



Influence of electrical field on COD removal and filterability of activated sludge

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ABSTRACT

Three batch reactors were operated in order to investigate the effects of electrocoagulation and electrical field on chemical oxygen demand (COD) degradation rates, efficiencies, and filterability. The first reactor was operated with iron electrode, the second reactor was operated with relatively inert titanium electrode, and the last one was run as control without any electrical field. The electrical field application with iron electrodes increased the COD degradation rates at 0.5 and 2.5 V/cm with COD removals around 90%. However, the removal efficiencies decreased down to 70% at 5 V/cm. Increases in degradation rates were observed with titanium electrodes with time at all the three voltages. Activated sludge from the reactor with iron electrodes showed excellent filtration properties at 2.5 and 5 V/cm experiments. This was attributed to iron precipitates screening membrane foulants and preventing the formation of high resistive cake layer and pore structure rather than floc size increase. Electrocoagulation showed promising results in reducing membrane fouling. A critical voltage gradient should be maintained and the magnitude of iron electrode dissolution should be controlled in the reactor to obtain optimum COD degradation and to enhance filterability. Sludge samples from reactor with titanium electrodes showed poor filterability as voltage gradient increased (at 5 V/cm). This was attributed to the elevated soluble protein content (SMP_p) concentration due to cell disruption.

Keywords: Electrocoagulation; Electrical field; Membrane filtration; Membrane fouling

1. Introduction

Membrane bioreactor applications (either side stream cross-flow or submerged) have been widely applied in both domestic and industrial wastewater treatment due to their advantages over conventional biological processes. They require smaller footprint, eliminate sedimentation tank, and offer various operational benefits. However, the rapid membrane fouling is the main operational concern in membrane

applications, causing higher operational and maintenance costs. Membrane fouling parameters have been extensively investigated in the literature. One group of researchers identified extracellular polymeric substances (EPSs) and soluble microbial products (SMPs) to be the major compounds causing membrane fouling [1,2]. These compounds have large molecular weights and are released by bacteria, containing proteins, polysaccharides, lipopolysaccharides, lipoproteins or complex mixtures of these biopolymers with a variety of functional sites including carboxyl, amino, and

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phosphate groups. EPS is an integral part of the bio-film structure and responsible for bacterial attachment [3,4]; whereas, SMP may potentially cause internal fouling of membranes, decreasing membrane filterability [5]. The other group of researchers identified the colloidal particles as the major foulants in addition to the biopolymers [6,7].

The research on membrane bioreactors (MBRs) has been directed toward controlling compounds (i.e. colloids and microbial products) that cause membrane fouling. The application of antifouling agents such as diatomaceous earth; powdered activated carbon, iron, and aluminum salts; inorganic polymeric substances; and organic polyelectrolytes has shown promising results in reducing membrane fouling [8–13]. The addition of these compounds to activated sludge decreases the concentration of SMP [14] and increases floc size, causing greater cake porosity [15]. In recent years, application of electrocoagulation has been investigated by few researchers to enhance the filtration and to reduce the membrane fouling [16–19]. Electrocoagulation has few advantages over conventional coagulation including no chemical use, easy operation, and no anion (i.e. chloride and sulfate) addition to the water [20]. However, electrical field application may negatively influence the microbial community. This may occur either due to the hydrolysis of water and change of pH [17] or disruption of cell structure on the electrode surface [21]. It has been shown that electrical field with 2 and 4 V/cm application by inert electrodes enhanced the chemical oxygen demand (COD) degradation; whereas, at 6 V/cm, COD concentration increased within an activated sludge reactor [21]. Simultaneous disruption of the cell and the oxidation of cell membranes on the electrode surface were suggested as the potential mechanism in the presence of electrical field.

