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Modified electrocoagulation process with porous nonwoven partition for advanced phosphorus control

Sungyoun Kim, Kwang Ho Ahn, Kwang-Soo Kim*

Environmental Engineering Research Division, Korea Institute of Civil Engineering and Building Technology, 283 Goyangdae-Ro, Ilsanseo-Gu, Goyang-Si, Gyeonggi-Do 411712, Korea, Tel. +82 31 910 0299; Fax: +82 31 910 0291; email: kskim@kict.re.kr (K.-S. Kim)

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ABSTRACT

Modified electrocoagulation (EC) process was applied in the treatment of phosphorus in municipal wastewater through laboratory and pilot-scale experiments. Porous nonwoven partition was equipped between electrodes in the EC system to separate chemical reactions in anode and cathode. The modified EC system consisting of partitioned electrocoagulation tank, slow-mixing tank, and membrane filtration tank was investigated to treat municipal wastewater from first and secondary settling tanks. Analyses of the water quality in partitioned cathodes and anodes and the following treatment tanks showed that each electrode provided different reaction conditions in pH, SS, S-P, and T-Fe concentrations efficient for the treatment of soluble P and suspended solids. Comparative SS levels in the treated waters from conventional and partitioned EC system showed 49–63% decrease in sludge production in the partitioned EC, and the S-P removal efficiency was improved from 22% in conventional EC to 94% in the modified EC system.

Keywords: Cathode and anode; Electrocoagulation; Electrode passivation; Fe dissolution; Nonwoven partition; Phosphorus removal

1. Introduction

Electrical coagulation (EC) is a technology to remove contaminants by applying electrical current in drinking or wastewaters without addition of chemical coagulant. The process requires number of conductive metal plates, namely electrode, connected to electric source to produce positive metal ions during the reaction in anodes, to precipitate with negatively charged contaminants in wastewaters [1,2]. The performance of electrocoagulation can be impacted from electrode material, and researches have investigated the effect of individual electrode materials in several treatment studies. Among these, aluminum and iron appeared as the most common metal plates used in electrocoagulation process [3–5]. There is wide range of contaminants which can be treated by EC process, including heavy metals as oxides, suspended and colloidal solids, oil emulsions, fats, oils and greases, complex organics, as well as bacteria and viruses [6]. Removal mechanism of EC is generally explained as the production of aluminum or iron hydroxide flocs by the metal ions dissolved from anode electrodes followed

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*Corresponding author.

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by the reaction with contaminants at the anodes and the same time by the hydrolysis at cathodes. During the EC treatment, hydrogen gas and hydroxide ion (OH^-) are produced by water hydrolysis in cathode electrodes, resulting in the additional precipitation by reacting hydroxide and the metal ions dissolved from cathodes. This may deteriorate entire EC performance by interfering contact between contaminants and metal ions, and also by adding unwanted sludge production [7]. Several researchers reported that addition of sodium chloride could prevent the anodic passivation as the chloride ions enhanced the dissolution of metal ions so that it contributed to the formation of contaminant flocs [8,9]. The voltage applied for the electrolysis is another important variable in the anode reaction, and this is mainly dependent on the current density, conductivity of the wastewater to treat, the distance between electrodes, and the electrode surface state [10,11]. It is well understood that the increase in voltage on EC system has positive effect on the removal efficiency, as the higher amount of metal ions in anodes promotes the destabilization of pollutant molecules. Despite the simple chemistry involved in the mechanism, high treatment performance for the variety of contaminant type, the EC technology has never been employed as a major water treatment process over the years, and this is mainly due to the electrodes passivation over time. In order to alleviate the limitation, this study suggests partial separation in the chemical reactions taking place in the anode and cathode by applying partition in between the electrodes. Porous nonwoven media were equipped in the space between individual electrodes to partially separate the chemical reactions in anodes and cathodes, expecting that OH^- ions by water hydrolysis are kept in the cathodes and do not react with metal ions in anodes. In this study, Fe(III) ions dissolved from the anode were intended to solely form iron phosphate, but not to produce metal hydroxide, and thus can be used for longer periods. Decline in the sludge production by unwanted metal hydroxide floc is another beneficial result. This paper shows the result of laboratory and pilot-scale experiments to treat municipal wastewater by cathode and anode partitioned EC plant for soluble phosphorus removal.

