electrodes at magnification of ×2,000. (a) Aluminum electrode, (b) aluminum cathode, (c) aluminum anode, (d) coated aluminum electrode, (e) coated aluminum cathode, and (f) coated aluminum anode.

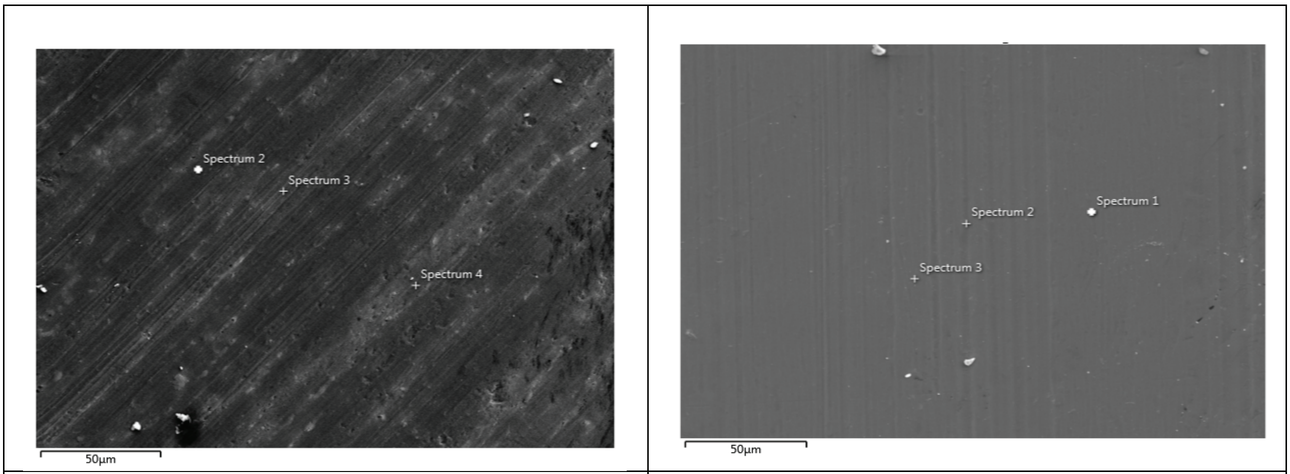
the electrodes before EC in Fig. 4(a). This was caused by the electrolysis reaction of water (Eq. (2)) at the cathode, which is composed of the remaining organics and nonorganics after removing the oxidation film on the surface. This suggests that Al was reduced on the electrode. Spectrum 8 shows the analysis results of the portion of the electrode without scales. According to EDS, this has a compositional ratio of elements similar to that shown in Fig. 4.

Fig. 6(b) shows the analysis results for the aluminum electrode coated with TiO_2 after EC. This electrode has a cleaner surface as compared with that shown in Fig. 4. According to EDS, the electrode contained aluminum, oxygen, and carbon only—in other words, it had no titanium. This shows that the reduction of the reaction on the surface and the removal of titanium from the electrodes prevented other substances from attaching to the electrode. As few scales are on the electrode, use of the coated aluminum electrode can decrease the electrical power required owing to the decrease in resistance. Consequently, this study demonstrates that the coated aluminum cathode can reduce the scaling on its surface and that the reduction reaction in EC can prevent scaling, thus maintaining its composition ratio of aluminum.

3.2. Microfiltration

3.2.1. Optimum conditions

Pressure is an important factor in the membrane process because it affects the flux volume [9]. Kim et al. [3] observed that the flux increased when the pressure difference was large, but sufficient flux was achieved at 2 kgf/cm^2 of pressure. Thus, considering the economic feasibility, a



(wt%)

	Spectrum 2	Spectrum 3	Spectrum 4
Al	73.5	72.4	74.1
Ti	0	0.1	0
O	1.6	2.6	1.8
C	24	24.8	24.1