Better filterability and high COD removals do not necessarily occur simultaneously. There is still need for the fundamental understanding of the effects of electrical field and electrocoagulation on the microbial activity and filtration process. The objective of this study was to investigate the effects of electrical field application with iron electrodes and relatively inert titanium electrodes on filterability and microbial activity of activated sludge process. Enhancement in filterability implies reduction in membrane fouling. Three parallel reactors were operated in batch mode. The first reactor was operated with iron electrode placed inside the reactor to perform electrocoagulation. The second reactor was operated with a titanium electrode which was almost insoluble throughout the experiment, and the last reactor was operated without any electrical field. The effects of electrical field on

COD removal kinetics as well as removal efficiencies were determined. This was performed to investigate the electrical field effect on microbial activity. Then, daily dead-end filtration experiments were conducted by using sludge samples taken from each reactor.

2. Materials and methods

2.1. Experimental set-up

The experiments were conducted by using three 1 L batch reactors. A schematic representation of the experimental setup is presented in Fig. 1. Various electrical fields (0.5, 2.5, and 5 V/cm) were applied in the first two reactors. At each electrical field, the reactors were operated for three days. Iron electrodes were used as anode and cathode material in the first reactor (Reactor I) and titanium electrodes were used in the second reactor (Reactor II). The surface area of the electrodes was 17.6 cm² and the distance between the electrodes was 2 cm. All electrodes were nonperforated. The third reactor was operated as a control without electrical field (Reactor III). The activated sludge used in the reactors was obtained from a pilot plant operated continuously in our laboratory. Six hundred milliliters of synthetic wastewater was added to 400 mL of settled activated sludge daily. At each voltage gradient, fresh activated sludge was used. Hundred milliliters of sludge was wasted from the reactor to adjust the sludge age to 10 days which was the same at the pilot plant where the activated sludge was obtained. The change of COD was observed with time. MLSS, MLVSS, SMP, and particle size of sludge were observed daily. In addition, daily filtration experiments were performed for each sludge sample obtained from the reactors. The filtration experiments were performed using a dead-end filtration cell (Sterlitech) at 1 atmosphere transmembrane pressure. PES membrane with pore size 0.05 μm (Microdyn Nadir) was used in the filtration experiments. The reactor was fed by using synthetic wastewater. The wastewater contained 2,000 mg/L glucose, 154 mg/L peptone, 308 mg/L urea, 154 mg/L KH₂PO₄, 15.4 mg/L K₂HPO₄, 154 mg/L (NH₄)₂SO₄, 54 mg/L MgSO₄·7H₂O, 76 mg/L NaHCO₃, 31 mg/L CaCl₂·2H₂O, 154 mg/L NaCl, 31 mg/L KCl, and small concentrations of other trace elements.

2.2. Analytical methods

Measurements of COD, mixed liquor suspended solids (MLSS), and mixed liquor volatile suspended solids (MLVSS) were performed as defined in standard method [22]. All COD measurements were

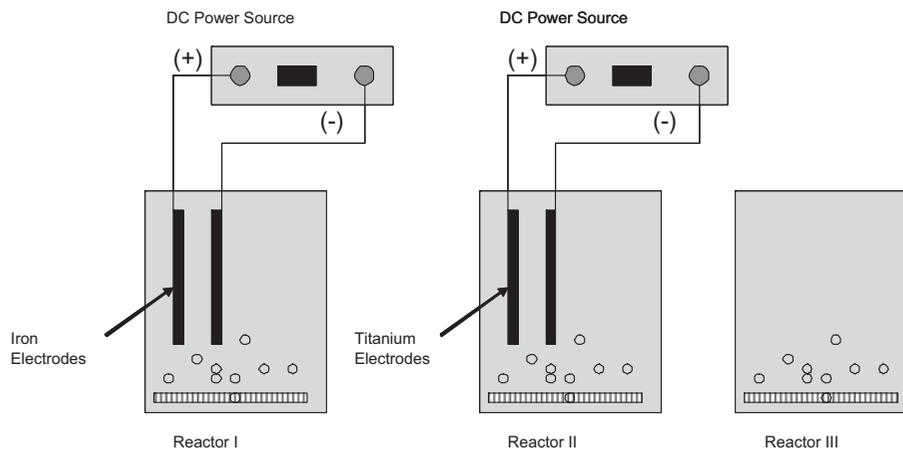


Fig. 1. Schematic representation of experimental setup.