2. Materials and methods

The performance of modified electrocoagulation process was investigated by laboratory and pilot-scale experiments, each by batch and continuous types. In every experiment, pH, SS, soluble phosphorus (as PO_4^{3-}-P), and total Fe levels are analyzed by pH meter (Model 915PDC, ISTEK, Inc.), Standard Methods [12], and spectrophotometer (HACH DR-4000), respectively.

2.1. Lab-scale experiment

For the preliminary test, batch-type EC process was operated. Influent water was taken from the first and secondary settling tanks in Il-san municipal wastewater treatment plant, Gyeonggi, Korea. Hydraulic loading rate was controlled as 10, 5, and 3 min, with other parameters fixed as 4 cm electrode distance, 30 V/0.6 A electric current, and 1,000 μS conductivity. Influent P concentration was 5 and 10 mg PO_4^{3-}-P/L.

The EC system for continuous test was composed of three parts: electrocoagulation, slow-mixing tank, and settling for discharge (Fig. 1). Working volume of the electrocoagulation tank was 2 L, equipped with iron electrodes of 100 mm × 100 mm × 2 mm. Electrical voltage was controlled from 1 to 30 V. The distance between cathode and anode was 20 mm, and the two electrodes were partitioned by nonwoven fabric of same size as electrodes in the center. Fine bubble blower was installed in the EC tank to create complete mixing condition in part for the phosphate in wastewater and iron ions from anode electrode. After treated by electrical coagulation, the wastewater was mixed slowly for complete coagulation and discharged after settlement. The identical lab-scale EC reactor without nonwoven partition was operated as control, and the results were compared each other. All the lab-scale experiments were carried out at room temperature.

2.2. Pilot-scale experiment

Batch-type pilot-scale experiment was carried out in Il-san municipal wastewater treatment plant, with the volume of 1.7 m^3 electrocoagulation tank treating first and secondary settling tank effluent. Levels of pH, SS, S-P, and T-Fe were monitored along with time passage. The surface area of each electrode was 1.4 m^2, as installed in 40 mm distance. Each electrode

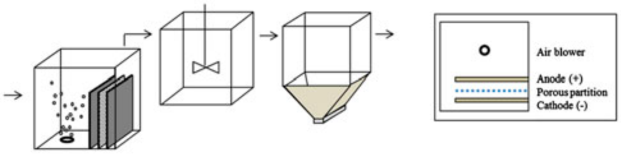

Fig. 1. Lab-scale EC plant consisted of partitioned EC tank, slow-mixing, and settling tanks. Details of EC electrodes and partition are illustrated in box.

was connected to a fixed 20 V with 10 A of electric current. In between the cathode and anode, nonwoven partition was applied as same manner in the lab-scale experiment.

Continuous-type pilot-scale EC plant was composed of influent storage tank, electrocoagulation tank, slow-mixing tank, and hollow fiber filtration tank for effluent. The slow-mixing tank was employed to enlarge the floc size after iron phosphate was formed in EC tank, and the filtration unit was applied as a post treatment, to completely remove remaining suspended solids. Hydraulic loading rate of the plant was 100 m^3/d, and the HRTs of EC, slow-mixing, and filtration tanks were 5–20, 20, and 20 min, respectively. The volume, electrode size, distance, and the partition of the EC reactor were exactly same as the batch test. Individual volumes of slow-mixing and filtration tanks were identical to 1.7 m^3.

