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Short Communication

Combination of Electrolysis Technology with Membrane for Wastewater Treatment in Rural Communities

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In this study, the electrolysis technology using copper electrode and ceramic membrane for the microfiltration were consisted to the system and applied for the treatment of domestic wastewater generated from small and rural communities. When the system was operated at the conditions of under voltage of 24V, current ranges of 0.14~0.36A, and TMP of 79.98kPa, the system showed the average removals of organics as chemical oxygen demand, total nitrogen, and total phosphorus as high as 88.5%, 66.7%, and 100%, respectively. Rapid organics removal was observed through electro-oxidation and electro-coagulation during the electrolysis procedure in shorter retention time of 5 minutes, and the residual particulate organics could be also removed through the membrane filtration. Most total Kjeldahl nitrogen was removed by electrolysis, and it could be performed by removal of ammonia which consists of total kjeldahl nitrogen. Phosphorus was also removed successfully through the formation of copper phosphate by electro-coagulation in electrolysis adopting the copper electrode. Increased suspended solids and turbidity by the effect of electro-coagulation in electrolysis were clearly removed during the membrane filtration.

Keywords: Electrolysis, Membrane, Wastewater, Rural communities

1. INTRODUCTION

In general, wastewater refers to liquid wastes containing a mixture of human feces and wastewater from non-industrial human activities such as bathing, washing, and cleaning. Untreated wastewater contaminates severely the groundwater and water bodies [1,2]. Untreated wastewater also poses a major risk to human health since it contains waterborne pathogens that can cause serious human illness. In particular, nutrients such as nitrogen and phosphorus in wastewater can lead to accelerated eutrophication, depleted dissolved oxygen level in water bodies, and unexpected derangement in aquatic system [3,4]. Therefore, the importance of wastewater treatment has been increasingly recognized. However, the wastewater treatment is very difficult, especially in rural communities, for several reasons such as severe inflow fluctuation, lower concentration of pollutants, wide pollution sources, and so on.

The removal of the substances causing eutrophication can be promoted by using of biological nutrient removal processes. Even if the processes are effective and economical, however, its limitations of longer hydraulic retention time and large area requirement (i.e. large volume of the bioreactor) make sometimes the processes less attractive than physic-chemical treatments, which require shorter retention time [5]. The capacity of microorganisms degrading some of the contaminants is limited, moreover, which does not allow the complete removal of these compounds by a biological process [6]. In case of chemical treatment, it has some disadvantages, e.g., problems associated with the handling of chemicals, higher maintenance cost, and the disposal of large amounts of generated by-product [7].

In recent years, thus, the applications of electrochemistry has been proposed to overcome the disadvantages of existing wastewater treatment methods [8]. A number of electrolysis processes such as electro-oxidation and electro-coagulation has been successfully applied for the treatment of many kinds of wastewaters such as tannery [6,9], olive mill [10], agro-industrial [5,11], and domestic [12]. In comparison with conventional treatment methods, the electrolysis has many advantages such as shorter retention time, easy operation and maintenance, no influence by the environment factors (weather, temperature, etc.), and so on. In spite of their many advantages, however, the electrolysis also has a limitation of by-product generation, because the particles and suspended matters in wastewater are removed by electro-coagulation separating coagulated pollutants with eluted ion from electrodes [13].

Membrane separation technology has great popularity over the last 30 years and is becoming a promising technology [14]. It has been widely used for the separation of particulate matters that bigger than pore size in water and wastewater treatment [15-19]. Membrane separation has many advantages such as stable effluent quality and small area requirement [14], and it can guarantee excellent purified effluent by combination with other wastewater processes [20-23]. Even though membrane is already established in commercial application for more than a decade, fouling remains the most crucial problem limiting wider application of membrane filtration [24]. In recent years, therefore, the membrane made of ceramic materials is newly spotlighted to supplement its disadvantages, being presented as the best alternative for existing membrane. We combined the electrolysis technology using copper electrode with ceramic membrane for the microfiltration, in this study, and evaluated the applicability of wastewater generated from small and rural communities.

2. EXPERIMENTAL

As illustrated schematically in Figure 1, combined electrolysis-membrane system used in this study consisted of electrolysis reactor, ceramic membrane reactor, and power supply, respectively. The electrolysis reactor made of plexiglass consists of three trains, and the trains were installed in parallel. Each train was piled as cassettes, of which modules were stacked with floors. Each floor had electrodes array which was composed of a cathode and anodes at both sides of the cathode.

