



Enhanced phosphorus and COD removals for retrofit of existing sewage treatment by electrocoagulation process with cylindrical aluminum electrodes

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ABSTRACT

A series of experiments were performed in a study designed to investigate the retrofit ability of removing total soluble phosphorus (sTP) and soluble chemical oxygen demand (sCOD_{Mn}) from wastewater through electrocoagulation technology utilizing cylindrical aluminum electrodes in batch-operating modes (BOMs) and continuous operating modes (COMs). By varying the operating conditions and the effects of various experimental parameters such as pH, NaCl concentration, hydraulic retention times (HRT), initial phosphorus concentration, and temperature, etc, the applied electric potentials (AEPs) range between 3, 4, and 5 volts (V), with current densities from 7.04 to 16.08 A/m² in BOM and from 7.48 to 21.69 A/m² in COM. Electrolysis times in the limits of 1 to 20 min were tried for different types of wastewater, including synthetic wastewater and municipal wastewater. According to experimental results, it was demonstrated that superior performance in removing phosphorus from wastewater can be achieved, with experimental data indicated that more than 99% (<0.2 mgTP/L) of phosphorus, and 75% (<10mgCOD/L) of sCOD_{Mn} can be removed using this method. Additionally, it was also found that TP and sCOD_{Mn} removal efficiencies were also increased with the addition of more NaCl to the wastewater. It has also been determined from the data that with optimum operating conditions and electrolysis time, this method can be used in existing municipal wastewater treatment plants to enhance treatment efficiencies of phosphorus and sCOD_{Mn} when properly retrofitted.

Keywords: Phosphorus removal; Electro-coagulation; Aluminum electrode; Sewage treatment

1. Introduction

The handling of phosphorus in wastewater using conventional biological treatment systems (CBTSs) has difficulties and limitations which make these treatments less effective. Phosphorus discharge standards

for municipal wastewater in all developed and developing countries have become increasingly stringent, while the phosphorus concentrations in final effluent using CBTSs can exceed more than 2 mgTP/L, creating an urgent need for a better mitigative technology. Thus, the need for innovation and applied advanced technology is needed to create

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efficiencies in phosphorus removal, using less space, lower capital investment; less installation; lower operating and maintenance costs, and eliminating the need for additional, frequent chemical use [1–4]. For these reasons and others, this research was carried out.

Using electricity to treat water was first proposed in England in 1889 [5]. The EC process using aluminum and iron electrodes was invented and patented in the USA in 1909. The first large-scale water treatment plant application of EC was first operated in the US in 1946 [5–7]. For a long time, electrochemical technologies in water and wastewater treatment were not implemented on a wider scale due to a lack of capital and greater operating costs. Nowadays, electrochemical technologies have reached such a state that they are not only comparable with other technologies in terms of cost but also are more efficient and more compact [5–8].

Phosphorus can exist in the environment in three main forms: orthophosphate, polyphosphate, and organic phosphorus, but the principal phosphorus compounds in wastewaters are generally orthophosphates [9]. Phosphate discharged into surface waters accelerates eutrophication and algal blooms in lakes, reservoirs, and ponds, which consequently leads to the depletion of oxygen levels as the algae decay. Reduction of dissolved oxygen levels can have harmful effects on fish and other aquatic life, causing reductions in biodiversity. Eutrophication can also affect the recreational value of natural resources [4,9,10]. Main sources of phosphate discharges are effluents from municipal or industrial plants, agriculture and animal husbandry [11].

The treatment of phosphate can be achieved by several physical, chemical, and biological methods [12]. In a biological treatment plant, it is necessary to transfer phosphate from liquid to the sludge phase, and the removal efficiency usually does not exceed 30%, which means that remaining phosphate should be removed by other techniques [4]. Absorption and chemical precipitation are among the physicochemical methods that have been widely used for phosphate removal. The removal of phosphate from aqueous streams requires the conversion of soluble phosphate into an insoluble solid phase. This solid phase can be separated from water by means of sedimentation or filtration [2,12]. EC involves the generation of coagulants in situ by dissolving electrically either aluminum or iron ions from aluminum or iron electrodes respectively. The metal ions generation takes place at the anode; hydrogen gas is released from the cathode. The hydrogen gas also helps to float the flocculated

particles out of the water. This process sometimes is called electroflocculation [11].

When aluminum is used as electrode material, the dominant reactions involved might be summarized as follows [4,13–15]:

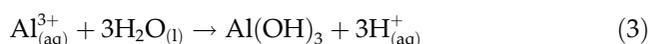
At the cathode:



At the anode:



In the bulk solution, multi-core coordination compounds and AlPO_4 , $\text{Al}(\text{OH})_3$ precipitates can be formed:



The objectives of this study were to (1) investigate the feasibility of phosphorus and COD_{Cr} removal from wastewater by EC using aluminum electrodes in batch operating mode (BOM) and continuous operating mode (COM); (2) determination of the reaction order, reaction rate constant for standard TP solution; (3) the efficiency of TP removal on initial concentrations of TP; (4) hydraulics retention times, and the effect of the current density upon them; (5) the optimal operating conditions for TP removal as well as determination of the reaction rate constant.

2. Materials and methods

2.1. Preparing wastewater

The municipal wastewater used in this study was collected from the Hybrid Pilot Plant–Advanced Sewage Treatment using the rotating hanging media bioreactor (RHMBR) and MBR combined system (Table 1), located in Yongin, South Korea. and the synthetic wastewater was prepared by mixing either distilled water or tap water with dissolved sodium chloride (NaCl , 99.5%, South Korea) and potassium dihydrogenophosphorus (KH_2PO_4 , 98%, South Korea). It required an initial concentration of phosphate solutions prepared by diluting the appropriate volumes of the stock solution (Table 1). At the start of each experiment, NaCl was added to adjust the conductivity to the equivalent level of conductivity found in domestic wastewater.

