Evaluation of bipolar electrocoagulation applied to biofiltration for phosphorus removal

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Abstract To reduce the residual organic matter and phosphorus contained in secondary effluent, a biofiltration system combined with electrocoagulation using bipolar iron elegctrodes was evaluated as a supplementary treatment to existing small-community sewage treatment. Based on the results of batch tests, bipolar electrocoagulation (BEC) was found to be more effective on phosphorus removal than monopolar electrocoagulation (MEC) but energy consumption was less in monopolar electrocoagulation. Optimum conditions of BEC to treat the secondary effluent were current density 15 A/m^2, electrode spacing 1 cm and pH < 8. The removals of COD_{Cr} and phosphorus by biofiltration system without BEC were 69.1% and 9.6%, respectively. However, biofiltration system combined with BEC showed 76.6–83.7% and 70.7–93.0% removal for COD_{Cr} and phosphorus respectively. Extraordinary increase in phosphorus could be achieved by introducing electrocoagulation to biofiltration, and BEC/biofiltration system was evaluated to be applicable to existing small-community sewage treatment plants as a supplementary process.

Keywords Biofiltration; bipolar electrocoagulation (BEC); phosphorus removal; small-community sewage treatment plant

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Introduction

Application of small-community sewage treatment plants has become a preferred treatment method in many rural areas where it is not possible or economically feasible to connect to a large wastewater treatment system. However nutrient removal such as nitrogen and phosphorus is negligible because most small-community sewage treatment plants were originally designed to remove organic matter and suspended solid (SS). Also, due to the increasingly stringent standard of total nitrogen and phosphorus on wastewater discharge, many existing sewage treatment plants need to be upgraded for removal of nitrogen and phosphorus.

Biological nutrient removal (BNR) processes are common methods for removing nutrients in wastewater. However they have a difficulty in operating, a limit in phosphorus removal and poor removal efficiency of nitrogen in low temperature. Physicochemical processes also have some problems in that the chemicals cost is high, secondary pollution can be caused by the chemicals used and it produces a great quantity of sludge (Mollah *et al.*, 2001).

It has been reported that electrocoagulation (EC) has potential to extensively eliminate the disadvantages of physicochemical and BNR processes. And it has been used to treat potable water (Vik *et al.*, 1984) and a variety of wastewaters (Mameri *et al.*, 2001; Tsai *et al.*, 1997; Kobya *et al.*, 2003). The characteristics of EC process are simple equipment, easy operation, short reaction time and low operating cost (Gurses *et al.*, 2002; Jeong and Lee, 2002). It also produces less amount of sludge than chemical coagulation

(Mollah *et al.*, 2001; Pouet and Grasmick, 1995). It can clearly be an economical and environmental choice for meeting discharge standards and compliance requirements.

In this study, to reduce the residual organic matter and phosphorus contained in secondary effluent, a biofiltration system combined with bipolar electrocogulation using iron electrode was evaluated as a supplementary treatment to be applied to existing small-community sewage treatments.

Experiments

Electrocoagulation

Batch tests were conducted in two stages. In the first stage, monopolar electrocoagulation (MEC) and bipolar electrocoagulation (BEC) were compared running three steps of experiment S1, S2, and S3. Reactors used in this experiment consisted of a 2.5 L acryl ones with four (S1, S2) or six (S3) 30 × 50 × 10 mm iron electrodes as shown in Figure 1. The electrodes were connected to a digital DC power supply (NS-8601, Han Il T&M CO, Korea) which had capacity of 2 A and 20 V. Experimental conditions were electric current 23 mA, initial conductivity 670 μS/cm and initial pH 7. Electrode spacing was 1 cm, 2 cm and 2 cm for S1, S2, and S3 respectively.

In the second stage, optimum operating conditions such as current density, electrode spacing, pH and conductivity were investigated for BEC which showed a higher phosphorus removal efficiency than MEC. The experiments were conducted in a bipolar electrocoagulation unit as shown in Figure 1(b) with six iron electrodes.

The synthetic wastewater was made of potassium dihydrogenphosphate (KH_2PO_4) as 8–10 mg PO_4-P/L. Sodium sulphate (Na_2SO_4) was used to adjust the wastewater conductivity and sulphuric acid (H_2SO_4) or sodium hydroxide (NaOH) was used to adjust pH. Before each test, electrodes were washed with acetone to remove surface grease followed by washing with water, dried and weighed. At the end of test, electrodes were washed thoroughly with water to remove any residues on the electrode surface, dried and re-weighed.

