ELECTROCOAGULATION: IS IT COST-EFFECTIVE FOR WASTEWATER TREATMENT

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ABSTRACT

Electrocoagulation (EC) is a wastewater treatment technology capable of removing suspended solids, dissolved organic matter and nutrients, faecal indicator bacteria as well as heavy metals, oils and other organic contaminants. EC has been most widely used for the treatment of industrial wastewater, including textile, oil, paper, and dye wastewaters.

EC generates coagulants in situ by electrolytic oxidation of metal anodes. Iron or aluminium plates are commonly used for the anodes, releasing iron (Fe^{2+}) and aluminium (Al^{3+}) ions into the wastewater which hydrolyse to polymeric hydroxides. Polymeric hydroxides are excellent coagulants for the removal of various wastewater pollutants. Coagulation involves charge neutralization of negatively charged contaminants followed by the formation of flocs that either settle or float. Therefore, a subsequent solids removal stage (e.g., clarifier or Dissolved Air Flotation) is required.

EC efficiency can be improved by optimizing operational parameters including: electrode spacing, electrode orientation, periodic electrode polarity reversal, current density (A/m^2) and contact time. Particularly, the removal efficiency of TSS and particulate BOD (including algae which are negatively charged) by EC is mainly dependent on the amount of Fe^{2+} or Al^{3+} ions generated from the anode. Therefore, greater removal can normally be achieved at higher current density. Phosphate ions (PO_4^{3-}) are neutralized by the polymeric metal hydroxides which also directly bind to suspended P contaminants. These then aggregate and settle with the flocculated solids. All nitrogen compounds can be removed to some degree by EC. For example, organic nitrogen is removed with the flocs of TSS. EC can promote inactivation of microorganisms including faecal coliforms and viruses by rupturing their membranes and then coagulating them into settleable flocs. Furthermore, EC can remove heavy metals as metal hydroxides and other organic compounds including pesticides and halogenated hydrocarbons.

Many laboratory-scale EC trials have been conducted to determine optimum design and operation parameters for efficient wastewater treatment. However, currently there is no information on full-scale application of EC technology available in peer-reviewed scientific literature. This study tested a laboratory-scale EC unit for the treatment of wastewater pond effluent. Pond water samples (~20 L) were collected from an oxidation pond on three occasions and each sample was used on the same day for the laboratory experiments. The effect of different EC currents (between 0.4A and 3A) on the water quality of the wastewater pond effluent was investigated in terms of the removal of organic matter (TSS and BOD_5), nutrients (nitrogen and phosphorus), and faecal coliforms. Physico-chemical parameters including temperature, pH, dissolved oxygen (D.O.), conductivity, turbidity and %UV transmittance (UVT) were also measured before and after the EC treatment.

This study showed that the laboratory-scale EC unit typically achieved >90% removal of TSS, BOD_5 and TP, >95% removal of DRP, 50-80% removal of TKN, and 2-3 log removal

of *E. coli* at a EC current of 0.8-1.6A. Full-scale EC unit power consumption would be ~0.4 kWh/m^3 wastewater which would cost NZ$0.12/m^3 wastewater (based on the current average power cost of NZ$0.30/kWh). This research indicates that EC is an efficient and potentially cost-effective option for treating wastewater pond effluent since the EC can provide a combined removal of organic matter, phosphorus and disinfection (replacing chemical flocculation/coagulation and UV treatment) and produce a readily dewaterable sludge.

KEYWORDS

Solids removal, phosphorus removal, disinfection, sludge dewatering, sludge management

PRESENTER PROFILE

Rupert is a Principal Scientist and Manager of the Aquatic Pollution Group at NIWA in Hamilton. He has undertaken research and consultancy for >20 years on pond and other ecotechnology treatment of domestic, agricultural and industrial wastewaters. His research specifically focusses on resource (energy, nutrient and water) recovery from wastewater.

1 INTRODUCTION

Electrocoagulation (also referred to as Electro-flocculation) has been widely applied for the removal of various contaminants from domestic and industrial wastewaters including: solids (TSS/turbidity), organic matter (BOD/COD/TOC/DOC), nutrients (nitrogen and phosphorus), faecal coliforms, heavy metals (Cd, Ni, Cu, Cr, Zn, As, Sb, etc.,), oils and other organic contaminants (Emamjomeh and Sivakumar 2009; Mook et al. 2012; Kuokkanen et al. 2013; Mook et al. 2014; An et al. 2017). EC generates coagulants in situ by using electrolysis (usually driven by a Direct Current (DC) voltage) to release ions from metal anode (positively charged) electrodes (Figure 1). Both aluminium (Al) and iron (Fe) plates are commonly used as the anode for EC.