Table 1
The characteristics of municipal and synthesis wastewater used in experiments

Parameter	Units	Municipal wastewater		Synthesis wastewater		
		Average	STD	Sol. 1	Sol. 2	Sol. 3
pH		7.72	0.23	7.25	7.25	7.25
SS	mg/L	290	58.07			
BOD ₅	mg/L	199.11	10.01			
COD _{Mn}	mg/L	143.2	6.45	37.3–72.8		
T-N	mg/L	29.56	8			
T-P	mg/L	4.12	1.12	5	10	20
Conductivity	μS/cm	460	24.7	1,200–4,700		
Alkalinity	mgCaCO ₃ /L	169.19	19.11			

Table 2
Prediction of TP removal rate and effluent TP concentration

Electrolysis time (min)	TP removal rate (%)		TP concentration (mg/L)	
	4 V	5 V	4 V	5 V
2	76.7%	89.5%	1.16	0.50

2.2. Experimental

During the experiment, the temperature and pH were not altered much between 16.8 to 25.4°C and 6.5 to 7.8 (Fig. 3), respectively. Electrodes were connected in monopole mode to a dual, DC power supply (Sunchang Electronic Co., Ltd., South Korea) which includes (1) meters to monitor the voltage and current; (2) an on-off switch and a rheostat used to allow vary the desired output voltage; (3) in each channel of the dual power supply, there are digital voltage meters with a voltage response (0–30 V) monitor and current meter to set the applied potential and current level.

The creativity and innovation in this electrolysis device is evident in the use of an arrangement of a pair of aluminum electrodes. Their concentric cylindrical shape increases the upward flow rate.

2.2.1. Batch mode experimental

A flow diagram of the batch mode experimental system used in this study is shown in Fig. 1. The EC reactor consisted of a rectangular shaped holding tank, ($L \times W \times H = 15.5 \text{ cm} \times 12 \text{ cm} \times 19 \text{ cm}$, with a working volume of 3000 ml), that was made from polyacrylate and a pair of aluminum electrodes, cylindrically shaped and placed in concentric cylinders together (see Fig. 1(a)), with inside electrode dimensions of $ID \times H = 5 \text{ cm} \times 10 \text{ cm}$, and a geometric area of 157 cm^2 ; and outside electrode dimensions of $OD \times$

$H = 9.5 \text{ cm} \times 10 \text{ cm}$, with a geometric area of 298.6 cm^2 . The gap between the electrodes was 2.25 cm (see Fig. 1), and a stirrer M was set at a stirring speed of 150 rpm in order to generate high turbulence to increase contact between the aluminum ions and pollutants (especially for orthophosphate) in the electrolyte.

2.2.2. Continuous mode experimental

A flow diagram of the continuous mode experimental system used in this study is shown in Fig. 2 (a). Raw wastewater (including municipal wastewater or synthetic wastewater) was contained in 200-liter polypropylene tank; from this tank, wastewater was pumped continuously through the flow meter at various flow rates in an upward axial flow in an annular region between two coaxial cylinders of radius 5 cm and 9.5 cm in the EC reactor as shown in Fig. 2(b). After passing through the electrolysis device, the wastewater gravity flowed through the inline mixer in order to increase contact between the aluminum ions and pollutants (especially the orthophosphate) and flocculants formed combined and absorbed suspended solid particles in the wastewater.

The EC reactor in continuous experimental mode consists of a pair of aluminum electrodes, cylindrical shaped and placed in concentric cylinders together with inside electrode dimensions of inside of $ID \times H = 5 \text{ cm} \times 26.25 \text{ cm}$ and a geometric area of

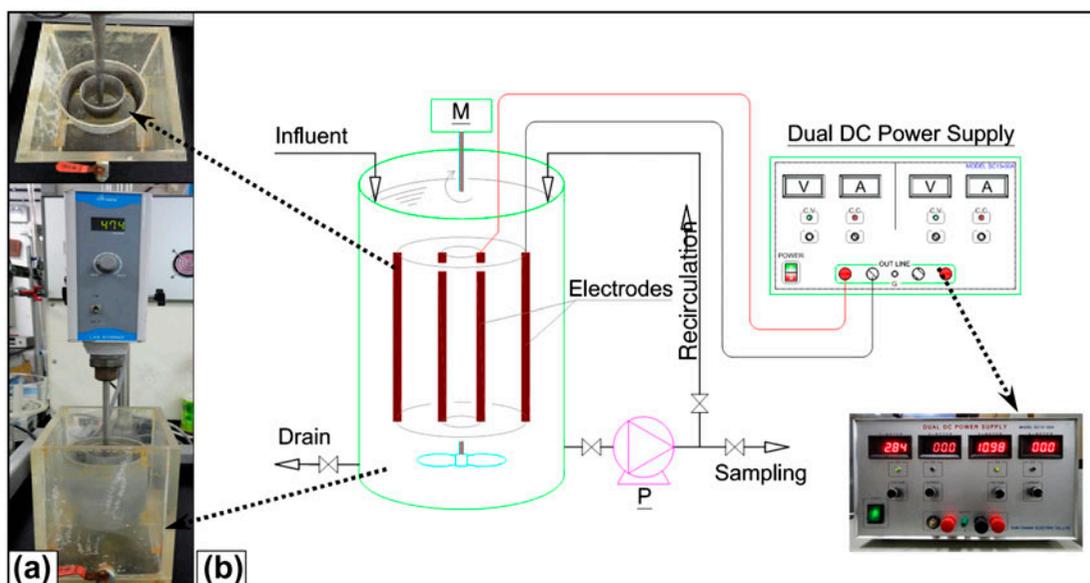


Fig. 1. Schematic diagram of the electro coagulation system for phosphorus removal in BOMs.