BEC/biofiltration

Continuous pilot test was conducted to evaluate BEC as a combined process to biofiltration based on the results of electrocoagulation experiment. A schematic diagram of BEC/biofiltration system used for the continuous pilot test is shown in Figure 2. The system was composed of a feed water tank, a BEC unit (2.5 L) with a DC power supply (20 V/2A), an inclined plate clarifier (18.3 L) and a biofilter (ϕ 100 mm × 1000 mm). The hydraulic retention times in BEC unit, clarifier and biofilter were 0.3 h, 2.2 h and

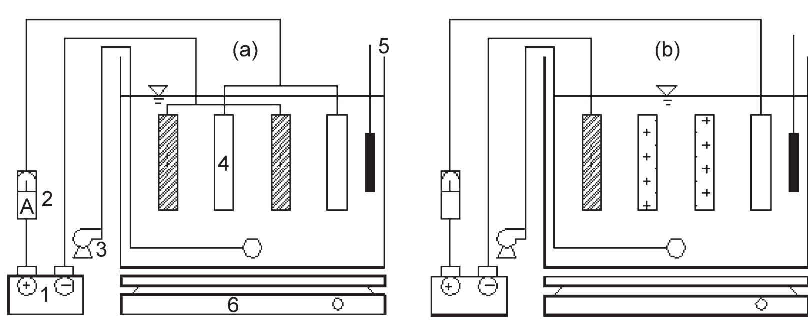

Figure 1 Schematic of electrocoagulation reactors. Monopolar electrocoagulation unit (a) and Bipolar electrocoagulation unit (b). 1: DC power supply, 2: Digital multimeter, 3: Aerator, 4: Electrodes, 5: DO and pH meter, 6: Stirrer

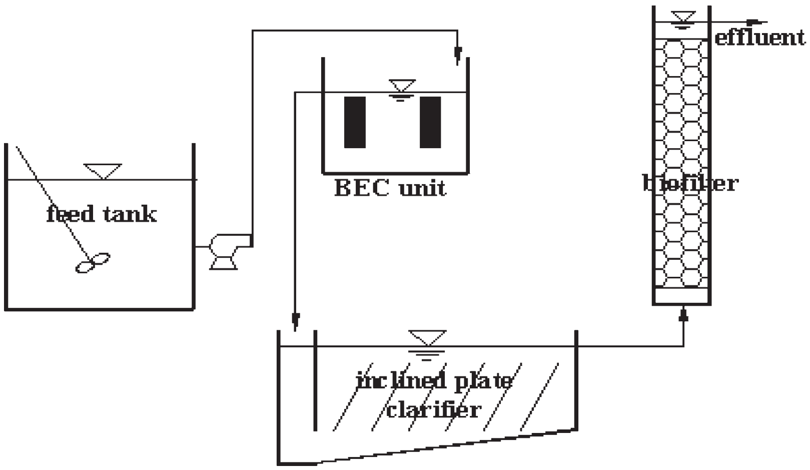

Figure 2 Schematic of BEC/biofiltration system for continuous flow test

0.94 h, respectively. The biofilter was filled with media (2 × 2 × 2 cm) like sponge form made of polyurethane.

Prior to continuous pilot test, biofilter was inoculated with activated sludge obtained from a municipal wastewater treatment plant to attach microorganism on the media. In the first stage, biofilter not applied with BEC was evaluated. And then BEC/biofiltration system was tested at the experimental conditions as shown in Table 1. Synthetic wastewater was made as shown in Table 2. The biofiltration and BEC/biofiltration system were fed at an average daily flow of 200 L/d using synthetic wastewater and operated by upward flow.

Results and discussion

Comparison between monopolar electrocoagulation and bipolar electrocoagulation

Removal efficiencies and energy consumptions of MEC and BEC were compared differing electrode spacings and the number of electrodes. Figures 3 and 4 show the phosphorus removal and energy consumption for MEC and BEC. Because of additionally released iron from sacrificial electrodes at the same current, phosphorus removal was faster in BEC than MEC. However the energy consumption of BEC was more than that of MEC. Change of electrode spacing and the number of electrodes did not affect phosphorus removal in MEC but phosphorus removal increased with increasing the number of electrodes in BEC.