performed after filtering the samples through 0.45 μm membranes. The measurement of protein content was carried out according to Lowry methods [23]. Bovine serum albumin (BSA) was used as a standard and the results expressed in mg equivalent of BSA per liter. The concentrations of all samples were determined by using a UV–Vis spectrophotometer (GBC, Cintra-20) at the wavelength of 660 nm for protein. Carbohydrate contents were determined according to Dubois method [24]. Each sample was analyzed at 490 nm. Simple sugars, oligosaccharides, polysaccharides, and their derivatives give an orange yellow color when treated with phenol and concentrated sulfuric acid. The intensity of the orange color is proportional to the concentration of total carbohydrates present in the suspension (glucose equivalents). The particle size of mixed liquor was measured by a particle size analyzer (Mastersizer 2000).

3. Results and discussion

3.1. Effect of electrical field on COD removal

The change of COD concentration with time is presented in Fig. 2 for 2.5 V/cm experiment. A rapid decline followed by gradual reduction was observed in all experiments. Initial degradation of organics was assumed to follow the first-order kinetics and the rate constants were determined for each experiment by fitting the first 3-h data to the rate equation as follows:

$$\ln C = \ln C_0 - kt \quad (1)$$

where C is the COD concentration at any time (mg/L), C_0 is the initial COD concentration (mg/L), k is the first order rate constant (day^{-1}), and t is the time (day). The fitted rate constants for each experiment

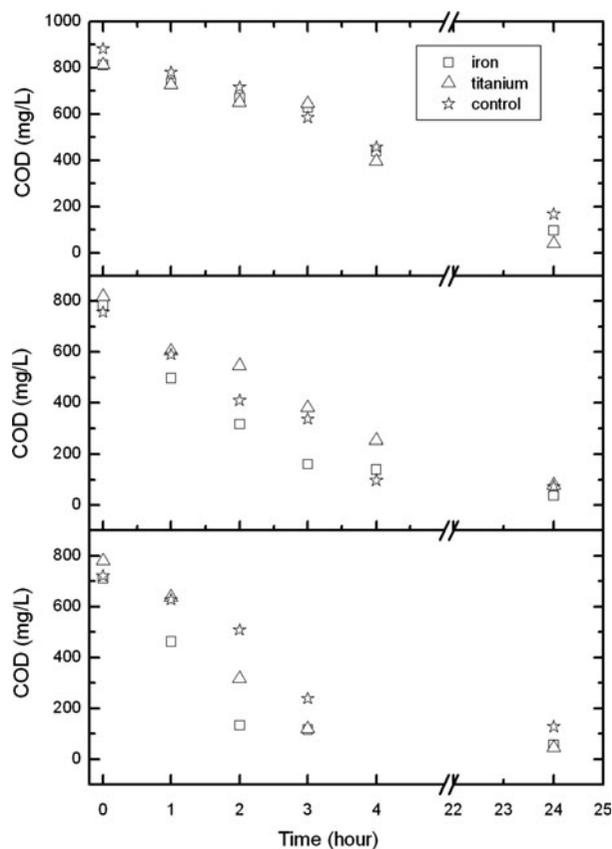


Fig. 2. Change of COD concentration with time at 2.5 V/cm.

are presented in Table 1. The rate constant of Reactor I was relatively smaller than that of Reactor II and Reactor III in the first loading (day 1). In the second and third loading, the rate constant of Reactor I increased substantially from 0.11 to 0.3 day^{-1} and 0.57 day^{-1} , respectively. Relatively smaller but