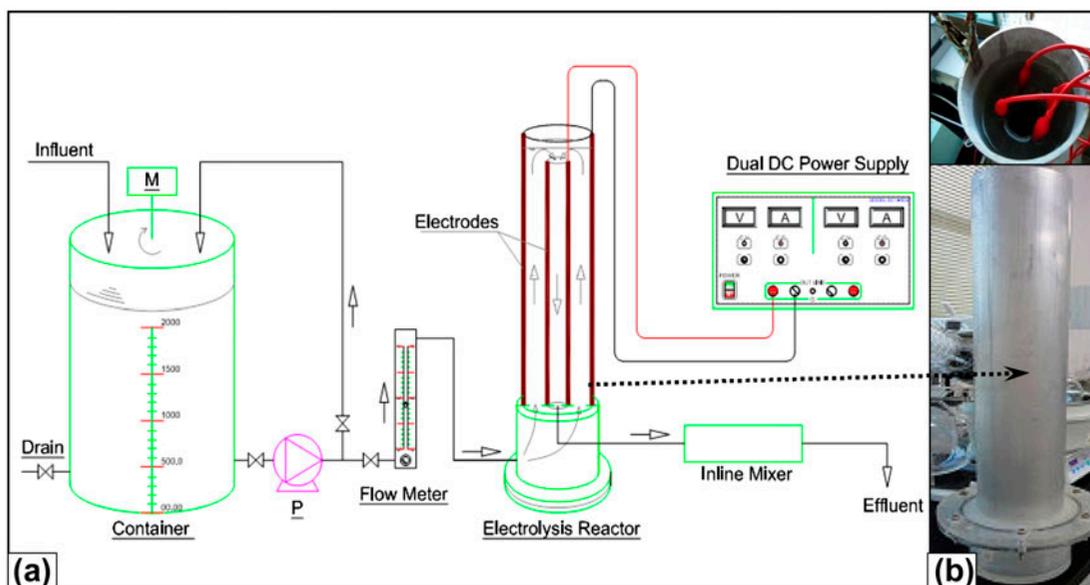


Fig. 2. Schematic diagram of the electrocoagulation system for phosphorus removal in continuous mode.

351.7cm², and outside electrode dimensions of OD × H=9.5 cm × 22.4 cm, with a geometric area of 783.4 cm²; but the working surface area was 668.52 cm². The gap between the electrodes was 2.25 cm (see Fig. 2(b)).

2.3. Analytical methods

During the experiments, the water quality was determined by measuring conductivity, temperature, pH, TP, and COD_{Mn}. All samples were analyzed

according to the standard methods [16]. The electrical conductivity of wastewater was measured by Conductivity Meter (ORION Model 130, Germany). TP and COD was analyzed by using a furnace reactor (Model HS-R200, Humas Co., Korea) and UV/visible Spectrophotometer (model HS-DR 3,300, Humas Co., Korea), the pH values and temperature were measured by using Fisher Scientific Accumet Excel XL60 (Accumet[®] XL60, Thermo Fisher Scientific Inc.). All samples were filtered using Whatman GF/C glass microfiber filters (Whatman, Germany) with a pore

size of 1.2 μm under vacuum pump before the analysis. Treated wastewater was collected over a desired period of time from the reactor, and also effluent of the reactor (continuous mode). And in these experiments, all the reagents were analytical grade and used without further purification. In addition, each sampling port valve was opened for 30 secs to 1 min prior to sampling to flush any contaminants present in the sampling lines.

3. Results and discussion

3.1. The effect of pH

The change of pH over electrolysis time for various NaCl concentrations, and initial TP concentration is presented in Fig. 3. The results presented in Fig. 3 show the variation of pH during electrolysis is not significant and is in a range near neutral pH. Thus, pH is not needed to be adjusted.

The cause of pH changes in the EC process is due to hydrolysis and polymerization reactions which form different complexes, such as polymer compounds, and also formed are $\text{Al}(\text{OH})^{2+}$, $\text{Al}(\text{OH})_2^+$, $\text{Al}(\text{OH})_2^{4+}$, $\text{Al}_{13}(\text{OH})_{32}^{7+}$, $\text{Al}(\text{OH})_3$, AlPO_4 [15], and chlorine ions present in the wastewater [6].

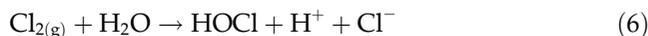
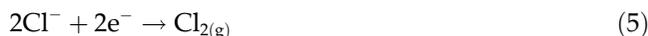
Theoretically, acidic and alkaline conditions during electrochemical oxidation could help organics removal. Acidic conditions significantly decrease the concentration of CO_3^{2-} and HCO_3^- , both well-known scavengers of OH generated on anodes, and so enhances oxidation. While alkaline condition boosts the $\text{Cl}^- \rightarrow \text{Cl}_2 \rightarrow \text{ClO}^- \rightarrow \text{Cl}^-$ redox circulation enhances indirect oxidation [17–19].

3.2. The effect of NaCl

The voltage and amperage are the key variables in controlling the efficiency of the EC process [20]. To increase the electrical conductivity in the water or wastewater to be treated, NaCl was added at different concentrations (see Fig. 4). The addition of NaCl leads to a reduction of phosphate treatment time because of a corresponding increase of the dissolution of the aluminum electrode, and NaCl concentration also has variable affects on electrical power consumption [20].

Moreover, [4,5,21–25] the electrolysis process also generated chlorine (Cl_2 , HOCl , OCl^-) (Eqs. (5)–(7)), hydroxyl radicals ($\cdot\text{OH}$) (Eq. 8), and peroxide (H_2O_2) (Eq. 9) were the agents during water disinfection.

Main reactions related to present of Cl^- ion are [4,25]:



Undergoes dissociation into ions



Generated hydroxyl radicals are [25,26]:



Oxygen dissolved in solution [22]

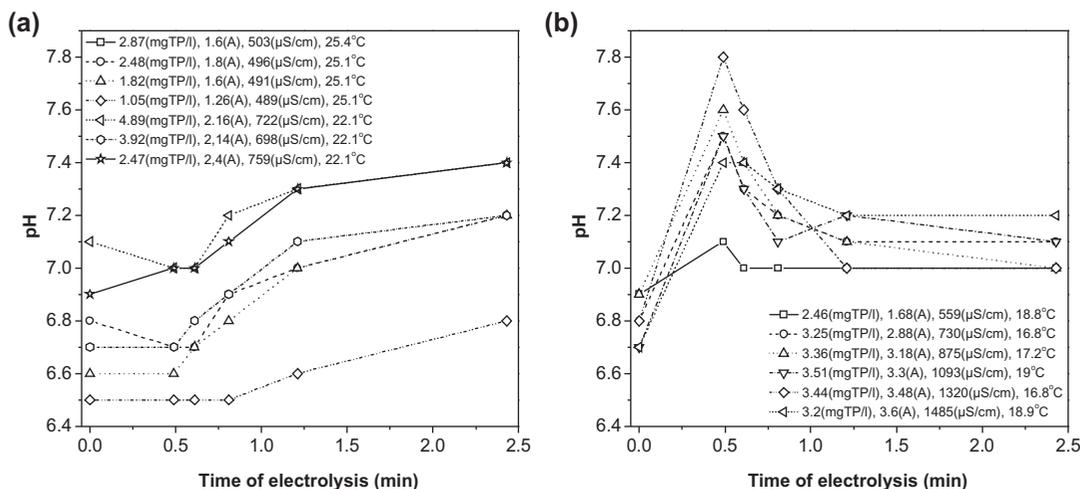


Fig. 3. Variation of wastewater pH with time, (a) synthetic wastewater and (b) real wastewater.