Other parameters could influence the selection of electrode configuration in practice, notably the more mechanically facile connection of multiple electrodes using the bipolar configuration. Also, BEC had advantages of maintenance in regard to installation and replacement of electrodes, and adjustment of electrode spacing. Therefore BEC was

Table 1 Experimental conditions of BEC/biofiltration system

Phase	Electrocoagulation					Biofiltration
	current density (A/m^2)	polarity reversal (hr)	electrode spacing (cm)	conductivity (μS/cm)	pH	
RunI	×	×	×	×	7	o
RunII	10	12	1	650	7	o
RunIII	15	12	1	650	7	o
RunIV	15	24	1	650	7	o
RunV	15	12	1	650	7	o

Table 2 Characteristics of synthetic wastewater

Compound	Dosage (mg/L)	Parameter	Concentration (mg/L)
Starch	10	COD_{Cr}	30
Glucose	9.38	SS	20
NH_4Cl	57.32	NH_3-N	15
KH_2PO_4	13.16	NO_3-N	15
$NaNO_3$	91.07	PO_4-P	3
$CaCl_2$	5		
$MgSO_4$	5		

selected to combine with biofilter considering phosphorus removal efficiency and maintenance advantages.

Bipolar electrocoagulation

Effect of current density. The effect of current density on phosphorus removal, dissolved iron and energy consumption is shown in Figure 5 and Table 3. As shown in Figure 5, the phosphorus level decreased fast with increasing the current density. With increasing the current density, the amount of dissolved iron ions increased as shown in Table 3 and phosphorus removal accordingly increased. In the investigated current density range from 5 to 20 A/m^2, phosphorus was efficiently removed at 15 A/m^2, which

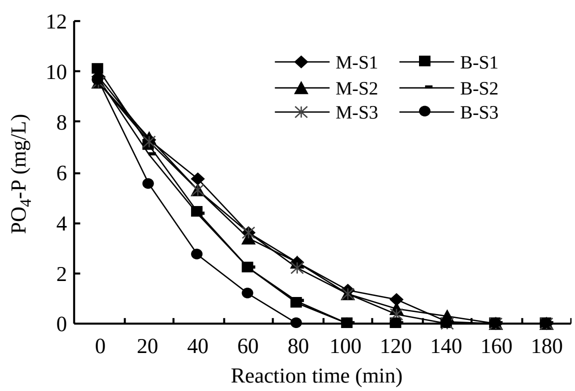

Figure 3 Comparison between phosphorus removals of MEC and BEC. M: monopolar electrocoagulation, B: bipolar electrocoagulation, S1: 1 cm electrode spacing, 4 electrodes, S2: 2 cm electrode spacing, 4 electrodes, S3: 2 cm electrode spacing, 6 electrodes

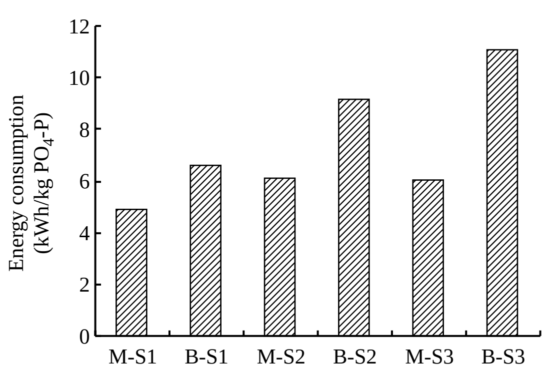

Figure 4 Comparison between energy consumptions of MEC and BEC. (See Figure 3 for abbreviation)

Table 3 Dissolved iron and energy consumption with current density

Current density (A/m^2)	5	10	15	20
Required voltage (V)	3.1	5.3	8.8	12.3
Dissolved iron (mg)	23.7	61.8	113.1	142.8
Energy consumption (kWh/kgPO$_4$-P)	4.71	11.23	14.41	17.89

showed low energy consumption and short reaction time for achieving a residual concentration less than 0.5 mg of PO$_4$-P/L.