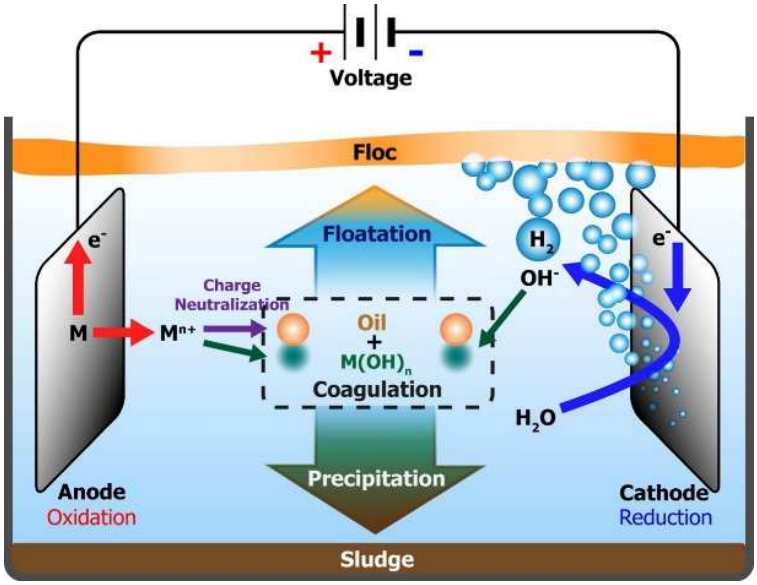

Figure 1: Schematic diagram of the electrocoagulation (EC) process (An et al. 2017).

The anions can remove suspended, emulsified, and dissolved contaminants from the wastewater (Chen 2004; Mollah et al. 2004; Emamjomeh and Sivakumar 2009; Butler et al. 2011). The metal ions are released from the anode electrode as electrons pass into the wastewater, while water is hydrolyzed to hydroxyl ions and hydrogen (H_2) gas bubbles at the cathode (negatively charged) electrode where electrons are taken up from the wastewater to complete the electrical circuit (Figure 1).

EC is very effective in removing many contaminants from wastewaters and is characterized by ease of operation and reduced sludge production compared with chemical coagulation (Mollah et al. 2001; Chen 2004; Sahu et al. 2014; Korving et al. 2019).

EC involves many chemical and physical reactions that are involved in three successive stages of contaminant removal (Figure 2) (Mollah et al. 2004; Emamjomeh and Sivakumar 2009; Elazzouzi et al. 2017):

(i) Dissolution: generation of coagulants by electrolytic oxidation of the metal 'sacrificial anode' (e.g., Al or Fe plates)

(ii) Coagulation: charge neutralization of negatively charged contaminants present in the wastewater by the positively charged metal cations, and

(iii) Flocculation: aggregation of the destabilized phases to form flocs that can either settle or float.

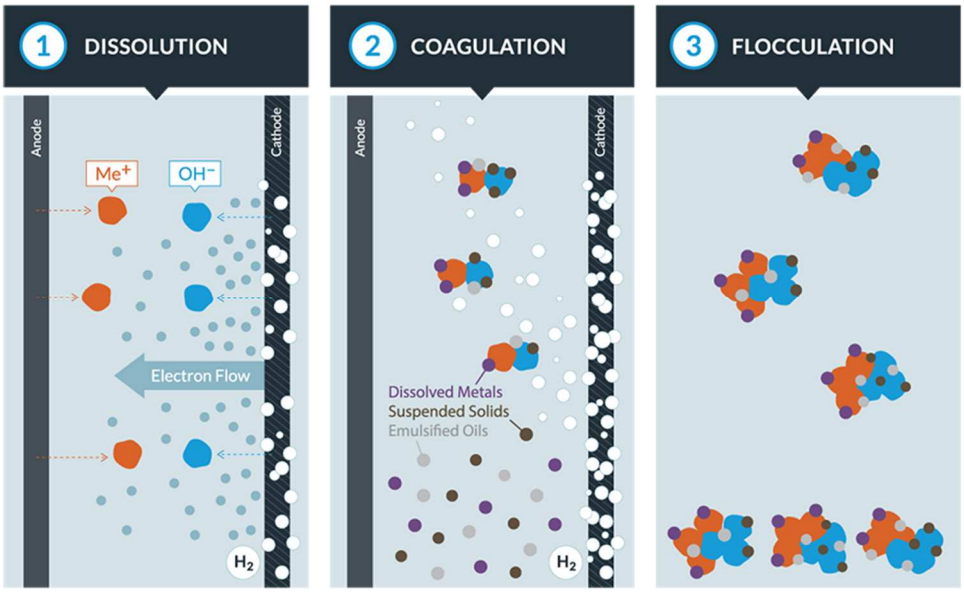

Figure 2: Three stages of electrocoagulation (EC) (http://www.watertectonics.com/electrocoagulation/)

More specifically, metal anodes (Fe or Al) cause a series of physiochemical reactions below (Shen et al. 2003; Mollah et al. 2004; Chen 2004; Kuokkanen et al. 2015; Franco et al. 2017):

- Once formed at the anode, iron (Fe^{2+}) and aluminium (Al^{3+}) ions almost immediately hydrolyse to polymeric hydroxides which are excellent coagulants. For example, hydrolysed aluminium ions form large polymers of Al–O–Al–OH.
- Coagulation occurs when these metal cations combine with the negatively charged contaminants (e.g., algal cells) that are attracted to the anode by electrophoretic motion. The contaminants are removed from solution either by chemical reactions and precipitation or by physical and chemical attachment to colloidal materials.