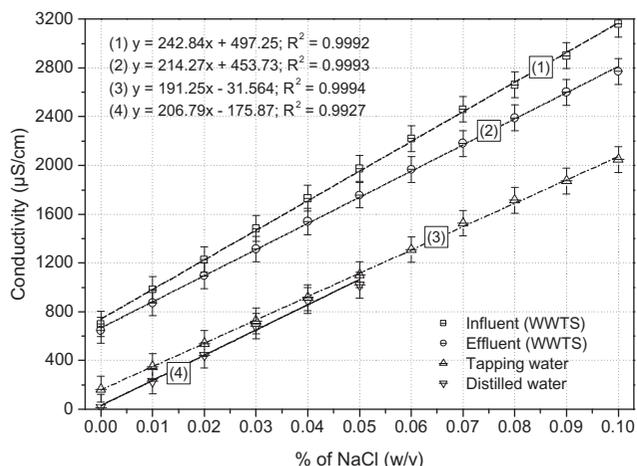


Fig. 4. Depending of conductivity as a function of NaCl concentration.

The variable conductivity of distilled water, tap water and influent and effluent from the hybrid municipal wastewater treatment pilot plant, which was the sample source, are as shown in Fig. 4.

As seen in Fig. 4, the obtained results presented a nearly linear relationship between conductivity of aqueous solutions and concentration of NaCl.

To study the effects of NaCl on removal of phosphate, the synthetic wastewater had been prepared with weight concentration of NaCl of 0.05, 0.1, and 0.2% and the initial concentration of TP was 20 mg/L. The conductivity of solutions was about 1,200, 2,000, and 4,700 µS/cm, respectively.

As it may be desirable to shorten the electrolysis time and to increase the electrolyte concentration (increase current density), the concentration of salt is

increased. This can increase the flow rate to reduce the operating costs, but result is faster electrode corrosion, and consequently, the electrode replacement cycle will be shortened [27].

Therefore, accordingly [13,20], the EC removal of TP can be simulated by formulae:

$$-V \frac{dC}{dt} = KAC \tag{10}$$

Eq. (10) can be integrated and then rearranged and modified to give the following:

$$\ln\left(\frac{C_t}{C_0}\right) = -\frac{k_1 I t}{r_2 - r_1} + C_1 \tag{11}$$

where k_1 : removal rate constant (cm/Ah), t : time (min), C_0 : initial concentration of TP at time $t=0$ (mg/L), C_t : concentration of TP at time t (mg/L), I : current (ampere, A), A : area of electrode (cm²), V : volume of cell (liter), r_1 : radius of a inside electrode (cm), r_2 : radius of an outside electrode (cm).

Then, Eq. (11) can be applied to removal of phosphate from wastewaters by EC.

The results are presented in Fig. 5 as a function of operating time. Fig. 5(b) illustrated the TP removal rate with various NaCl concentrations and linear regression fit, according to Eq. (11). From the slope of the linear equations, the values of the TP removal rate constant (k_1) are easily calculated.

The results showed that the TP removal rate constant is increased with concentration of NaCl due to the increase in electrical current, which was set at 0.2, 0.33 and 0.63 A, respectively. In terms of energy, the highest removal constant obtained was with a

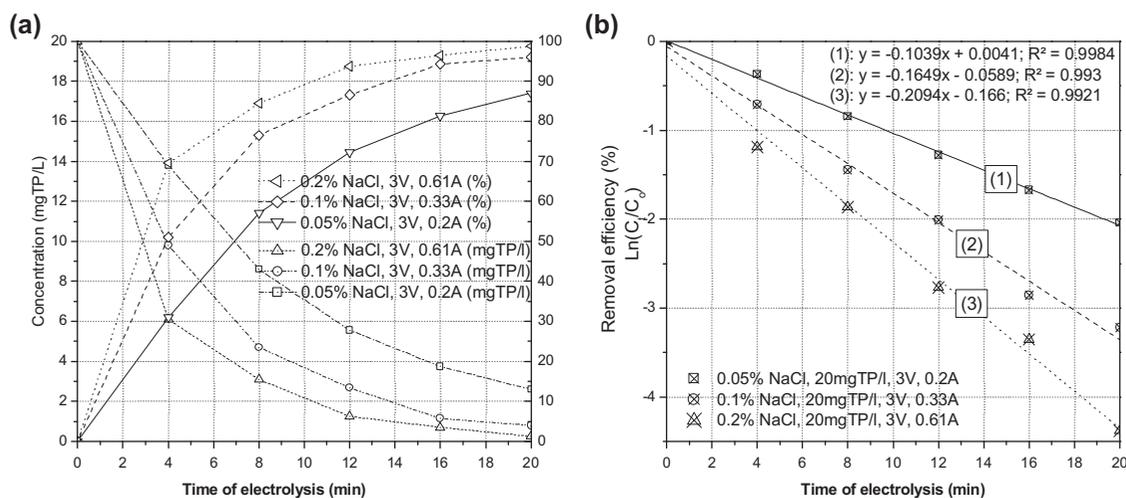


Fig. 5. Comparison TP removal versus electrolysis time for various NaCl concentration at 3 V in BOMs.

concentration of 0.05% NaCl. And in the same conditions, TP removal efficiency increased as the electrolysis time increased.