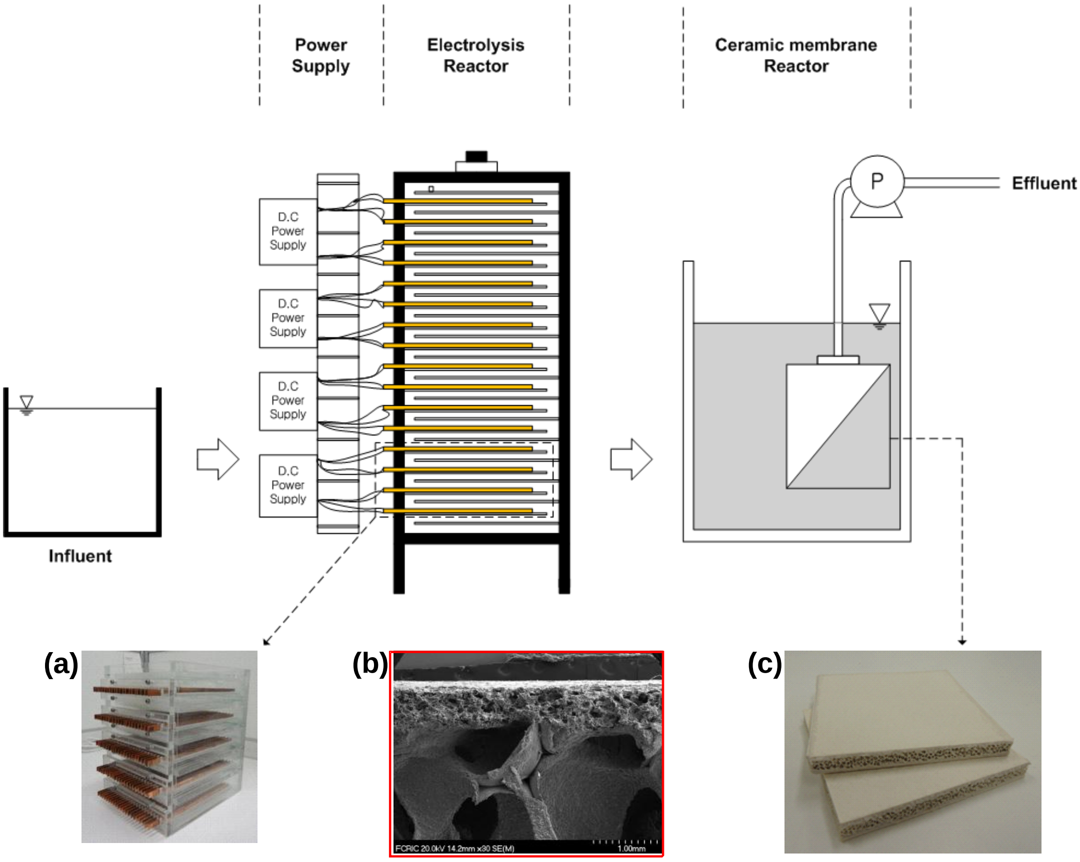

Figure 1. Combined electrolysis-ceramic membrane system (a) Copper electrode cassette (b) Cross-section of ceramic membrane (c) Installed ceramic membrane

For safety and energy saving, the electrolysis reactor was designed to adjust the current to the concentration of pollutants in influent at low bias voltage. The electrolysis reactor was operated under voltage of 24 volts (V) and variable current with an upper limit of 10 amperes (A) by supporting of twelve power supplies. The current value between the anode and cathode was measured simultaneously during the electrolysis procedure. The cleaning of electrode was conducted by scraping with a brush for all cassettes. The specifications of electrolysis reactor was summarized, as presented in Table 1. Influent wastewater was controlled by a submerged fluid pump, and entered first train of the electrolysis reactor. The effluent from first train was introduced to subsequent trains in turn.