Adsorption of mineral cations to 'in situ' generated hydroxides is 100 times greater than to added metal hydroxide chemical coagulants (Mollah et al. 2004).

- Water molecules are also electrolyzed in a parallel reaction, producing small bubbles of oxygen and positively charged hydrogen ions (protons) at the anode ($2H_2O \rightarrow O_2 + 4H^+ + 4e^-$). These hydrogen ions are attracted to the cathode where they combine with electrons from the electrical circuit to form additional hydrogen gas ($4H^+ + 4e^- \rightarrow 2H_2$).
- Gas bubbles attach to the flocculated particles and pollutants floating them to the surface (Chen 2004; Mollah et al. 2004; Emamjomeh and Sivakumar 2009; Butler et al. 2011; An et al. 2017).

Many operation parameters including electrode materials, spacing, plate orientation, and water pH, conductivity and temperature affect the performance of the EC treatment (Chen 2004; Mollah et al. 2004; Katal and Pahlavanzadeh 2011; Attour et al. 2014; An et al. 2017). In particular, the amount of metal ions (i.e., coagulant) released into the wastewater from the anode is proportional to the 'current density' (the total amount of electric current flowing through the anode surface area that is in contact with the wastewater, A/m^2) and contact time (Chen 2004; Mollah et al. 2004). Therefore, greater organic matter removal (TSS and BOD) can normally be achieved at higher current density (Ebeling and Ogden 2004; Bukhari 2008; Makwana and Ahammed 2016; Elazzouzi et al. 2017).

Phosphate ions (PO_4^{3-}) are neutralized by the polymeric metal hydroxides which also directly bind to suspended P contaminants (Ebeling and Ogden 2004; Kuokkanen et al. 2015; Franco et al. 2017). These then aggregate and settle with the flocculated solids. EC can promote inactivation of microorganisms including faecal coliforms and viruses by rupturing their membranes and then coagulating them into settleable flocs (Diao et al. 2004; Elazzouzi et al. 2017). Furthermore, Emamjomeh and Sivakumar (2009) reported that EC can remove heavy metals as metal hydroxides and other organic compounds including pesticides and halogenated hydrocarbons.

All nitrogen compounds can be removed to some degree by EC (Koparal and Öğütveren 2002; Mook et al. 2012). Nitrate can be reduced to nitrogen gas (N_2) through sequential electrochemical reduction ($2NO_3^- + 5H_2 + 2H^+ \rightarrow N_2 + 6H_2O$). Nitrate may also dissociate to N^{3-} ions that react with iron (II) and iron (III) cations to form iron nitride precipitates ($Fe^{3+} + N^{3-} \rightarrow FeN$ or $3Fe^{2+} + 2N^{3-} \rightarrow Fe_3N_2$), which settle with the flocculated solids. Nitrate/nitrite removal increases with higher current density or longer contact time. Ammonium ions (NH_4^+) may react with anions in hard water (containing many cations) to form insoluble precipitates. However, this is less likely in NZ, as freshwater is relatively soft.

Many researchers (Chen 2004; Mollah et al. 2004; Mook et al. 2014) pointed out that there is a critical current density and treatment time above which there is little improvement in EC performance, and efficiency is reduced as electrical power is wasted by heating up the wastewater. Moreover, dissolved metal ions may be reduced at the cathode resulting in fouling, which must be removed to maintain treatment performance. Therefore, selection of optimal current density for a particular wastewater should be based on the wastewater characteristics (e.g., conductivity, pH, and temperature) and operation parameters such as wastewater flowrate and the type of pollutants to be removed.

This study investigated the treatment of wastewater pond effluent using a laboratory-scale electrocoagulation unit using Fe-Fe electrodes in terms of TSS (mainly algal solids), organic matter (BOD_5), nutrients (N and P) and *E. coli* removal.

2 MATERIAL AND METHODS

2.1 LABORATORY SCALE ELECTROCOAGULATION UNIT

A schematic of the laboratory-scale EC unit is illustrated in Figure 3. This EC unit was provided by Powell Water Systems (PWS) in New Zealand and has been used extensively in the US to provide design, size and contact time information for installation of commercial EC units.