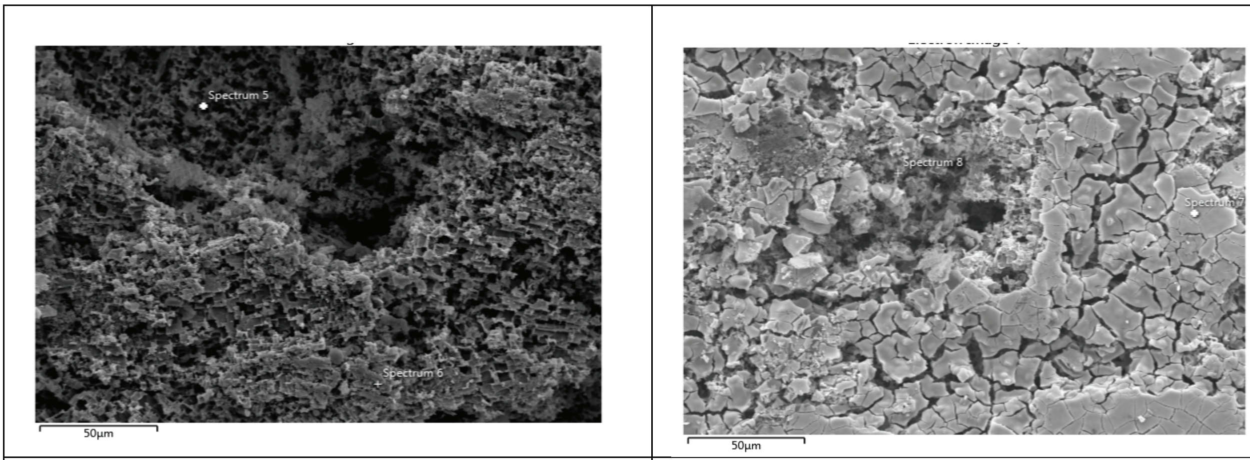
(wt%)

	Spectrum 1	Spectrum 2	Spectrum 3
Al	66.9	61.1	67.5
Ti	2.3	4.6	1.9
O	8.1	10.3	7.6
C	22.5	23.5	22.6

(a)Aluminum electrode

(b)Coated aluminum electrode

Fig. 4. EDS analysis of the aluminum and coated aluminum electrodes before EC. (a) Aluminum electrode and (b) coated aluminum electrode.



(wt%)

	Spectrum 5	Spectrum 6
Al	88.9	81.2
Ti	0	0.3
O	2.4	2.7
C	8.6	15.6

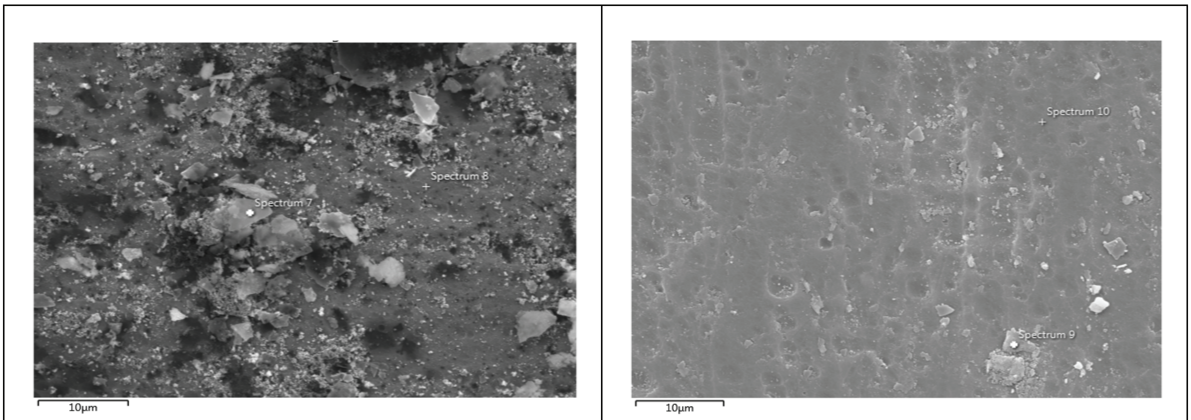
(wt%)

	Spectrum 7	Spectrum 8
Al	8.8	32.6
Ti	36.9	22.7
O	37.5	35.7
C	16.7	9

(a) Aluminum anode

(b) Coated aluminum anode

Fig. 5. EDS analysis of the aluminum and coated aluminum anodes after EC. (a) Aluminum anode and (b) coated aluminum anode.



(wt%)

	Spectrum 7	Spectrum 8
Al	43.8	73.6
Ti	0	0
O	31.1	2.9
C	24.5	21.6

(a) Aluminum cathode

(wt%)

	Spectrum 9	Spectrum 10
Al	65.8	79.2
Ti	0	0
O	11.4	1.8
C	22.4	18.9

(b) Coated aluminum cathode

Fig. 6. EDS analysis of aluminum and coated aluminum cathodes after EC. (a) Aluminum cathode and (b) coated aluminum cathode.

flux of 2 kgf/cm^2 was selected. Also, pH and temperature are important factors for the MF process. According to the advantages of EC, pH and temperature were maintained at the pretreatment conditions, that is, at a pH of 7 and a temperature of 15°C.