3. Results and discussion

3.1. Laboratory-scale experiment

Fig. 2 shows the result of batch-type EC operation, with and without nonwoven partition between anodes and cathodes. Water samples were collected separately at cathodes and anodes in the case where the electrodes are partitioned by nonwoven medium. In the conventional EC reactor without partition, increase in pH was minor and sustained in neutral to slight alkaline ranges. This is because the OH^- ions from cathode are consumed by the reaction with Fe ions. Soluble P removal occurred with time passage, as a result of the reaction with Fe(III) dissolved from anode. The increase in the total Fe indicates that dissolution of the iron plate took place in anode. The metal dissolution results in the increment in SS level, either by metal pollutant combination or metal hydroxide formation. The buildup of SS concentration in the conventional EC reactor is attributed by phosphorus removal as well as iron hydroxide formation, and this is clearly compared with the SS level in the partitioned EC result. In the right-hand side, comparison in the same parameters from electrode partitioned EC experiment is provided. The pH values in anode (+electrode) and cathode (−electrode) are distinguished in this case, and this is because the porous partition in between the electrodes was successfully hampered the traveling of OH^- ions from cathode to anode. As water hydrolysis took place in cathode, accumulation of OH^- ion contributed the pH rise, while the pH in anode sustained or even slightly lowered. The low pH condition in anode is beneficial for the dissolution of iron and also for the formation of iron phosphate flocs. As OH^- ions are isolated from iron by the partition, unwanted iron hydroxide formation can be prevented and this is an additional benefit for the treatment process. Soluble P removal was more efficient in anode as expected but also occurred in cathode. Dramatic increase in the total Fe indicates that iron dissolution took place actively in anode, but did not consumed by OH^- from cathode, due to the porous partition between the two electrodes. The iron ions can be solely used for phosphorus removal so that the removal efficiency and the life span of the EC system can be improved. As the reaction between iron and OH^- is prevented, there should be decrease in the sludge production, and this is shown in the right-side SS graph. After 30 min operation of the batch-type EC systems, comparison in the effluent quality between conventional and partitioned EC was made, and it revealed that partitioned EC produced better quality effluent than conventional, containing maximum 63% less sludge, and the S-P removal efficiency was improved from 22% in the conventional to 94% in the partitioned EC.

Fig. 3 shows the relation between iron mass dissolved from the electrode and the electrode surface area, and the electric voltage supplied. For the relation between electrode surface and iron dissolution, electric voltage was supplied at fixed 10 V and the electrode surface was controlled from 20 to 150 cm^2. The iron dissolution increased with time and showed positive relation with electrode surface area. The velocity of iron dissolution for unit electrode surface area is found to be 0.02 mg Fe/min cm^2. Using a 100 cm^2 electrode, dissolved iron mass was monitored with increasing electric voltage from 5 to 30 V. The velocity of iron dissolution by electric voltage was found to be 0.3 mg Fe/min V.

Tables 1 and 2 list the wastewater quality parameters as compared between the conventional EC performance and the partitioned EC found by lab-scale continuous test. In Table 2, it is shown clearly that phosphate was efficiently precipitated with iron so the removal efficiency was high, and the SS was dramatically lowered by blocking the contact between the iron and OH^- ions. In the effluent of conventional EC-treated water (Table 1), formation of $Fe(OH)_3$ was even noticed by the reddish color. In Table 2, lowered pH condition in the anode also provided good environment for the precipitation of $FePO_4$. In the conventional EC plant, dissolution of iron was active and showed high concentration values, but did not used efficiently in the removal of phosphate. This resulted in higher iron as well as phosphate concentrations in the settling tank. In the partitioned EC plant, water quality parameters show that P removal condition was