The EC unit was tested for the treatment of wastewater pond effluent. Pond water samples (~20 L) were collected from a 1 ha wastewater treatment HRAP at Cambridge wastewater treatment plant in New Zealand and used on the same day for the laboratory experiments. Three experiments were conducted to investigate the effect of different EC currents (between 0.4A and 3A), which determine the amount of metal ion coagulant released into the wastewater from the anode, on the water quality of the pond effluent.

EC outflow samples were collected, gently mixed to degas the floccs and allowed to settle for 1 hour. The supernatant was then analysed for Total Suspended Solids (TSS), nitrogen (TKN, NH_4-N, and NOx-N (NO_2-N+NO_3-N)), phosphorus (TP and DRP), and *E. coli* according to Standard Methods (APHA 2017). The results were compared to the initial TSS, nitrogen, phosphorus and *E. coli* concentrations to determine the removal efficiency. Physico-chemical parameters including temperature, pH, dissolved oxygen (D.O.), conductivity, turbidity and %UV transmittance (UVT) were also measured before and after the EC treatment.

The EC operation conditions of the thee laboratory experiments including current density, voltage, interelectrode voltage, total power, power use, and flow rate are summarized in Table 1.

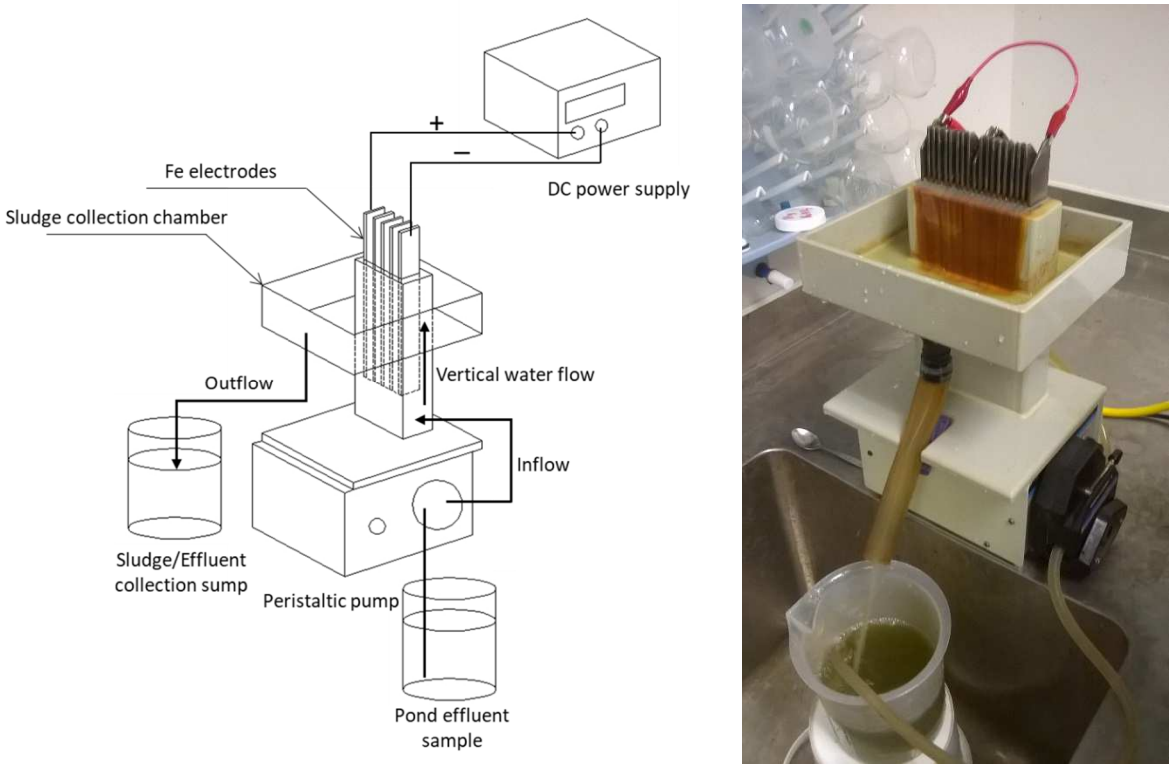

Figure 3: Diagram and photograph of the laboratory-scale electrocoagulation (EC) unit.

Table 1: The EC operation conditions of the thee laboratory experiments including flow rate, EC current, current density, voltage, interelectrode voltage, total power, power use.