The main reactions that occur during the electrocoagulation are as follow.
Reaction at anode,

$$Fe_{(S)} \rightarrow Fe^{2+} + 2e^- \quad (1)$$

Reaction in bulk solution,

$$Fe^{2+} + \frac{1}{4}O_2 + \frac{1}{2}H_2O \rightarrow Fe^{3+} + OH^- \quad (2)$$

$$Fe^{3+}PO_4^{3-} \rightarrow FePO_4 \quad (3)$$

One Fe/P molar ratio will be required if only main reactions occur. However not only FePO$_4$ but also Fe(OH)$_3$ is produced and actual Fe/P molar ratio will be different. Amounts of dissolved iron at each current density were 23.7, 61.8, 113.1 and 142.8 mg, respectively in 180 min. Fe/P molar ratios became 0.55, 1.45, 2.64 and 3.34 at 5 A/m^2, 10 A/m^2, 15 A/m^2 and 20 A/m^2 respectively at the end of experiment. Therefore PO$_4$-P still remained at 5 A/m^2 and 10 A/m^2 where Fe/P molar ratios were less than two at the end of experiment. On the other hand, PO$_4$-P in solution disappeared after lapses of 80 min and 100 min respectively at 15 A/m^2 and 20 A/m^2 where final Fe/P molar ratios were more than two. From the results, required Fe/P molar ratio appeared to be around two to remove PO$_4$-P effectively. However Fe/P ratio should be determined depending on acceptable residual phosphorus level in effluent.

The amount of iron dissolved from feeder anode were similar to theoretical amount but the amount of iron dissolved from sacrificial electrode in the low current density (< 10 A/m^2) was close to zero and increased about 50% at current density of more than 15 A/m^2. Therefore, the current density required for applying sacrificial electrode was more than 15 A/m^2. Also, the higher current density applied, the more energy consumption was required.

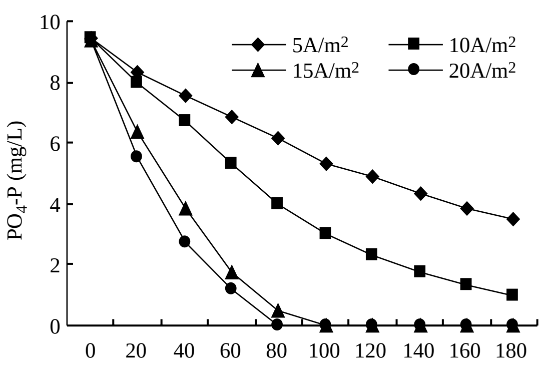

Figure 5 The effect of current density on phosphorus removal

Effect of electrode spacing. In the range of electrode spacing from 0.5 to 3 cm, energy consumption increased due to the increase of required voltage as the spacing increased (Table 4). Narrow electrode spacing of less than 1 cm required low energy consumption but phosphorus removal decreased because dissolved Fe ions could not effectively contact with phosphorus in the solution. However, high removal efficiency was observed at the electrode spacing between 1 and 2 cm (Figure 6). Consequently, 1 cm was selected as optimum electrode spacing considering phosphorus removal efficiency and energy consumption.

Effect of pH. To examine the pH effect on phosphorus removal in electrocoagulation, the wastewater was adjusted to the desired pH for each experiment by using 0.1 N-sodium hydroxide or 0.1 N-sulfuric acid. The effect of initial pH on phosphorus removal was presented in Figure 7. There was not any significant difference in phosphorus removal below pH 7, but phosphorus removal dropped considerably at the pH higher than 8. PO_4^{3-} ions must compete with OH^- ions to be precipitated as $FePO_4$. When the pH is high, there is high concentration of OH^- and PO_4^{3-} could lose the competition, $Fe(OH)_3$ may precipitate instead of $FePO_4$. Therefore, the phosphorus removal is low at the pH higher than 8.

The pH of wastewater changed during the electrocoagulation as shown in Figure 8. Electrocoagulation caused pH increase at the low pH, however, pH dropped at a pH above 9. These results suggested that electrocoagulation presented some pH buffering capacity.