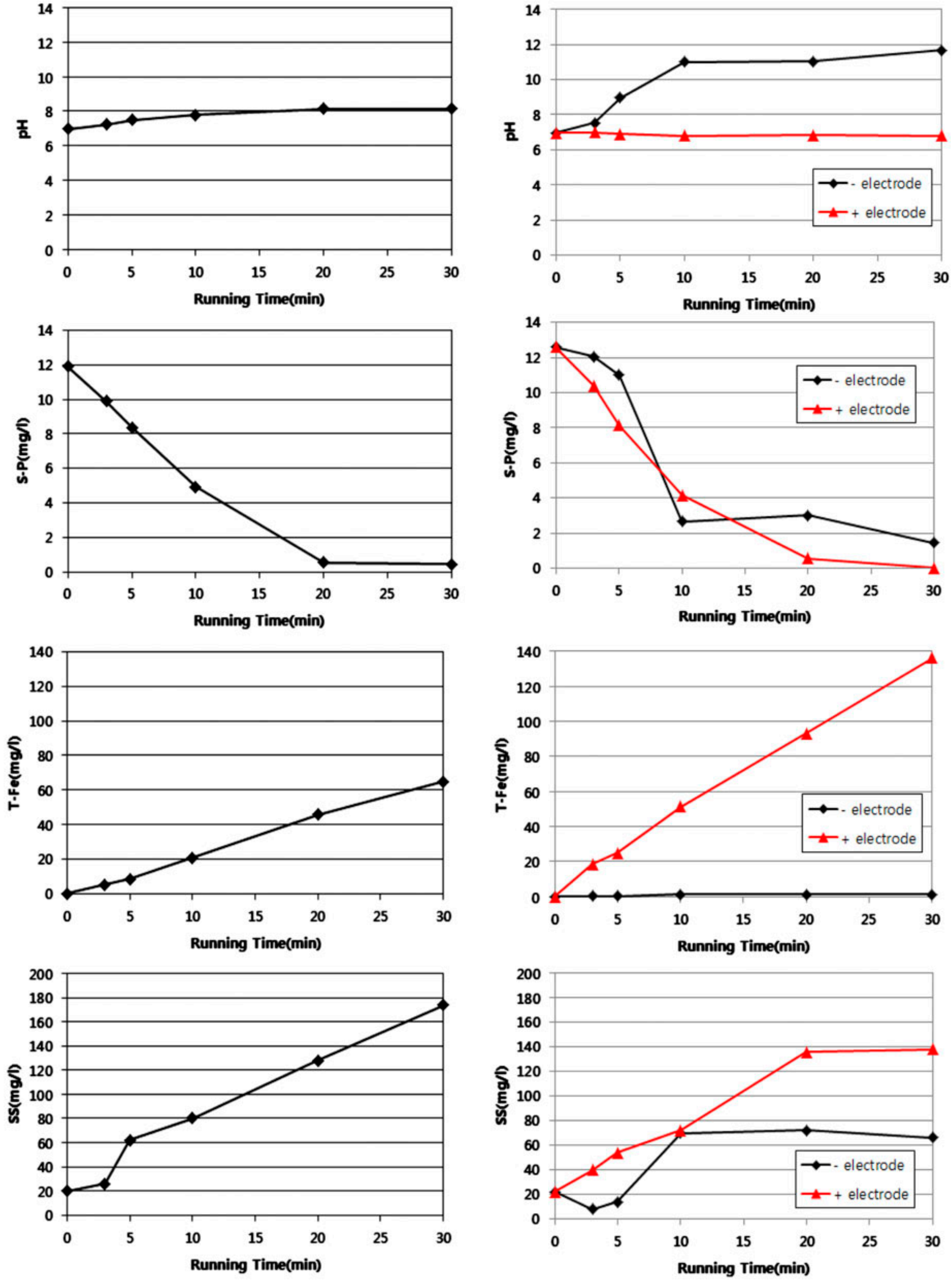

Fig. 2. Comparison of major water quality parameters in batch test: conventional EC (left), partitioned EC (right).

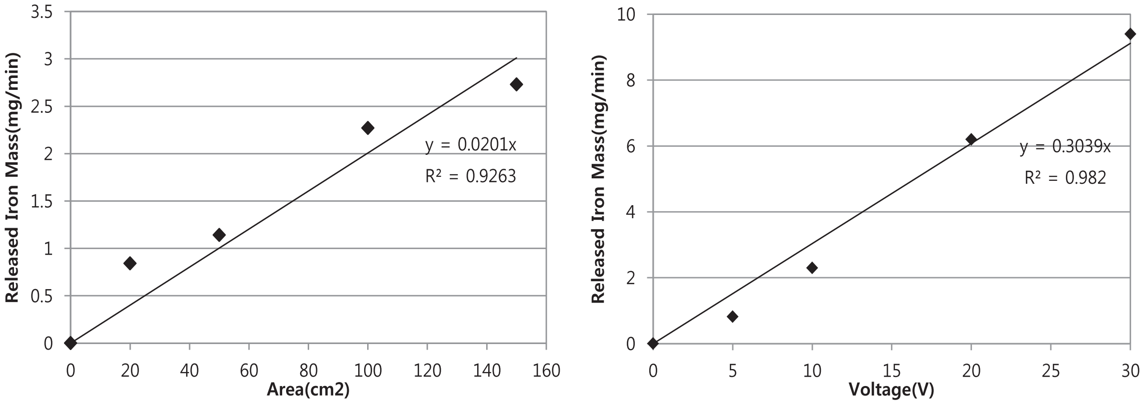

Fig. 3. Iron dissolution rate by increase in electrode area and electric voltage supplied.