Table 1
First-order COD removal rate constants

Voltage (V/cm)	Loading (day)	k (day ⁻¹)		
		Iron	Titanium	Control
0.5	1	0.115 ± 0.000	0.194 ± 0.014	0.241 ± 0.022
	2	0.351 ± 0.113	0.247 ± 0.073	0.370 ± 0.084
	3	0.566 ± 0.058	0.397 ± 0.061	0.306 ± 0.020
2.5	1	0.086 ± 0.006	0.081 ± 0.017	0.131 ± 0.016
	2	0.522 ± 0.039	0.240 ± 0.034	0.280 ± 0.022
	3	0.670 ± 0.146	0.632 ± 0.122	0.352 ± 0.102
5	1	0.192 ± 0.006	0.240 ± 0.009	0.249 ± 0.076
	2	0.358 ± 0.036	0.235 ± 0.038	0.302 ± 0.015
	3	0.378 ± 0.028	0.558 ± 0.159	0.504 ± 0.086

significant increase was also observed in Reactor II. At the end of the third day, the rate constant was the highest for the Reactor I and the lowest for the Reactor III. Similar results were also observed at 2.5 V/cm experiment. In the first loading, the rate constants for all reactors were relatively low. In subsequent loadings, significant increases in the rate constants were observed in all three reactors. Much faster degradation occurred in both reactors with iron and titanium electrodes than the reactor without electrical field. Degradation rates also increased from the first loading to the subsequent loadings in the experiments at 5 V/cm. However, the degradation rate in Reactor I was not as fast as the experiments at 0.5 and 2.5 V/cm in the second and third loadings. Results clearly suggested that electrical field application stimulated the degradation kinetics of COD. This occurred at all voltage applications when titanium electrode was used. However, the increase was relatively lower at 5 V/cm

for the reactor with iron electrodes compared to those at 0.5 and 2.5 V/cm. Similar findings were observed also in literature. The rate of COD removal was stimulated for voltage gradient at 0.28 and 0.57 V/cm; however, it became harmful at 1.14 V/cm [25]. Low electrical gradient stimulated microbial activity due to changes in DNA and protein synthesis, cell membrane permeability, and cell growth [26]. A critical current density was also reported in the literature in which microbial activity was stimulated below 25 A/m² for the MLSS concentration of 6,000 mg/L [27].

The initial and final COD concentrations and removal efficiencies are also presented in Table 2. The removal efficiencies were similar varying from 80 to 99% in all reactors at 0.5 and 2.5 V/cm. However, a significant reduction in removal efficiency was observed in Reactor I, decreasing down to 70% in the third loading.

Both kinetics and removal efficiency results showed that 5 V/cm electrical field hindered the process efficiency. Decrease in removal performance in Reactor I was attributed to the dissolution and accumulation of high iron concentration since higher degradation rates and removal efficiencies were observed in reactor with titanium electrode at the same electric field. Two competing mechanisms were reported in the literature for the variations of COD in activated sludge under an electrical field. Disruption of cells releases the EPSs and increases the COD concentration; whereas, at the same time, oxidation at the electrode surface decreases the COD content [21]. The electrochemical oxidation either occurs directly at the electrode surface or occurs due to electrochemically generated oxidants such as hydrogen peroxide and chlorine gas [28]. As the voltage gradient increases, the current also increases resulting in a

Table 2
COD removal efficiencies in all experiments

Voltage (V/cm)	Loading	Iron electrode			Titanium electrode			Control		
		Influent COD (mg/L)	Effluent COD (mg/L)	Removal efficiency (%)	Influent COD (mg/L)	Effluent COD (mg/L)	Removal efficiency (%)	Influent COD (mg/L)	Effluent COD (mg/L)	Removal efficiency (%)
0.5	1	616	64	89.6	722	118	83.7	690	70	89.9
	2	724	144	80.1	666	140	79.0	608	110	81.9
	3	658	8	98.8	628	24	96.2	676	18	97.3
2.5	1	812	96	88.2	814	38	95.3	882	166	81.2
	2	786	38	95.2	818	78	90.5	758	74	90.2
	3	712	56	92.1	780	44	94.4	722	128	82.3
5	1	902	80	91.1	918	100	89.1	1,024	78	92.4
	2	736	206	72.0	954	202	78.8	868	116	86.6
	3	866	264	69.5	874	136	84.4	888	100	88.7