The experiment was conducted by using aluminum electrodes and aluminum electrodes coated with TiO_2. The results are shown in Fig. 7.

According to Fig. 7, treated water using aluminum electrodes was filtered faster than that by aluminum electrodes coated with TiO_2. When using aluminum electrodes, the greater amount of eluted aluminum resulted in the high removal efficiency of EC.

However, when backwashing MF, Fig. 8 shows that the aluminum electrodes coated with TiO_2 recovered faster than the ones without coating, as evidenced by the gap of 1,800 s between the filtration rates of the two types of electrodes.

3.2.2. Backwashing MF with clean-in-place

Treated water after EC includes numerous pollutants that contribute to membrane contamination, which blocks the pores of the film with microflocs. Thus, an experiment to identify reversible and irreversible contamination of the membrane was performed in the current study according to Raffin et al. [10]. Backwashing with distilled water was examined to verify reversible contamination. An irreversible experiment was conducted with CIP on contaminated membranes. CIP was conducted using 0.1 N HCl because the treated water after EC containing a number of micromaterials such as organics and inorganics should be implemented using an acid rather than a base to remove the contaminants from the film [10].

The flux presented an 85% recovery rate with backwashing. MF with CIP showed a 97% recovery rate as compared with the initial MF. Consequently, backwashing with CIP is more appropriate than backwashing using distilled water, as shown in Fig. 9.

3.3. Phosphorus removal and other contaminants

This study satisfied the recommended water quality standard of TP in the lake (≤0.05 mg/L for TP and ≤0.6 for total nitrogen [TN]). Both the aluminum electrodes and coated aluminum electrodes were used under two different conditions: one was with a molar ratio of Al to P of 5:1 for 0.2 mg/L of phosphorus in the lake and the other was with a ratio of 20:1, which achieved the highest removal efficiency, that is, 98.41% shown in Table 3 in section 3.1.1. The results are represented in Fig. 10.

As shown in Fig. 10, the removal efficiency of phosphorus in EC was much higher when Al:P of 20:1 was used. The EC molar ratio (Al:P of 5:1) showed removal of less than 80% of TP, which further increased up to 90% with the addition of MF. Using MF, the removal efficiency of phosphorus by molar ratio of 5:1 was slightly higher or similar to the case whose molar ratio was 20:1. Thus, in terms of economics, it is suggested that the concentration of TP in a lake should be assessed before applying the molar ratio of aluminum to phosphorus. The removal efficiency of phosphorus between the two types of electrodes (aluminum vs. TiO_2-coated aluminum) was similar. This suggests that the TiO_2-coated

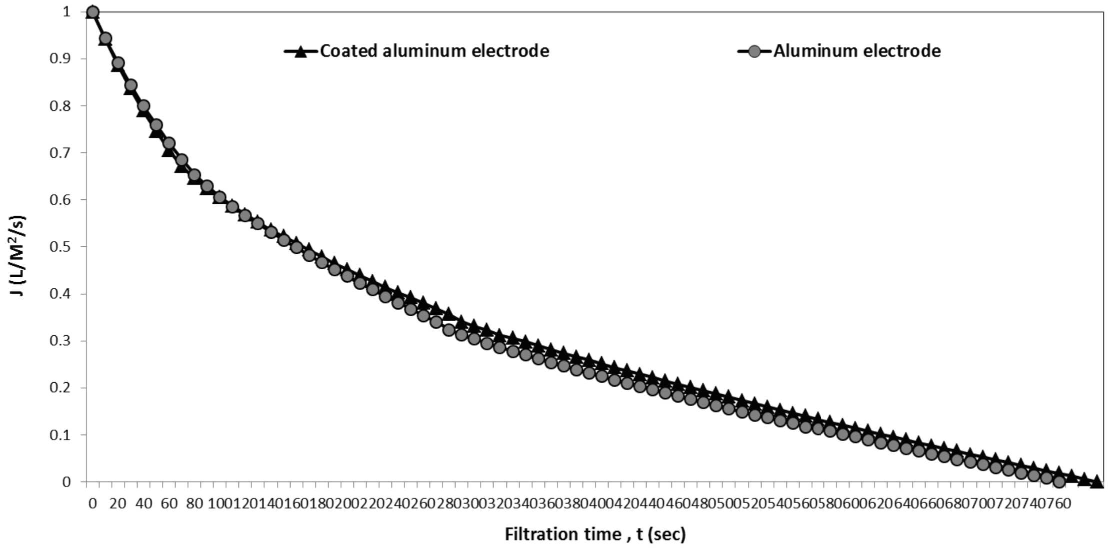


Fig. 7. Flux of MF process according to time using the two types of electrodes.