The current ratio was 0.2:0.33:0.63=1:1.65:3.15, and meanwhile, the conductivity ratio was 1,200:2,000:4,700=1:1.67:3.92, or the currents were not increased proportionally with the conductivity of the solutions.

3.3. The effect of applied electric potential

The synthetic wastewater with 0.05 (wt)% of NaCl and TP concentration of 20 mg/L had been chosen to study the effect of applied electric potential (AEP) on

TP removal rates. The obtained results are shown in Fig. 6.

The current settings were 0.23, 0.33, and 0.48A; or the ratio was 1:1.43:2.09. It increased more rapidly than the AEP the ratio of potential was 1:1.33:1.67.

The ratio of the TP removal rate constant was increased 1.35 times and 2.21 times when the AEP increased from 3 to 4 and 5V. The removal rate constant seems to be more proportional to the current setting than AEP.

Fig. 7 illustrates the TP removal rate for practical wastewater with variable applied potential and electrolysis time or flow rate. Fig. 7(b) shows that the results in the COMs are not similar to obtained results

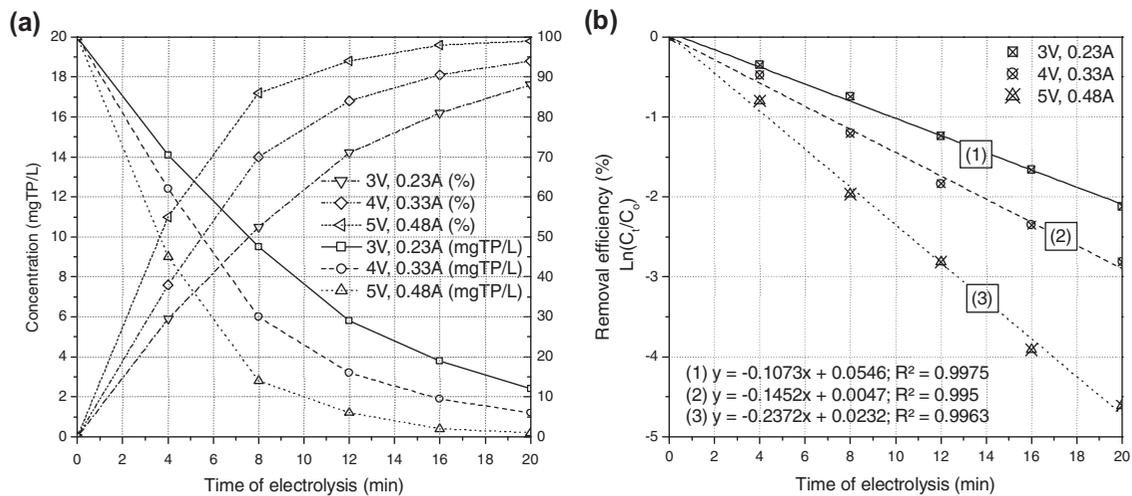


Fig. 6. Comparison TP removal versus electrolysis time for various AEP in BOMs.

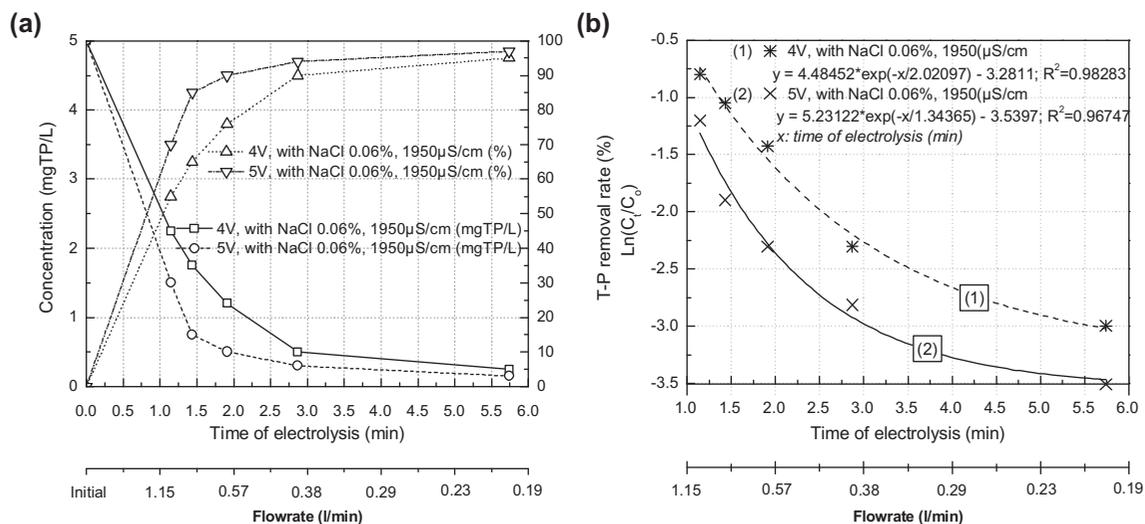


Fig. 7. Comparison TP removal rate and Ln(C_t/C₀) versus electrolysis time/flowrate for practical wastewater with initial TP concentration 5 mg/L and conductivity 1,950 μS/cm (NaCl 0.06%) in COMs.

in the BOMs. This means the relationship between $\ln(C_t/C_0)$ of TP concentration and electrolysis time in COMs was not followed in the first-order linear equations, but it was followed in the first-order exponential decay. And, with the same initial TP concentration, the time required for total removal of phosphorus in COM is much longer than in BOM.

According to the result shown in Fig. 7(a), under experimental conditions with practical wastewater in COMs, TP removal efficiency followed first order exponential decay.

Obtained results in BOMs and COMs showed the TP removal efficiency increased directly proportional to an AEP, electrolysis time, and concentration of dissolved NaCl.

3.4. The effect of initial TP concentration

To demonstrate the effect of initial TP concentration and time of electrolysis to the effective operation of the EC process, the studies were carried out with synthetic wastewater containing 0.05% NaCl, and with different initial TP concentration of 5, 10 and 20 mg/L. Fig. 8 illustrates the obtained results when AEP is 3, 4, and 5 V. The TP removal rate increased when AEP was increased, while initial TP concentration was decreased as a result. The TP also could be removed

totally after 20, 16, and 12 minutes, in respect to AEP of 3, 4 and 5 V with initial TP concentration of 5 mg/L.