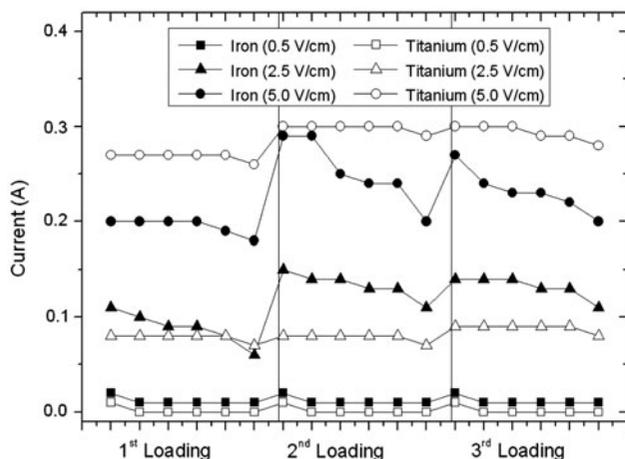


Fig. 3. Change of electrical current with time.

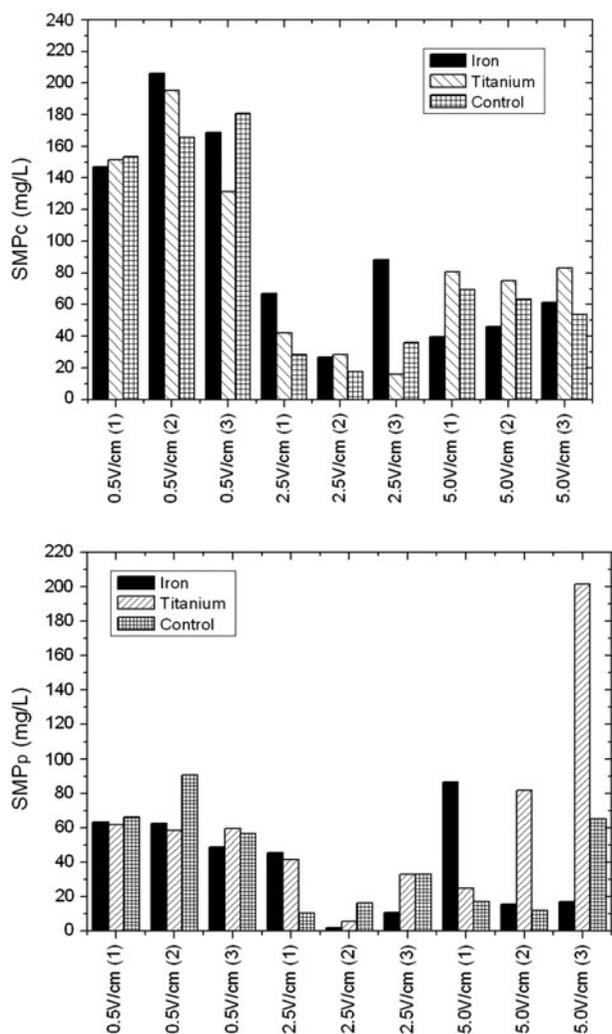


Fig. 4. Variations of SMP_p and SMP_c concentrations.

stronger driving force for the oxidation at the electrode surface (both cell disruption and organic oxidation). The change of current values with time at each loading is presented in Fig. 3. Very low current values were observed at 0.5 V/cm, whereas, substantially greater current values were reached at 5 V/cm. The current reached to 0.3 A for titanium electrode and ranged in between 0.2 and 0.25 A for iron electrodes at 5 V/cm. It was apparent that the increased current likely stimulated electrochemical oxidation at higher current values. This was more apparent for the reactor with titanium electrode since greater SMP_p values were observed in mixed liquor suggesting disruption of cells as discussed below.