Operation Variables		Experiment 1			Experiment 2			Experiment 3		
Flow rate (m^3/d)						1.4				
EC C\current (A)		0.4	0.8	3	0.4	0.8	1.2	0.8	1.6	2.4
Current density (A per L/min)		0.2	0.4	1.0	0.4	0.4	0.6	0.4	0.8	1.2
Voltage (V)		96	106	88	210	115	130	78	108	150
Interelectrode voltage (V)		2.7	2.9	1.6	11.7	3.2	3.6	2.2	3.0	4.2
Total power (kW)		0.04	0.08	0.26	0.08	0.09	0.16	0.06	0.17	0.36
Lab power use	(kWh/d)	0.9	2.0	6.3	2.0	2.2	3.7	1.5	4.2	8.6
	(kWh/m^3)	0.6	1.4	4.4	1.4	1.5	2.6	1.0	2.9	6.0

Table 2: Summary of the initial wastewater pond water characteristics for three laboratory-scale EC trials

	Expt 1	Expt 2	Expt 3
Temperature	21.3	17.8	17.3
pH	6.7	6.8	7.2
Conductivity (µS/cm)	411	302	377
D.O. (%)	99	95	66
Turbidity (NTU)	108	82	99.6
TSS (mg/L)	240	240	210
COD (mg/L)	90	-	187
BOD_5 (mg/L)	45	58	107
Soluble BOD_5 (mg/L)	4	5	5
TKN (mg/L)	40	24	44
NH_4-N (mg/L)	13	0.01	22
NO_x-N (mg/L)	0.1	0.1	0.09
TP (mg/L)	2.8	2.4	3.2
DRP (mg/L)	0.86	0.02	1.1
E. coli (MPN/100ml)	$7.9x10^4$	$5.4x10^4$	$1.6x10^5$

3 RESULTS AND DISCUSSION

3.1 EFFECTS OF ELECTROCOAGULATION ON PHYSICO-CHEMICAL WATER QUALITY PARAMETERS

Water temperature and pH increased, with a larger increase at higher EC current during the Experiments (Figure 4a and b). For example, in Experiment 3, an increase in the EC current from 0 (control) to 2.4A increased the water temperature by ~7˚C from 17.3 to 24.7˚C.

Water conductivity decreased slightly with an increase in EC current as a result of decreases in dissolved irons (e.g., PO_4^{3-}, NO_2^-, or NO_3^-) (Figure 4c). Water D.O concentration decreased with increasing current density (Figure 4d).

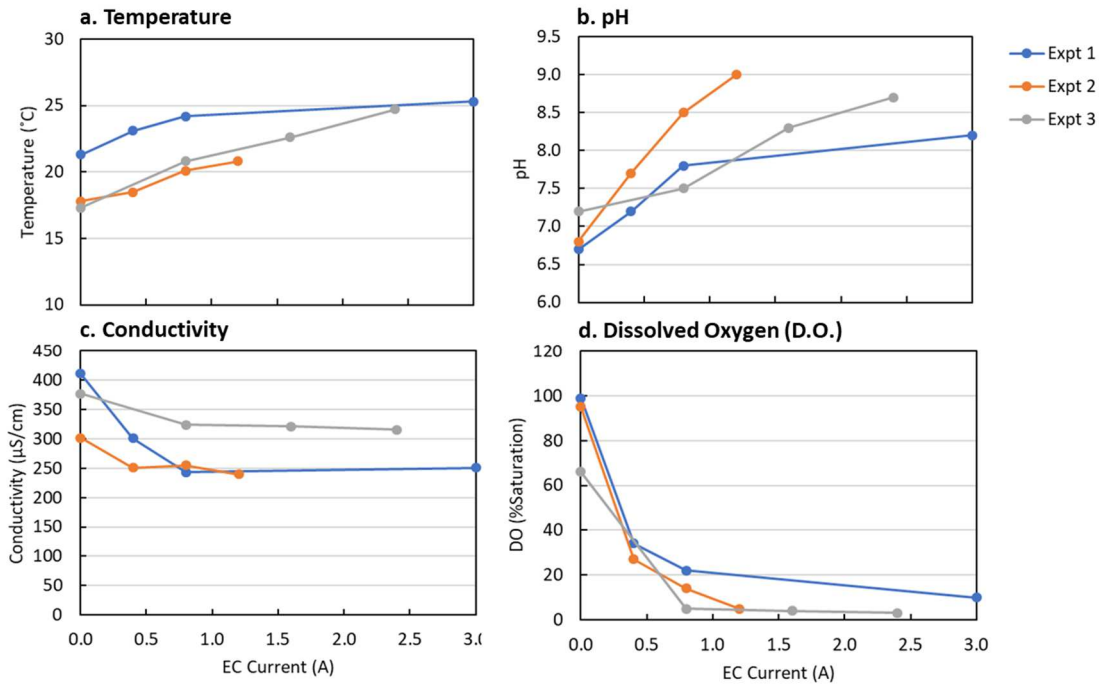

Figure 4: Effects of electrocoagulation current on the water temperature, pH, conductivity and dissolved oxygen (D.O).