3.5. The effect of co-coagulation and flocculation

Besides TP, the sewage contains other organic pollutants which may express as COD. The practical wastewater from the T-sewage plant had been tested with variable AEPs. NaCl was added to obtain conductivity of about 2,000 $\mu\text{S}/\text{cm}$. The results in Figs. 9–11 illustrate that the EC process is capable of removing TP and sCOD_{Mn} .

From the results of the relationship between TP removal efficiency and electrolysis time (contact time) for the case of BOM complicit with first-order reactions, and for the case of COM complicit with first-order exponential decay, shows that the effectiveness of different treatments between different treatment regimes (BOMs and COMs).

Per results shown in Fig. 8, the TP may be removed totally after 10, 8, and 6 minutes, corresponding to AEP of 3, 4, and 5 V, respectively, base on initial TP of 5 mg/L.

In real wastewater, some part of TP is associated with organic matter in dispersed particle form. In EC process, the main reaction accomplished is phosphate

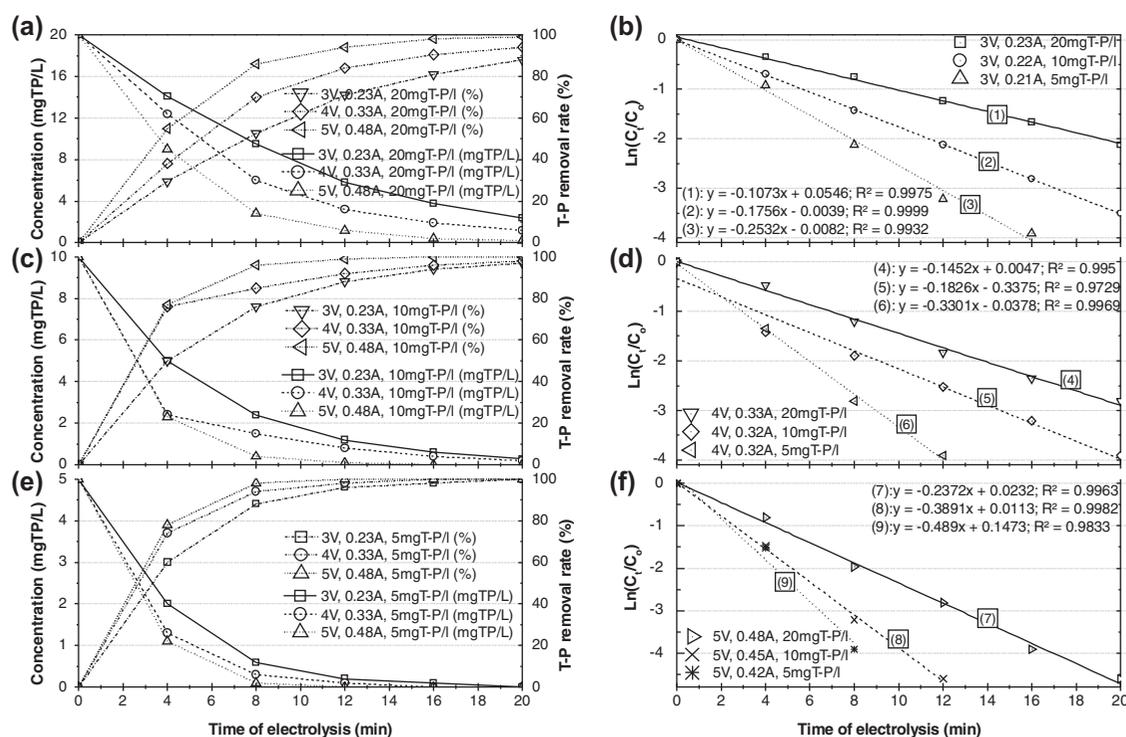


Fig. 8. Comparison TP removal versus electrolysis time for various initial TP concentration and AEP 3, 4 and 5 V in BOMs.

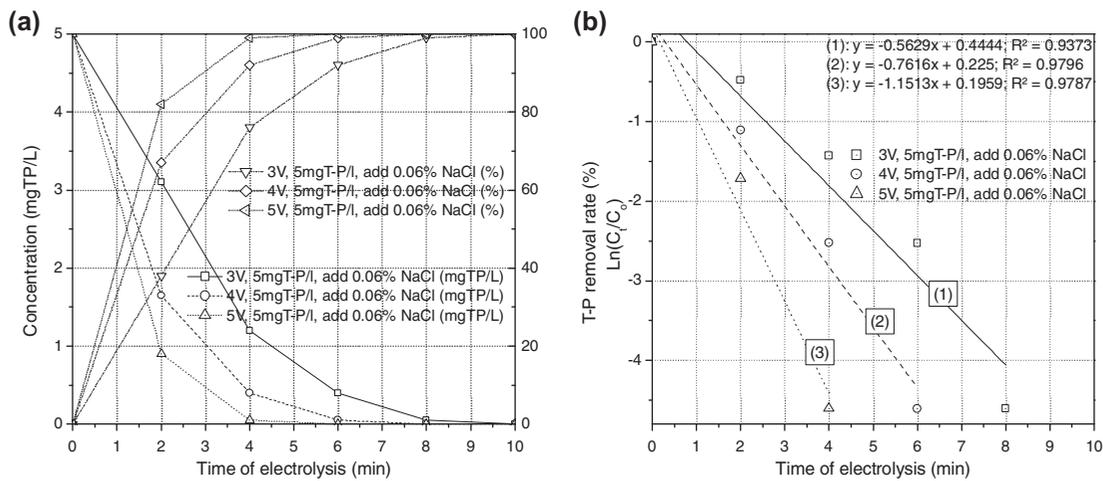


Fig. 9. Comparison TP removal versus electrolysis time for practical wastewater with various AEPs in BOMs.

removal accompanied by the precipitation of $Al(OH)_3$ and $AlPO_4$, although the $AlPO_4$ precipitation is favored over $Al(OH)_3$. The precipitation of $Al(OH)_3$ creates precipitation of dispersed particles that leads to increases in the TP removal rate. The $sCOD_{Mn}$ reduction with time is the evidence of this effect. The $sCOD_{Mn}$ removal rate by EC was not high: a decline in $sCOD_{Mn}$ from 89 to 50 mg/L after 2 min at AEP 5 V as shown in Fig. 10.