Table 1
Summary of conventional EC-treated wastewater quality

	Anode/cathode			
	pH	SS (mg/L)	PO_4^{3-} (mg/L)	Iron (mg/L)
Electrocoagulation tank	7.8	–	4.5	12
Slow-mixing tank	7.8	185	3.8	10
Settling tank	7.8	25	3.5	8

Table 2
Summary of EC-treated wastewater quality with nonwoven partition between electrodes

	Anode/porous partition/cathode							
	pH		SS		PO_4^{3-} (mg/L)		Iron (mg/L)	
	Anode	Cathode	Anode	Cathode	Anode	Cathode	Anode	Cathode
Electrocoagulation tank	7.2 → 6.1	7.2 → 10.8	0	62	7	0.1	8	0.3
Slow-mixing tank	7.3		73		1.2		4.1	
Settling tank	7.3		2		0.4		0.1	

well controlled through the pH and iron dissolution, and provided optimal environment for iron phosphate formation. Low and stable SS, phosphate and iron concentrations in the settling tank proved the success of electrode partitioning.

3.2. Pilot-scale experiment

Table 3 lists the summary of treatment performance of batch-type pilot-scale plant operation for P removal. The pilot-scale experiment was carried out only by the partitioned EC plant operation, to confirm the performance as found by lab-scale test. For the influent of batch test, first and secondary settling tank effluent was used in the Il-san municipal wastewater treatment plant. The batch test was conducted twice and the results were averaged. Monitoring the iron and phosphate concentration over time showed that the plant removed phosphorus in the ranges of 2.3–4.7 PO_4^{3-}-P g/h velocity, at 20 V and 13 A conditions. Regardless of Fe dissolution rate, P removal rate became higher when P loading concentration was high. In the first and secondary settling tank effluent, P concentrations ranged 12–21 and 5–7 mg P/L, respectively. The ratio between Fe dissolved/used for P removal and P removed in mass was found to be in the range around 2–3.

The pilot-scale partitioned EC plant was operated continuously at three different HRTs, and Fig. 4 shows the results of HRT 20 min quality parameters. The

Table 3
Summary of batch-type pilot-scale plant operation result for phosphorus removal

Influent to batch-type EC	Fe dissolution rate (Fe g/h)	P removal rate (PO_4^{3-}-P g/h)	Fe mass required for unit mass P removal (Fe g/PO_4^{3-}-P g)
Effluent from first settling tank	8.8	4.7	≈1.9
Effluent from secondary settling tank	7.2	2.3	≈3.1