Protein and carbohydrate fractions of SMP are presented in Fig. 4. The characteristics of sludge were different at each experiment since fresh sludge was obtained from the pilot plant at the beginning of each experiment; therefore, SMP variations at different voltages cannot be compared. At each electrical field, SMP of Reactors I and II can be compared with those of control (Reactor III) at which no electrical field was applied. At 0.5 V/cm, control and Reactors I and II had similar SMP values. Some variations in SMP fraction compared to the control were observed in experiment with 2.5 V/cm; however, there was no distinct pattern. The most apparent change was that the soluble protein content (SMP_p) in Reactor II which was much greater than Reactor III at 5 V/cm experiment suggested greater cell disruption in titanium electrode surface. Increase in carbohydrate concentration (SMP_c) was also observed at the same electrical field but the magnitude was not as large as that of SMP_p . This can be attributed to subsequent oxidation or degradation of carbohydrates reducing effluent COD concentration and increasing degradation rate. The current values were smaller for iron electrodes at 5 V/cm (Fig. 3). Furthermore, as opposed to titanium electrode, iron electrode itself dissolves substantially, reducing electron transfer to the cells through the electrode surface and formation of the oxidizing compounds. Therefore, cell disruption is not expected as much as in the Reactor II. The rate of electrochemical oxidation decreased substantially compared to the Reactor II. Hindrance in removal efficiency, as well as relatively smaller degradation rates at 5 V/cm, was attributed to excessive dissolved iron content, which was largely in colloidal form.

Enhancement in rate constant at lower electrical field is also a result of iron particle interaction with COD in addition to microbial degradation. Results clearly demonstrated that electrocoagulation enhanced the COD removal rates and efficiencies up to critical dissolved iron content. However, negative effect occurred at elevated iron concentrations. On the other

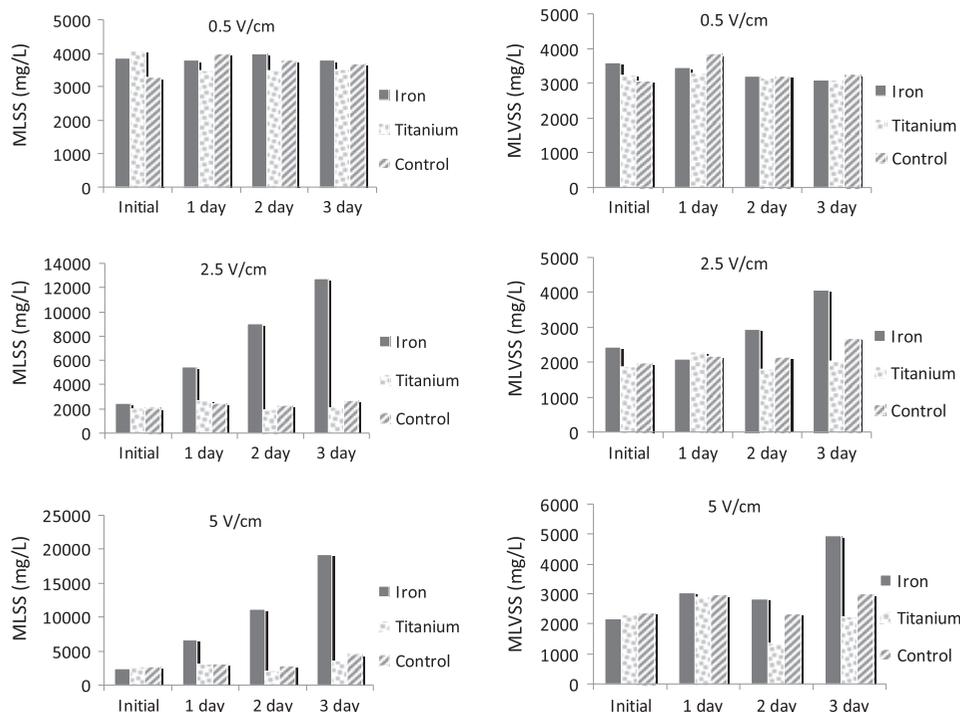


Fig. 5. Variations of MLSS concentrations.

hand, increases in COD removal rates were observed at each electrical field with titanium electrode.