The results shown in Figs. 10 and 11 illustrate variable $sCOD_{Mn}$ removal rates with electrolysis times, and contaminants removal efficiency has increased with an increase in AEP, with the best result being 75%. The $sCOD_{Mn}$ removal rate in BOM and COM are similar and obtained results follow first-order exponential decay equation.

According to results [5], the optimum current density of EC for a long period of time without maintenance is in a range of 20–25 A/m². Based on

these obtained results, we can estimate optimal operation in continuous mode for a combination of TP and $sCOD_{Mn}$ removal for the T sewage plant (Table 2).

We can conclude that the optimal operation is about two minutes of electrolysis at AEP 4V. In this case, the current density is about 16 A/m², with an efficiency of $sCOD_{Mn}$ removal of about 75%, with an effluent concentration of TP of about 1.15 mg/L.

From all the results presented in Figs. 4–11 above, it can be observed that effective contaminants (TP, COD_{Mn}) at the beginning of the EC process are higher and faster than at the later time. The mechanisms responsible for this enhanced EC process that generated iron hydroxides from the corrosion of the anode, form complexes with ions occurring in the wastewater, this results in charge neutralization and possibly creates sweep flocculation by the precipitated metal hydroxide complexes, and colloidal particles formed

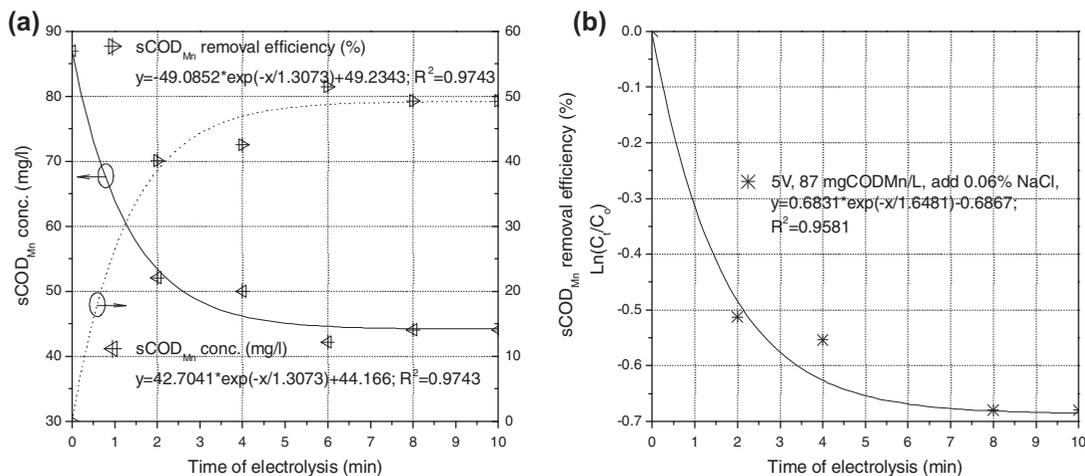


Fig. 10. Variation of $sCOD_{Mn}$ removal versus electrolysis time for practical wastewater at applied potential 5 V in BOM.

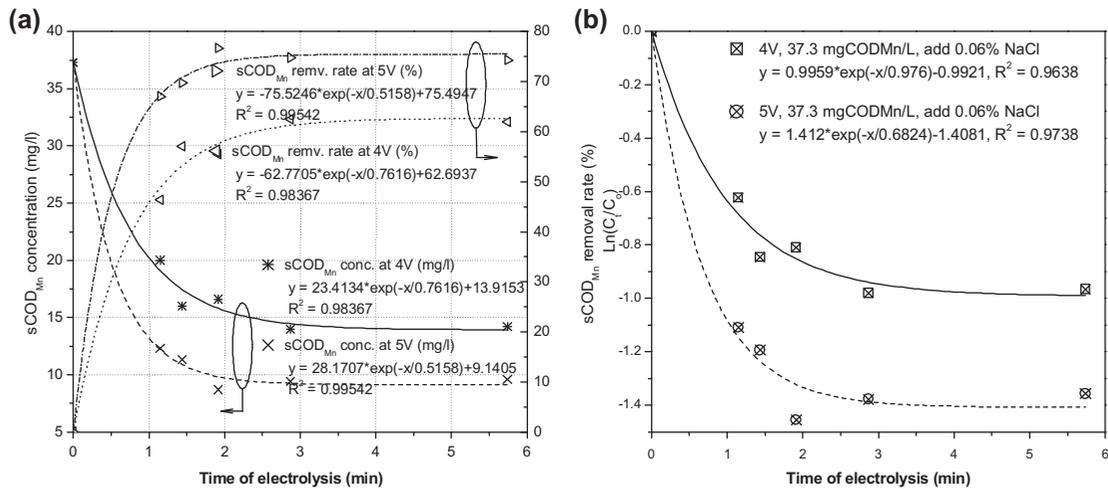


Fig. 11. Variation of sCOD_{Mn} removal versus electrolysis time for practical wastewater at applied potential 4 V and 5 V in COMs.

large aggregates, therefore rapidly removing contaminants [28].

3.6. The effect of electrolysis time and voltage on specific energy consumption and specific aluminum consumption

The variation of specific energy consumption (SEC) and specific aluminum consumption (SAC) and removal efficiency of TP increases proportionally to the electrolysis time and voltage in one operational condition. And from Fig. 12(a) and (b) we can

during 20 min and 6 min electrolysis time in BOMs and COMs, respectively at differential initial TP concentration and voltages and NaCl concentration were shown in Fig. 12.