Table 1. Specifications of electrolysis reactor

Total volume of train	2L
Number of train in a reactor	3
Size of a train	40mmW×250mmL×1,200mmH
Number of cassette in a train	4
Number of module in a cassette	5
Size of a module	40mmW×250mmL×50mmH
Number of electrode in a module	3
Size of an electrode	2mmW×190mmL×10mmH
Inter-electrode gap	7.5mm
Material of electrode	Copper

Membrane filtration reactor, which is one of major component in the electrolysis-membrane system, was also installed at the end of electrolysis reactor for the separation of all particle matters or by-products in effluent after electrolysis. In this experiment, Plate-type ceramic membrane was applied to membrane filtration. The size of membrane was 50mm long and 5mm wide with a height of 60mm, and the range of pore was 0.1 to 0.2μm. The ceramic membrane was fabricated from Alumina (Al_2O_3) of 99% for the guaranteeing of excellent mechanical strength and endurance on strong acid, alkali, and extremely high temperature. The membrane was operated in a dead-end filtration manner, usually used in a batch-type process, that flow direction was normal to the membrane surface by using of peristaltic pump (Model No. 7553-75, Cole-Parmer Instrument Company), and the flowrate of filtrated liquid was 56mL/min. Flux was calculated via automatic data acquisition equipment which measured filtrated water weight. Pressure gauge was also installed for the measurement of transmembrane pressure (TMP), and the TMP of 79.98kPa was observed in this experiment.

Influent wastewater was collected at the end of screen in wastewater treatment plant located in Yeongwol, Korea. Characteristics of the influent are summarized in Table 2. The influent was typical domestic wastewater generated from small and rural communities in Korea. All analytical determinations were performed as per procedure in the America Public Health Association (APHA) Standard Methods [25].

Table 2. Characteristics of the influent wastewater in rural communities

Parameters		Concentration*	Unit
Turbidity		58.2~65.8 (62.4)	NTU
Suspended Solids		148~154 (150)	mg/L
Chemical Oxygen Demand	Total	150~156 (152)	mg/L
	Soluble	14~20 (16)	mg/L
Total Nitrogen	Total Kjeldahl Nitrogen	18.7~21.4 (19.8)	mg/L
	Nitrate	4.1~5.2 (4.8)	mg/L
Total Phosphorus		2.3~2.5 (2.4)	mg/L
Orthophosphate		1.1~2.3 (1.7)	mg/L

* minimum~maximum (average)

3. RESULTS AND DISCUSSION

The combined electrolysis-ceramic membrane system was devised and evaluated for the treatment of wastewater generated from small and rural communities. When the system was operated at the conditions of under voltage of 24V, current ranges of 0.14~0.36A, and TMP of 79.98kPa, the system showed the average removals of organics as chemical oxygen demand (COD), total nitrogen (TN), and total phosphorus (TP) as high as 88.5%, 66.7%, and 100%, respectively.

As mentioned above, the combined electrolysis-membrane system is comprised of two main part: electrolysis reactor and ceramic membrane reactor. In electrolysis reactor, the elimination of pollutants in wastewater can be performed by electro-oxidation or electro-coagulation. The particulate contaminants like suspended matters or solids can be also removed by separation from wastewater in membrane filtration. The electro-oxidation of organics can occur directly at anodes through the generation of physically adsorbed active oxygen (adsorbed hydroxyl radicals, $•OH^-$) or chemisorbed active oxygen (oxygen in the oxide lattice, MO_{x+1}), and this process is usually called anodic oxidation or direct oxidation [26]. Prominent oxidants such as peroxide, Cl_2, hypochlorite, and ozone can also be produced electrochemically [27], and these oxidants react with the organics, whereas, it is called indirect oxidation. By anodic dissolution followed by hydrolysis, electro-coagulation can produces aluminum or iron hydroxide flocs which destabilize and aggregate the suspended particles or precipitates, and absorb dissolved contaminants [28].

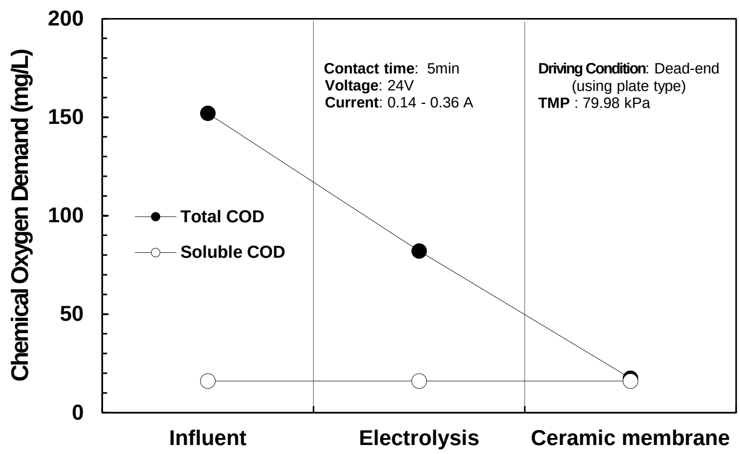

Figure 2. Organics removal characteristics in combined electrolysis-membrane system