3.2. Effect of electrical field on MLSS and MLVSS

MLSS and MLVSS data are presented for all experiments in Fig. 5. Both MLSS and MLVSS did not change significantly in the experiments conducted at 0.5 V/cm. Little iron was dissolved since the current was relatively low (Fig. 3). MLSS/MLVSS ratio was changing from 0.8 to 0.94, showing high micro-organism content of the suspended material. A significant increase was observed in MLSS due to the dissolution of iron electrode at 2.5 V/cm experiment. The increase was almost linear. MLSS content of Reactor with titanium did not change significantly. Similar observations were also made for the reactor without electrical field. While MLVSS/MLSS ratio was around 0.9 for all three reactors, it decreased down to 0.32 for reactor with iron electrode. Change in MLVSS was not significant. Slight increases were observed in Reactor I. MLSS/MLVSS ratio did not change significantly in reactor with titanium electrode and in control.

3.3. Effect of electrical field on filterability of activated sludge

Cumulative filtration volume change with time for 2.5 V/cm experiment is presented in Fig. 6. At the end

of each loading, 25 mL of sludge samples was taken from each reactor and filtered in a dead-end filtration module. The same experiments were also conducted by using the sludge samples obtained from 0.5 and 5 V/cm experiments at the end of each loading. In Table 3, time required to obtain 20 mL of filtrate in all experiments is presented. The filterability of the sludge obtained from the Reactor I was much better than those of Reactor II and Reactor III at 2.5 V/cm and 5 V/cm electrical fields at each loading. Twenty milliliters of filtrate was obtained in 3–5 min from the sludge of Reactor I; whereas, it took about 11–15 min for sludge from Reactor III and 12–19 min for sludge

Table 3
Time required to filter 20 mL of activated sludge samples

Voltage (V/cm)	Loading (day)	Filtration time (minutes)		
		Iron	Titanium	Control
0.5	1	5.9	4.2	4.2
	2	7.1	4.4	4.5
	3	4.8	3.4	4.5
2.5	1	5.8	14.1	11
	2	3.4	19	15
	3	3	11.8	11
5.0	1	4.8	11	8.5
	2	3.2	17.5	8.5
	3	4.8	43.5	11.1

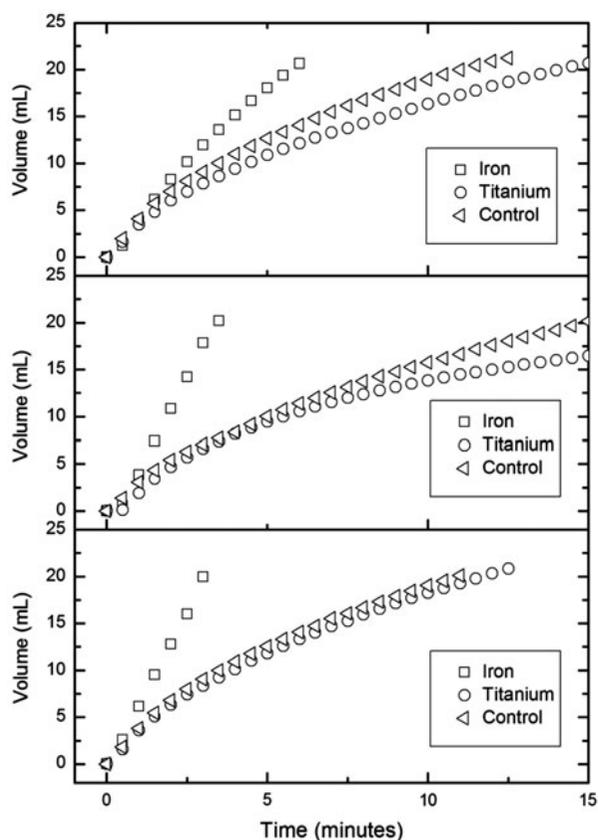


Fig. 6. Filtration performance of sludge samples taken from three reactors at 2.5 V/cm.