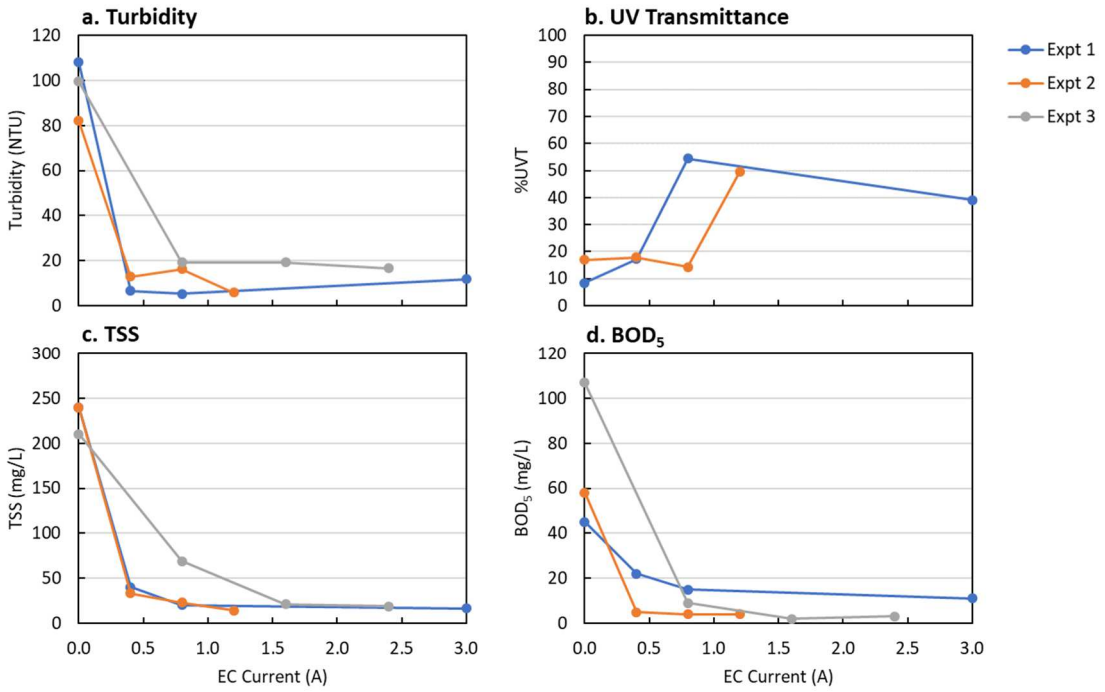

Figure 5: Electrocoagulation removal of turbidity, TSS and BOD_5 in wastewater pond effluent.