The organics removal characteristics of the combined electrolysis-membrane system and each unit processes are as shown in Figure 2. In common, all organic carbon with the exception of certain aromatics like benzene, which are not completely oxidized in the reaction, can be estimated by COD measurement. The COD can be also classified with soluble COD (SCOD) as chemical oxygen demand of soluble compounds and total COD (TCOD) including both particulate or soluble COD, respectively. In this study, we have measured the COD as a degree of the organics concentration in the water. As presented in Figure 2, rapid decline of influent COD concentration was observed during the electrolysis procedure despite of shorter retention time of 5 minutes. It might be explained that the higher COD removal was successfully achieved by both electro-oxidation and electro-coagulation in the electrolysis reactor. In addition, the residual particulate organics could be also removed through the ceramic membrane filtration. Insignificant reduction of SCOD representing soluble organic compounds has been observed, whereas, and it would be considered that the electrolysis reaction time was insufficient and the electric power was too low. The retention time for electrochemical oxidation and electro-coagulation to remove soluble organics in wastewater was just 5 minutes, in this study, and it was very shorter than that of other studies [5, 12, 29, 30]. The electric power used in this study also ranged 3.36 to 8.64 watts, and it was very lower than that of literatures [29, 30] mentioned that the electric power above 50 watts was necessary for improvement of organics removal.

Nitrogen compounds in wastewater is classified into organic nitrogen, ammonia (NH_3) or and ammonium (NH_4^+), nitrite (NO_2^-), and nitrate (NO_3^-). The sum of organic nitrogen, ammonia, and ammonium in wastewater is also classed as Total Kjeldahl Nitrogen or TKN. Biological removal of nitrogen in wastewater is performed via nitrification and denitrification under the conditions of aerobic and anoxic, respectively. However, it can lead to increment of areas in treatment facilities because each reaction needs a spatial separation where its functions are carried out in separated tanks for conditioning of aerobic and anoxic phases. Moreover, the biological nitrogen removal processes using microorganisms can be affected severely by environmental factors such temperature and influent characteristics. In comparison with biological nitrogen removal method, whereas, the electrolysis technology can be an good candidate for the removal of nitrogen in wastewater due to shorter reaction time, easier operation and maintenance, endurance on the environmental effects, and so on.

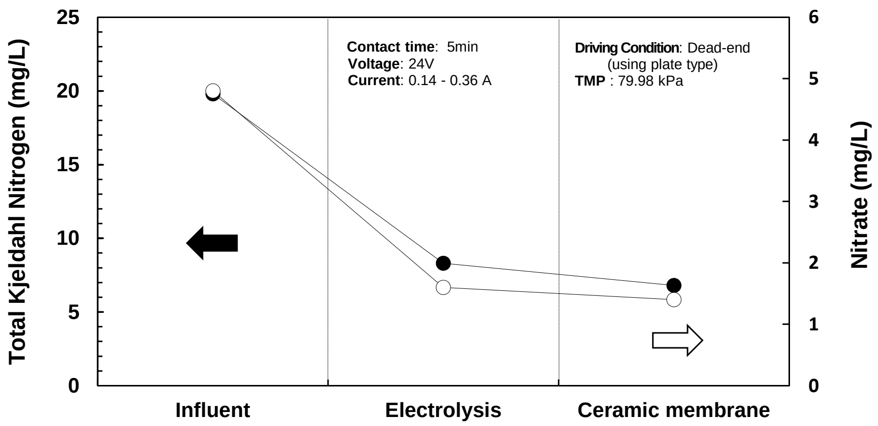

Figure 3. Nitrogen removal characteristics in combined electrolysis-membrane system

Figure 3 presents the profiles of nitrogen compounds in the combined electrolysis-membrane system. In electrolysis, the organic nitrogen can be eliminated by electro-coagulation, and the removals of nitrite and nitrate can be carried out by electrochemical reduction. Ammonia can be also removed by direct and indirect oxidation reactions. The direct oxidation of ammonia takes place at the anode and the indirect oxidation reaction also occur through oxidant such as hypochlorous acid.