Effect of conductivity. Figure 9 and Table 5 show the effect of wastewater conductivity on the performance of electrocoagulation. Conductivity was adjusted to the desired level by adding sodium sulphate (Na_2SO_4). The effect of wastewater conductivity on the phosphorus removal was negligible in the range from 350 μS/cm to 1000 μS/cm. However, required voltage decreased with increasing wastewater conductivity at the same current. High conductivity was desirable for saving energy consumption because low

Table 4 Dissolved iron and energy consumption with electrode spacing

Spacing (cm)	0.5	1	2	3
Required voltage (V)	2.9	5.5	8.8	12.4
Dissolved iron (mg)	105.9	116.4	113.1	116.1
Energy consumption (kWh/kgPO_4-P)	9.7	10.8	14.4	24.4

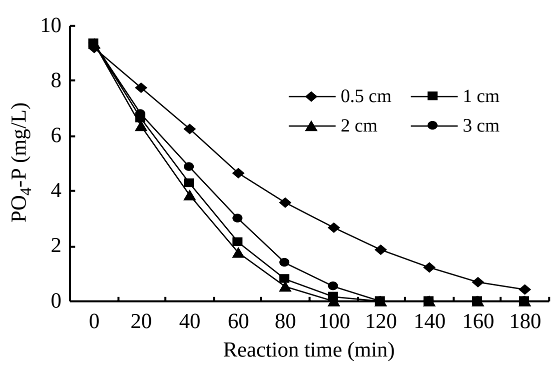

Figure 6 Effect of electrode spacing on phosphorus removal

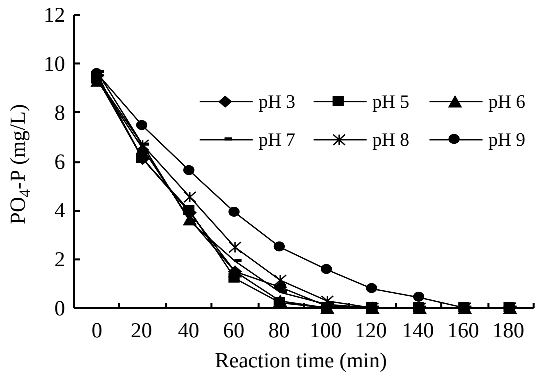

Figure 7 Effect of initial pH on phosphorus removal

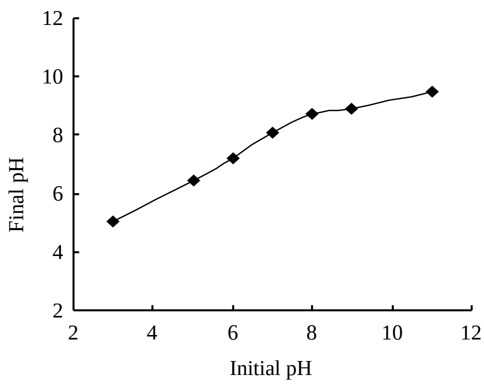

Figure 8 Variation of pH during EC

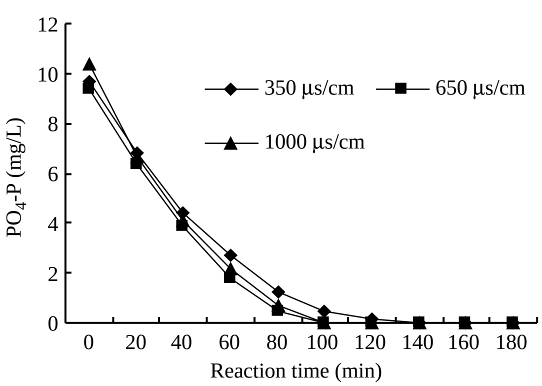

Figure 9 Effect of conductivity on phosphorus removal

Table 5 Dissolved iron and energy consumption with wastewater conductivity

Conductivity (μS/cm)	350	650	1000
Required voltage (V)	15.02	8.8	6.74
Dissolved iron (mg)	112.6	112.7	114.9
Energy consumption (kWh/kgPO_4-P)	33.4	14.41	9.89

conductivity required high voltage, therefore more energy consumption. Energy consumption at the conductivity of 350, 650 and 1000 μS/cm was 33.4, 14.41 and 9.89 kWh/kgPO_4-P, respectively. Accordingly, it may be said that high conductivity is desirable for saving energy consumption

BEC/biofiltration system

COD_{Cr} removal. COD_{Cr} removal efficiency in biofiltration and BEC/biofiltration systems is shown in Figure 10. Removal efficiency of COD_{Cr} by biofiltration system without BEC (RunI) was 69.1%. However, biofiltration system combined with BEC (RunII–RunV) showed 76.6–83.7% removal for COD_{Cr} as shown in Figure 10. The electrocoagulation or electrochemical reactions improved the removal efficiency of COD_{Cr} due to the direct oxidation or indirect oxidation. Direct oxidation occurred on the anodic surface and was due to surface adsorption and decomposition of pollutants. Indirect oxidation occurred in the bulk liquid phase and was caused by the oxidant produced during electrocoagulation. In this test, removal efficiency of COD_{Cr} by electrocoagulation or electrochemical reaction was approximately 13.9% to 27.3%.