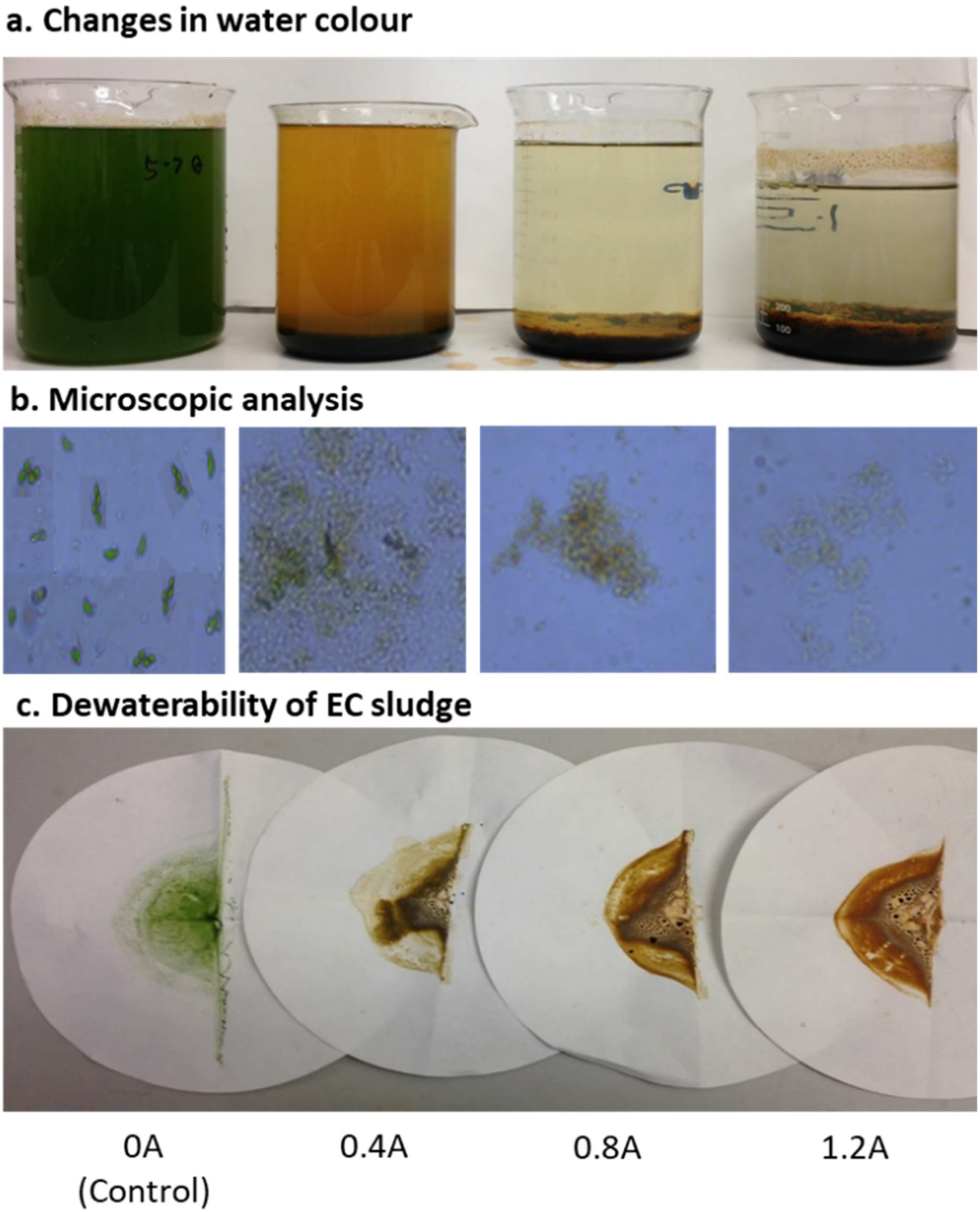

Figure 6: Changes in water colour, destruction of algal cells, and dewaterability of EC sludge with increasing EC current (Experiment 2).

3.2 SUSPENDED SOLIDS AND ORGANIC MATTER REMOVAL

The results of electrocoagulation experiments for the removal of turbidity, TSS and BOD_5 in wastewater pond effluent are shown in Figure 5.

Water turbidity decreased with increasing EC current (Figure 5a), resulting in an increase in the water UVT (ultra violet light transmittance) up to 55% with an EC current of 0.8A (Figure 5b). As shown in Figure 6, water colour changed from algae green to iron red (an indication of incomplete EC), and then to clear as the EC current increased up to 1.2A. Microscopic examination of these water samples showed that the algae cells were destroyed, leaving flocculated cell particles with increasingly less internal contents.

TSS and $cBOD_5$ concentrations all decreased with increasing current density during Experiments 1 to 3 (Figure 5c and d). For example, in Experiment 2, >90% removal of TSS (from 240 to 23 mg/L) and $cBOD_5$ (from 58 to 4 mg/L) was achieved with the EC current at 0.8A. Soluble $cBOD_5$ also showed some decrease (from 5 to 1.9 mg/L in Experiment 2), but this was much less than for the other water quality measures. To remove >90% of TSS and $cBOD_5$ required the power consumption of ~0.4-0.7 kWh/m^3 and ~0.3-0.4 kWh/m^3 respectively.

Increasing dewaterability of the EC sludge with greater higher current density is illustrated in Figure 7 2. Note the increasing separation of the dried sludge into granules (due to its hydrophobic nature) with higher current density.

3.3 NUTRIENT AND *E. COLI* REMOVAL

TKN was effectively removed from the wastewater pond effluent at an EC current of 0.4A (Experiment 1: from 40 to 11 mg/L; Experiment 2: from 24 to 4.9 mg/L). However, little further TKN removal was achieved at higher current densities (Figure 7a). This laboratory EC study showed that ~50-80% TKN removal required the power consumption of ~0.2-0.4 kWh/m^3. EC treatment slightly increased the NH_4 concentration (e.g., from 1 to 1.1 mg/L in Experiment 2) and did not affect the concentration of nitrate or nitrite. Both DRP and TP were effectively removed by EC (Figure 7b and c), (~99% DRP removal at ~0.2-0.4 kWh/m^3 and ~90% TP removal at ~0.2-0.7 kWh/m^3) with little further decrease at higher current densities. A 3-log removal of *E. coli* was possible with EC treatment at a power consumption of ~0.7-1.1 kWh/m^3. EC treatment removes *E. coli* by rupturing their membranes and then coagulating them into settleable flocs (Diao et al. 2004; Elazzouzi et al. 2017), with increasing *E. coli* reduction at higher EC current.

The results of EC treatment of the wastewater pond effluent, and power consumption and costs are summarized in Table 3. Overall, the laboratory-scale experiments showed that >90% removal of TSS, BOD_5 and TP, >95% removal of DRP, 50-80% removal of TKN, and 2-3 log removal of faecal coliforms can be achieved by EC treatment at a EC current of 0.8-1.6A tested. Assuming the full-scale power use of an EC unit is about a quarter of lab-scale (PWS 2018), a power consumption would be ~0.4 kWh to treat 1 m^3 of wastewater pond effluent. Based on the current average power cost of ~NZ$0.30/kWh (April 2018), the power cost of a full-scale EC unit would be only NZ$0.12/$m^3$.