The results indicated that the SEC, SAC, and removal efficiency of TP increases proportionally to the electrolysis time and voltage in one operational condition. And from Fig. 12(a) and (b) we can

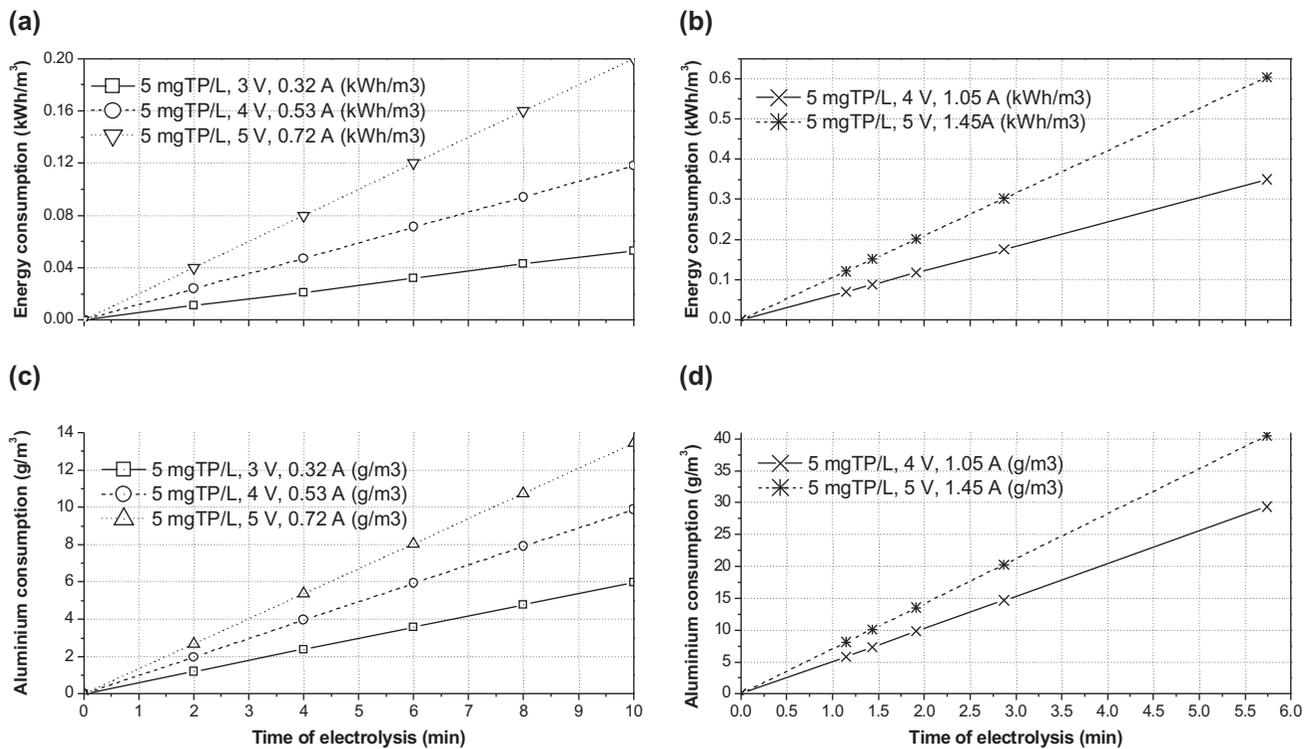


Fig. 12. Variation of specific energy consumption (a, b) and specific aluminum consumption (c, d) versus electrolysis time for practical wastewater in BOMs (a, c) and COMs (b, d).

calculate the energy consumption cost per cubic meter of wastewater, and from Fig. 12(c) and (d), we also calculate the amount of aluminum used per cubic meter of wastewater. From this, it is possible to determine the working time of the electrodes. This would enable operators to have a predictable plan for replacement of used electrode. For example, municipal wastewater with the initial TP concentration of 5 mg/L had a 96% efficiency (final TP effluent 0.2 mgTP/L) in operating conditions using 4 or 5 V, and adding NaCl 0.06% to the wastewater. For BOM, at least 3.8 and 5.7 min (Fig. 9(a)) of electrolysis time was needed, the corresponding specific energy consumption was 0.076 and 0.067 kWh/m³ (Fig. 12(a)), and then the corresponding to specific aluminum consumption was 5.101 and 5.632 g/m³ (Fig. 12(c)), respectively. And for COM, at least 3.3 and 5.1 min (Fig. 7(a)) of electrolysis time was needed. The corresponding to specific energy consumption was 0.347 and 0.311 kWh/m³ (Fig. 12(b)), and then the corresponding to specific aluminum consumption was 23.312 and 26.089 g/m³ (Fig. 12(d)), respectively.

4. Conclusions

The results demonstrated that the EC processes provide high TP removal from aqueous solutions. TP can be removed completely from wastewater by using this EC process, but for the effective removal of COD, post-treatment or pretreatment is needed.

The optimum conductivity of wastewater for EC of TP fluctuates around 2,000 $\mu\text{S}/\text{cm}$ and the reaction time for TP control, in practice, fluctuated around two minutes. The relationship between $\ln(C_t/C_0)$ of TP concentration and electrolysis time in the COM does not follow in the first-order linear equations, but it is followed in the first order exponential decay, and in the BOM, the first-order linear equations are also followed. The relationship between $\ln(C_t/C_0)$ of COD concentration and electrolysis time in BOM and COM are similar and obtained results following first order exponential decay equations.

The optimum EC for TP removal for the T-sewage plant sample using aluminum electrodes in continuous mode was two minutes of hydraulic retention time at an applied potential of 4 V.

The activity of the anode can decrease over time due to the existence of ions such as Ca^{2+} , Mg^{2+} , NH_4^+ , HCO_3^- , SO_4^{2-} , etc., in wastewater because of the precipitation of ions or the formation of insoluble hydroxides or sludge layers on the surface of the electrodes. These insulating layers on the surface of the electrodes cause reduced amperage and the

needed anode electrode dissolution in the electrolytic solution [4,5]. And the EC was designed in this study can be avoided by these above concerns.

Results showed that processing efficiency for TP, and COD is directly proportional to the AEPs, electric current, concentration of dissolved salts, electrolysis time, SEC and SAC and is inversely proportional to the initial TPs and COD_{Mn} . Understanding such features of the process allow for versatility in wastewater treatment plant design, installation, and operation.

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