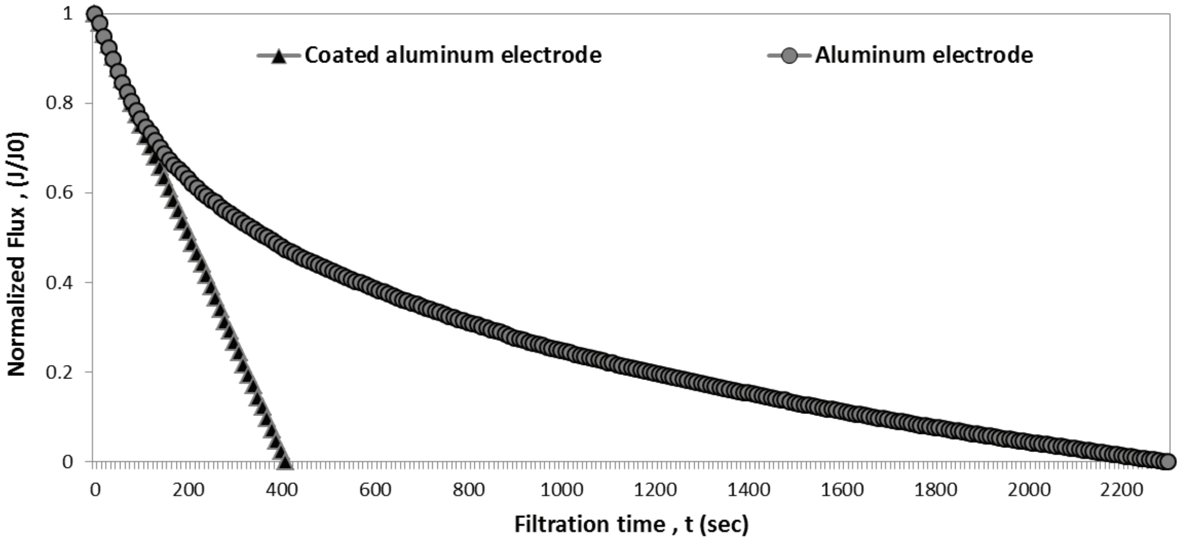


Fig. 8. Backwashing in J/J_o of MF according to time.

aluminum electrode, with decreased scaling on its surface, maintained its removal efficiency for phosphorus. Also, it was shown that in terms of electrical power, the aluminum electrodes coated with TiO_2 were more economical than aluminum electrodes without TiO_2.

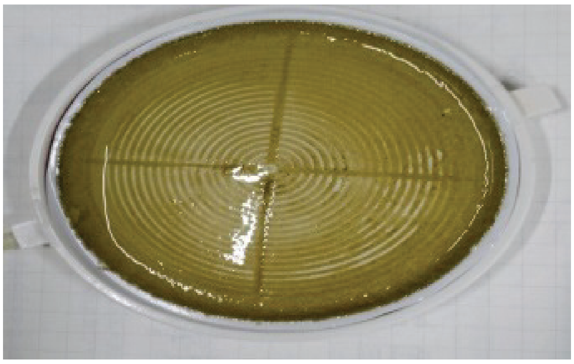
As shown in Fig. 11, with a molar ratio of Al:P of 5:1 for 0.2 mg/L of phosphorus in the lake, more than 80% of other pollutants (biochemical oxygen demand and suspended solid) were removed but less than 30% of TN was eliminated. The aluminum electrode showed lower removal efficiency of these pollutants except TN because the released titanium was dissolved in the raw water, not combined with other organics. However, using the coated aluminum electrode satisfied the recommended water quality standard for the lake.

Consequently, EC and MF are both highly efficient methods for eliminating phosphorus. To remove phosphorus, EC, with application of the molar ratio and TiO_2 coating of the electrode, is the best preprocess before MF in terms of cost and effectiveness to prevent fouling of MF.