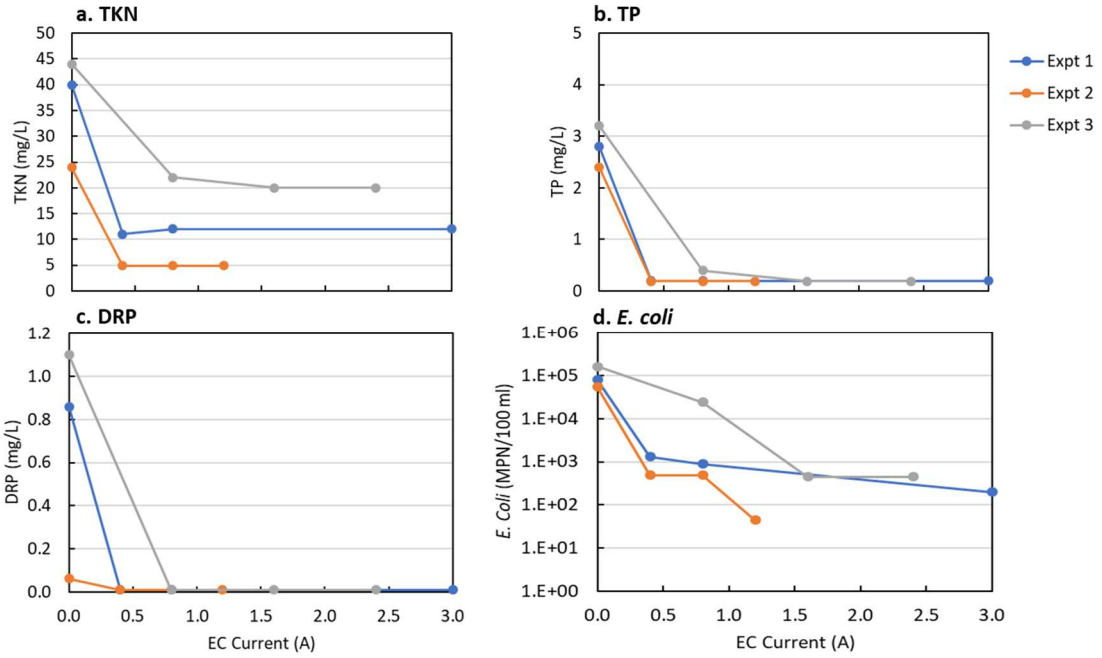

Figure 7: Electrocoagulation removal of TKN, TP, DRP and *E. coli* from wastewater pond effluent.

Park et al. (2019) reported that the cost of chemical flocculation (using cationic polyacrylamide, PAM) was ~NZ$0.05 NZD/m^3 of wastewater pond effluent to achieve >50% TSS and about 1-log *E. coli* removal. This suggests that the operation cost of the EC unit (excluding plate costs) would be about 2.5-fold more expensive than that of the chemical flocculation. However, EC provides combined removal of organic matter, phosphorus as well as disinfection, and the EC sludge is highly dewaterable.

Table 2: Summary results of EC treatment performance, full-scale power consumption and cost.

Water quality variables	% Removal	Full-scale power consumption (kWh/m^3)	Full-scale power cost ($/m^3)
TSS	>90	~0.4-0.7	~0.06-0.15
cBOD$_5$ (g/m^3)	>90	~0.3-0.4	~0.14-0.15
TKN (g/m^3)	50-80	~0.2-0.4	~0.06-0.15
TP (g/m^3)	>90	~0.2-0.7	~0.06-0.15
DRP (g/m^3)	>99	~0.2-0.4	~0.06-0.15
E. coli (MPN/100 ml)	3-log	~0.7-1.1	~0.15-0.26

4 CONCLUSIONS

This laboratory study investigated the use of electrocoagulation for the treatment of wastewater pond effluent in terms of TSS (mainly algal solids), organic matter (BOD$_5$), nutrients (N and P) and *E. coli* removal. This study showed that the laboratory-scale EC typically achieved >90% removal of TSS, BOD$_5$ and TP, >95% removal of DRP, 50-80% removal of TKN, and 2-3 log removal of faecal coliforms at an EC current of 0.8-1.6A tested.

A power consumption and cost of a full-scale EC unit would be ~0.4 kWh/m^3 and only NZ$0.12/m^3 (based on the current average power cost of NZ$0.30/kWh). The operation

cost of the EC would be about 2.5-fold more expensive than that of the chemical flocculation (based on the use of cationic polyacrylamide to achieve 50% TSS and >1 log *E. coli* removal). This research indicates that EC is an efficient and potentially cost-effective option for treating wastewater pond effluent, since the EC can provide a combined removal of organic matter, phosphorus and disinfection (potentially replacing chemical flocculation/coagulation and UV treatment), as well as producing a readily dewaterable sludge.

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