from Reactor II at 2.5 V. Much faster filtration occurred in Reactor I despite much higher MLSS concentration. Similar filtration profiles were also obtained at 5 V/cm for Reactors I and III; however, the filterability of Reactor II substantially got worse at each loading, reaching filtration time of 20 mL of permeate up to 44 min. This can be attributed to the elevated SMP_p concentrations (Fig. 3) that were likely a result of cell disruption on the electrode surface. These results suggested that iron electrocoagulation substantially increased the filterability at 2.5 V/cm and 5 V/cm; however, the electrical field application without coagulation had considerable negative effect on the filtration of activated sludge in the reactor with titanium electrode. The negative effect was much more noticeable as the electrical field strength increased. As the electrical field strength increased, the current density also increased and likely caused higher cell disruptions on the electrode surface releasing elevated SMP_p concentrations and resulting poor filterability.

In the literature, enhanced filterability (or reduced fouling) by coagulation and flocculation was attributed to either the increase in average floc size

Table 4

Average particle size of flocks in various

Voltage (V/cm)	Average particle size (μm) (loadings 1, 2 and 3)		
	Iron	Titanium	Control
0.5	58.6	68.6	66.5
2.5	36.4	40.5	38.6
5.0	38.3	37.4	40.8

[16,29,30] or reduction in the concentrations of microbial polymeric substances [9,30]. The average floc size variations in each reactor are presented in Table 4. The values represent the average of all three loadings. There were no significant variations ($<2\mu\text{m}$) at each loading. Although, some small particles (less than $1\mu\text{m}$) and some larger particles (greater than $100\mu\text{m}$) were observed in reactor with coagulation (Reactor I), the average floc size and the distribution in all three reactors were similar. Therefore, the enhanced filterability of Reactor I cannot be solely attributed to the particle size increase. Furthermore, change in SMP_p and SMP_c did not show decreasing tendencies (Fig. 4). Inorganic iron particles (i.e. iron hydroxide) are likely strained small colloids or soluble fractions reaching to membrane surface. Similar conclusions were also reported by other researchers. It was reported that the formation of aluminum hydroxide or iron hydroxide particles reduced fouling by preventing fouling-causing compounds to reach the membrane pores [18,19].

Somewhat different results were obtained at the experiments at 0.5 V/cm. For these experiments, filtration of all sludge samples occurred relatively rapidly. Reactor I yielded slightly slower filtration than Reactors II and III. The dissolution of iron electrode in 0.5 V/cm was relatively insignificant since there was no significant MLSS increase. Enhanced filterability in all reactors was attributed to the characteristics of the initial activated sludge seed obtained from the pilot plant. Significant variations occurred in the sludge characteristics with time in pilot plant in the laboratory. Then, 0.5 V/cm experiments were conducted several months later than the other two voltage gradient experiments. The particle size distribution showed distinct differences among activated sludge samples used as seed in 0.5 V/cm and the others. The average particle size of sludge samples was 67, 39, and $41\mu\text{m}$ for 0.5, 2.5, and 5 V/cm, respectively. It was apparent that the particle size had significant influence on filterability. The effect of dissolved iron at 0.5 V/cm was not apparent as was for the other voltage gradients. This is due to the fact that the filterability of the

sludge was already good and the dissolved iron content was much lower than the other voltage gradients.

4. Conclusion

The electrical field application with iron electrodes increased the COD degradation rates at 0.5 and 2.5 V/cm with COD removals around 90%. However, the removal efficiencies decreased down to 70% at 5 V/cm. Increase in degradation rates was observed with titanium electrodes with time at all three voltages. The current increased at higher voltages that caused both cell disruption as well as electro-oxidation simultaneously. Sludge obtained from the reactor with iron electrodes showed excellent filterability at 2.5 and 5 V/cm. This was attributed to the iron precipitates straining membrane foulants and preventing the formation of high resistive cake layer and pore structure. Electrocoagulation showed promising results in reducing membrane fouling. Poor filterability was observed as voltage gradient increased in reactor with titanium electrodes due to elevated SMP_p concentration.

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