4. Conclusions

The optimum conditions for the removal of phosphorus by EC included the following: an aluminum electrode with a 3 mm gap, a reaction time of 1 min, a temperature of 15°C, a molar ratio of Al:P of 5:1 for 0.2 mg/L of phosphorus in the lake, and a conductivity of 0.2 mM. The aluminum was coated with TiO_2 as a pretreatment, which helped prevent the occurrence of scaling on the electrode, and the Al^{3+} released

(a)

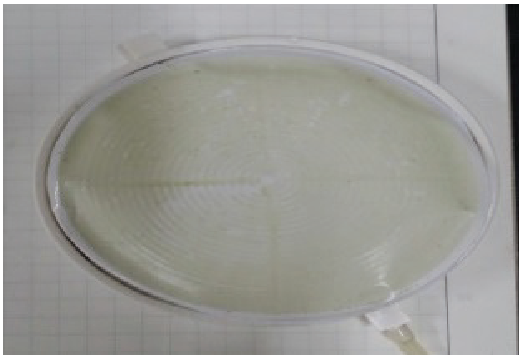


Before backwashing

(b)

After distilled water

(c)



After backwashing with CIP

Fig. 9. Comparison of films before and after backwashing. (a) Before backwashing, (b) after distilled water, and (c) after backwashing with CIP.

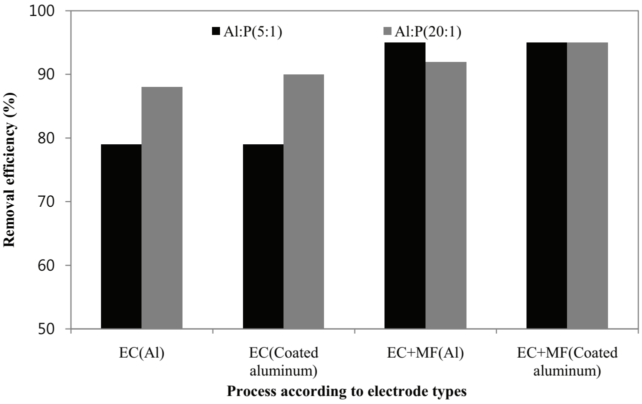


Fig. 10. Comparison of phosphorus removal using EC and MF according to molar ratio.

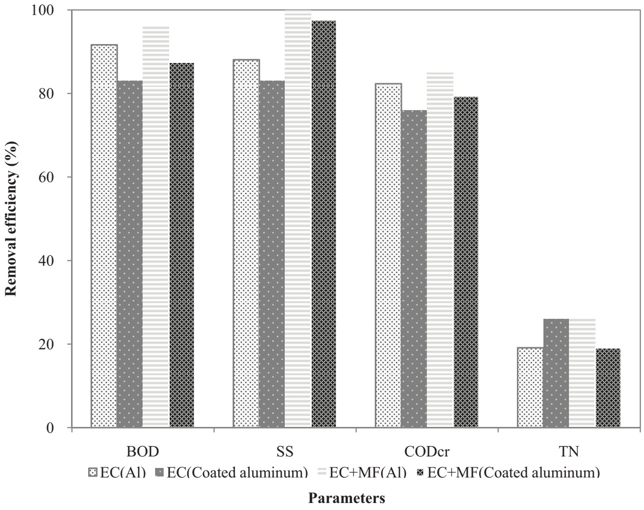


Fig. 11. Removal efficiency in terms of other parameters.

from the electrode helped remove phosphorus. The removal efficiency of phosphorus between the two types of electrodes (aluminum vs. TiO_2-coated aluminum) was similar, but the coated aluminum electrode showed decreased scaling on its surface while maintaining its phosphorus removal efficiency.

The conditions for implementing MF to secure stable water quality were as follows: 2 kgf/cm^2 pressure and 0 mV zeta potential. A recovery of 85% of the initial membrane was achieved by backwashing in distilled water, but CIP (0.1 N HCl) resulted in a 97% recovery. Thus, backwashing MF with CIP was considered to be appropriate for the recovery of the contaminated membrane. Consequently, EC and MF were both highly effective in the elimination of phosphorus, and their use satisfied the recommended water quality standard for the lake (<0.05 for TP and <0.6 for TN). EC, using the molar ratio and titanium coating, was considered to be the best pretreatment to prevent fouling of the MF in terms of its cost and effectiveness.

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