The direct and indirect oxidation reactions mechanism of ammonium or ammonium are suggested as follows, respectively;

$$NH_3 + 3OH^- \rightarrow 0.5N_2 + 3H_2O + 3e^- \quad (1)$$
$$2NH_4^+ + 3HOCl^- \rightarrow N_2 + 3H_2O + 5H^+ + 3Cl^- \quad (2)$$

In this experiment, the total kjeldahl nitrogen which was mainly presented as ammonium nitrogen and organic nitrogen with very lower concentration below 1mg/L were measured. Most total kjeldahl nitrogen was removed by electrolysis, and it could be performed by removal of ammonia which consists of total kjeldahl nitrogen. Most organic nitrogen existing a form of solids in wastewater could be eliminated by electro-coagulation, and the remaining organic nitrogen in effluent after electrolysis could be also removed successfully by subsequent membrane filtration.

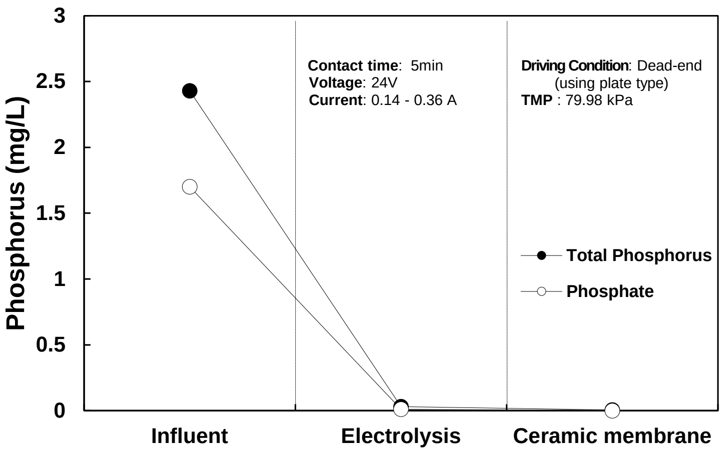

Figure 4. Phosphorus removal characteristics in combined electrolysis-membrane system

In common, eutrophication is well known as the ecosystem response to the addition of artificial or natural substances, such as nitrates and phosphates, through fertilizers or sewage, to an aquatic system. Phosphorus removal can be more effective method for the prevention of eutrophication because its concentration is relatively lower than that of nitrogen in wastewater. A number of biological phosphorus removal technologies such as A/O, A^2/O, and Bardenpho processes have been widely developed and applied to wastewater treatment plants. However, the processes were very difficult to operate, and had many considering factors. Phosphorus removal technology by chemical precipitation also has a severe disadvantage such as large amount of produced by-product. Figure 4 shows the phosphorus removal characteristics in combined electrolysis-ceramic membrane system. In very shorter retention time of 5 minutes, the excellent phosphorus removal performance of the system could be achieved. Most phosphorus was successfully removed through the formation of copper phosphate ($Cu_3(PO_4)_2$) by electro-coagulation in electrolysis adopting the copper electrode: $Cu^+ + 2PO_4^{3-} \rightarrow Cu_3(PO_4)_2 \downarrow$. Thus, the phosphorus removal by electrolysis using the copper electrode would be another good substitute of conventional phosphorus removal processes.

Figure 5 illustrates the turbidity and suspended solids removal characteristics in the combined electrolysis-membrane system. During the electrolysis procedure, the suspended solids concentration increased by the effect of electro-coagulation and the turbidity was also influenced by electrochemical oxidation increasing the dissolved solids concentration. However, both turbidity and suspended solids were perfectly removed during the membrane filtration step.

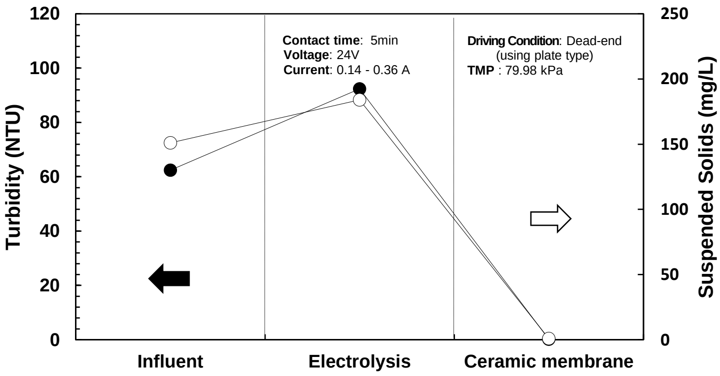

Figure 5. Turbidity and suspended solids removal characteristics in combined electrolysis-membrane system