When current density increased from 10 A/m^2 (RunII) to 15 A/m^2 (RunIII–RunV), removal efficiency improved by 6.9%. The current density was an important parameter affecting the removal of COD_{Cr}. Polarity reversal period did not affect the removal efficiency of COD_{Cr}.

Phosphorus removal. Removal efficiency of phosphorus by biofiltration system (RunI) without BEC was 9.6%. The effluent of biofiltration system did not satisfy the discharge standard and additional treatment was required. However, biofiltration system combined with BEC showed 70.7–93.0% removal depending on experimental conditions, and phosphorus was effectively removed by electrocoagulation.

It is evident from Figure 11 that high phosphorus removal was observed at the RunIII–RunV operated with current density 15 A/m^2. As current density increased from 10 A/m^2 (RunII) to 15 A/m^2 (RunIII–RunV), removal of phosphorus by BEC increased by 22.3%. More increase in phosphorus removal was caused by precipitation with more dissolved iron ions from sacrificial electrodes. Iron hydroxide and iron complex formed by electrocoagulation were effectively removed in the inclined plate clarifier. Electrode polarity reversal had little effect on the removal of phosphorus. However, as electrodes spacing increased, polarity reversal period lengthened and energy consumption increased with rise of electrolysis voltage.

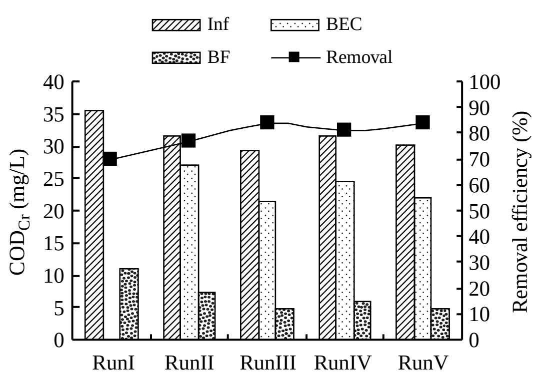

Figure 10 COD_{Cr} removal in biofiltration and BEC/ biofiltration systems. Inf.:influent, BEC:effluent of bipolar electrocoagulation, BF:effluent of biofiltration

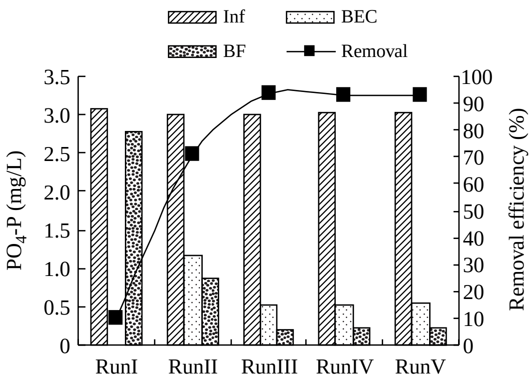

Figure 11 Phosphorus removal in biofiltration and BEC/biofiltration systems (abbreviations as in Figure 10)

Conclusions

This study was intended to examine the feasibility of electrocoagulation as a supplementary treatment to apply to existing small-community sewage treatment. Bipolar electrocoagulation was more effective on the phosphorus removal than monopolar electrocoagulation but required higher energy consumption. The current density was a more important parameter than others in the performance of electrocoagulation. The removals of COD_{Cr} and phosphorus by biofiltration system without BEC were 69.1% and 9.6%, respectively. However, biofiltration system combined with BEC showed 76.6–83.7% and 70.7–93.0% removal for COD_{Cr} and phosphorus respectively. The pollutant in the secondary effluent was effectively reduced by introducing BEC/-biofiltration system. Extraordinary increase in phosphorus removal could be accomplished by combining electrocoagulation with biofilter, and BEC/biofiltration system was evaluated to be applicable to existing small-community sewage treatment plants as a supplementary process.

Acknowledgements

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