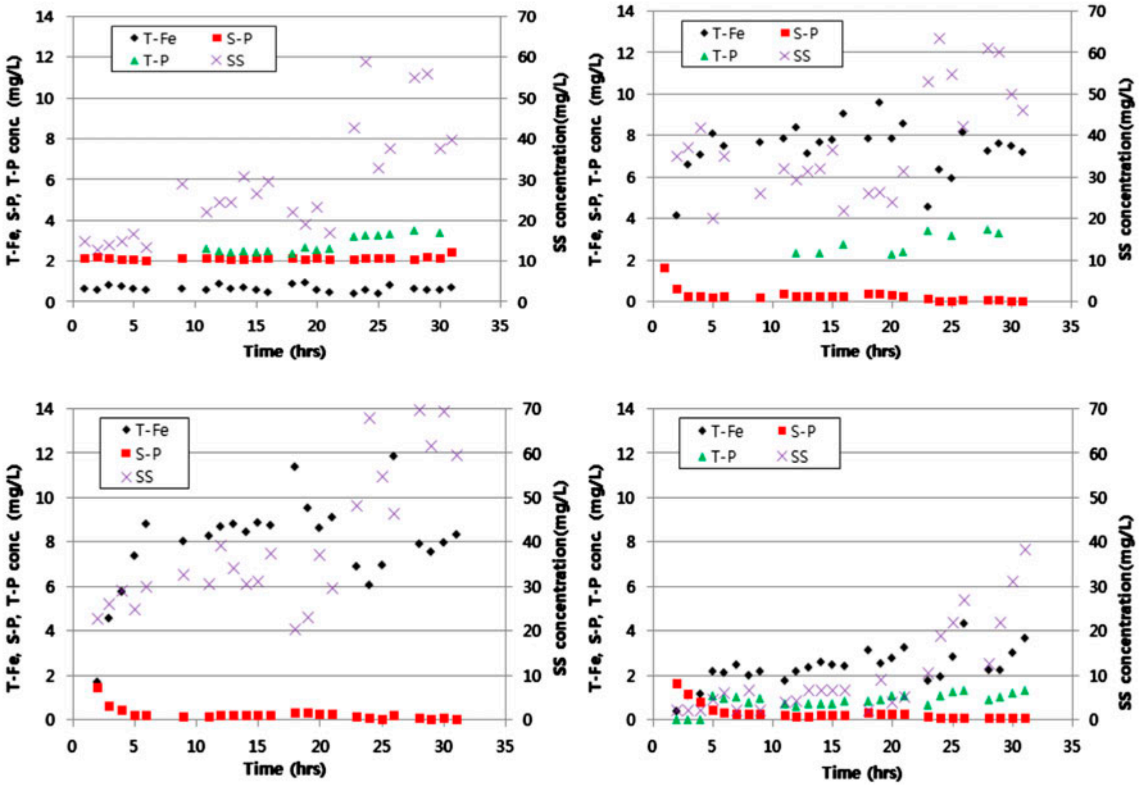

Fig. 4. Pilot-scale partitioned EC system operation result. Samples taken from influent as secondary settling tank effluent, EC tank, after filtration, and slow-mixing tank (from upper left, clockwise).

influent total and soluble P concentrations were around 3 and 2 mg P/L and maintained similar values during the experiment. As the wastewater reaches EC tank, iron concentration rises from 0.5 to around 8 mg Fe/L, and the removal of soluble P was observed. In the slow-mixing tank, fluctuation of Fe and SS concentrations are shown as at this step, the formed metal phosphate aggregates each other and makes larger flocs. The iron drop was little as it was analyzed in total Fe, and also because the influent P concentration was low. Soluble P concentration continued to be low and stable. After filtration, though SS and total Fe remained in some portion, but overall water quality was controlled satisfactory and both total and soluble P concentrations remained the levels less than 0.2 mg P/L.

4. Conclusions

Partitioned electrocoagulation process for phosphorus removal was carried out in laboratory and pilot-scale, each by batch- and continuous-type experiments. In the lab-scale experiment, the production of FeOH was cut down to half when nonwoven partition was placed between electrodes. This is because of the partitioning between anode and cathode successfully blocked the combination of iron with hydroxide ion but used only for iron phosphate formation. The partition between electrodes is proved beneficial in P removal, decrease in sludge production, and also will elongate the life span of the electrode system. At 10 V condition, the velocity of iron dissolution per unit electrode area was 0.02 mg Fe/min cm^2, and the dissolution velocity by electric voltage was 0.3 mg Fe/min V. Hydraulic retention time for stable phosphorus removal was found to be minimum 10 min. The iron mass required for every 1 mg of phosphorus removal was found to be 2–3 mg, and the volumetric loading rate for the treatment of 0.2 mg P/L effluent quality was 0.3 kg PO_4^{3-}-P/m^3 d. In the pilot-scale experiment, when the effluent from the first settling tank was used, phosphorus was treated to 1–2 mg P/L at 20 min of HRT and 10 mg P/L of P loading condition. This also required 10 mg Fe/L of dissolved iron concentration. When it used the secondary settling tank effluent, phosphorus was treated even better with 5 mg Fe/L dissolved iron, to the effluent phosphorus levels less than 0.2 mg P/L at 20 min HRT. At the shorter HRT conditions, the effect of dissolved iron dilution affected phosphorus removal performance, therefore the control of iron concentration to 5 mg Fe/L and higher was necessary. The volumetric P loading rate for the treatment of P to 0.2 mg P/L and less was found to be 0.04 kg PO_4^{3-}-P/m^3 d for the secondary settling tank effluent. Overall, partitioned EC system showed good S-P removal performance over conventional EC, improving the removal efficiencies from 22 to 94%, and the sludge production decreased to 63% compared to conventional EC system.

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