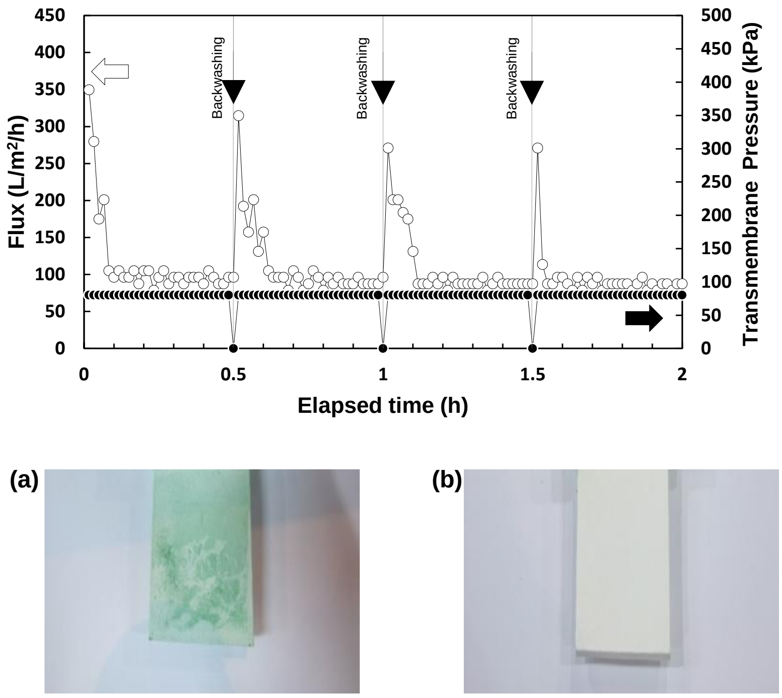

Figure 6. Changes of Flux and TMP (a) Membrane before backwashing (b) Membrane after backwashing

Flux is defined as the flow rate of a property per unit area, in membrane filtration, and transmembrane pressure (TMP) means pressure applied to inside of membrane. Both flux and TMP are expressed by U.S. EPA [31] as follows;

$$J = Q/A \tag{3}$$

where, J = flux ($L/m^2/h$)
Q = permeate flow (L/h)
A = surface area of membrane (m^2)

$$TMP = P_f - P_p \tag{4}$$

where, TMP = transmembrane pressure (kPa)
P_f = feed pressure (kPa)
P_p = permeate pressure (i.e., backpressure) (kPa)

Figure 6 shows the behavior of flux and transmembrane pressure during the procedure of membrane filtration. The flux sharply decreased to about 75 $L/m^2/h$, as the filtration time of electrolyzed effluent progressed, whereas the stable filtration could be maintained successfully. The transmembrane pressure increases during the filtration procedure, in general, the relatively constant pressure was observed due to its unique physical characteristics of ceramic membrane with excellent water permeability. In order to prevent the fouling on the surface of the membrane, the backwashing via air and water was carried out at 30 minutes intervals. As presented in Figure 6 (a) and (b), the backwashing could be successfully performed by simple operation using air and water. Just after the backwashing of membrane, the flux could be recovered as before and stable filtration could be also achieved effectively.

4. CONCLUSION

The electrolysis technology and ceramic membrane were combined and applied for the treatment of domestic wastewater generated from small and rural communities. The electrolysis reactor using copper electrode was operated under the conditions of voltage of 24V and variable currents of 0.14~0.36A, and plate-type ceramic membrane with pore sizes of 0.1 to 0.2μm was applied for the separation of all particle matters or by-products in effluent after electrolysis.

Rapid organics removal was observed through electro-oxidation and electro-coagulation during the electrolysis procedure, despite of shorter retention time of 5 minutes, and the residual particulate organics could be also removed through the membrane filtration. Most total kjeldahl nitrogen was removed by electrolysis, and it could be performed by removal of ammonia which consists of total kjeldahl nitrogen. Most organic nitrogen existing a form of solids in wastewater could be eliminated by electro-coagulation, and the remaining organic nitrogen in effluent after electrolysis could be also removed efficiently by subsequent membrane filtration. Phosphorus was also removed successfully through the formation of copper phosphate by electro-coagulation in electrolysis adopting the copper electrode. The suspended solids concentration and turbidity increased by the effect of electro-coagulation were removed completely during the membrane filtration step.

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