

Enhanced-Electrocoagulation For Organic Matter Removal From Surface Drinking Water Sources: Operational Parameters And Energy Consumption

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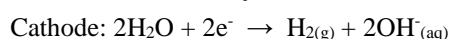
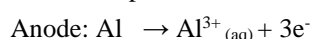
Abstract. In this study, an Enhanced Electro-Coagulation (En-EC) technique is described for the removal of dissolved organic carbon (DOC) from surface drinking water sources. Assessment of the En-EC technique included investigation of the impacts of operational factors on the organic removal efficiency (R%) and energy consumption (C_E). A decision support system for En-EC control was developed, referred to as Water Treatment Control for Electro-Coagulation (WTC-ECoag). Experiments were conducted using two surface water samples with contrasting water qualities. An electrocoagulation (EC) cell equipped with the capability for measuring consumed coulombs (C) during the process was used. Three different sets of experiments were conducted to investigate the impact of process parameters (i.e. the electric current (I_{EC}), the pH during EC process (pH_{EC}), and the coagulant dose) on the R% and C_E. The R% was determined during the process based on absorbance at 254 nm (A₂₅₄). The C_E was calculated through the monitoring of the I_{EC} and electric potential during the process. In the I_{EC} impact study, similar R% but different C_E results (under constant coagulant dose and pH conditions) were obtained for different applied I_{EC}. The investigation of pH_{EC} impact (under constant coagulant dose and I_{EC} conditions) indicated an optimum pH_{EC} of 6 for the process. Investigation of the impact of coagulant dose (under constant pH and I_{EC} conditions) indicated a maximum R% of 73% and 92% for low and high DOC waters, respectively. It is concluded that En-EC has potential as an alternative to conventional dosing processes for enhanced coagulation.

Keywords: Enhanced Electrocoagulation, Dissolved Organic Matter, Surface Drinking Water sources, Energy Consumption, Coagulant dose control system.

1. Introduction

In many parts of Australia, drinking water is supplied by treating surface waters including rivers and reservoirs. Depending on the catchment, surface waters contain various concentrations of dissolved organic matter (DOM). This includes organic compounds that are hydrophobic in nature, such as humic substances, and hydrophilic compounds such as protein-like compounds (Eish and Wells 2006; Sepp *et al.* 2019). The coagulation process removes more efficiently the hydrophobic and higher molecular weight compounds which are the major precursor for disinfection by-products (DBP) formation during the chlorination (disinfection) process as compared to the hydrophilic and lower molecular weight compounds (Tak and Vellanki 2018; Hussain *et al.* 2019). Some DBP's in chlorinated drinking waters have been reported to have the potential to cause cancer in digestive and urinary tracts in humans (Singer 1999). The majority of humic DOM can be removed by enhanced coagulation which is a procedure where a sufficient chemical coagulant dose along with pH control is applied to ensure maximum organic matter removal (Cui *et al.* 2020). However, enhanced coagulation (EnC) is reported to be associated with potential drawbacks including, a large amount of sludge production, higher safety risk associated with handling the chemicals supplies and low efficiency for protein-like compounds removal (Chafi *et al.* 2011; An *et al.* 2019). Electrocoagulation (EC) is an electrolytic process that involves the *in-situ* generation of a coagulant with the supply of an electric current and a sacrificial metal electrode such as aluminium or ferrous which liberates Al³⁺ or Fe²⁺ (Garcia-Segura *et al.* 2017).

The reactions during the EC for the aluminium electrode as an example can be described as follows:



In the solution: $\text{Al}^{3+}_{(\text{aq})} + 3\text{H}_2\text{O} \rightarrow \text{Al}(\text{OH})_3(\text{s}) + 3\text{H}^{+}_{(\text{aq})}$

The reported advantages and disadvantages associated with the EC process in comparison to the conventional chemical coagulation (Hashim *et al.* 2017) need more investigation in the case of DOC removal in drinking surface water sources. In this study, an Enhanced Electro-Coagulation (En-EC) technique is described that is based on the concept of maximizing DOC removal through sufficient dosing of an electrically produced aluminium (metallic) coagulant, with pH control during the EC process. The study aimed to determine the impacts of I_{EC} , pH_{EC} , and coagulant dose on the DOC removal efficiency (R%) and energy consumption (C_{E}) in the En-EC process.

2. Materials and methods

Water samples used in this study were collected in September 2020 from the Murray River (Murray Bridge, SA, Australia) and the Myponga River (Myponga, SA, Australia). The water qualities of the samples are presented in Table 1. Samples were maintained at 4°C and experiments were conducted within four weeks of the sample collection.

We used a laboratory-scale EC unit, consisting of a DC power supply (GPS-18500, GwInstek, Australia), eight aluminium electrodes, a 2-litre jar, magnet stirrer (NS100-S-V2, New Star Environmental, United States) and a developed coulomb counter device. The aluminium electrodes (Alloy no. 6063) were flat bar strips with an effective size of $3 \times 32 \times 105$ mm.

A compatible decision support system for coagulant dose control during the EC process termed WTC-ECoag was adopted and applied in this study. The WTC-ECoag was used to predict the optimum aluminium dose during the EC process and translating this datum to the number of required coulombs. WTC-ECoag was developed based on the existing WTC-Coag software¹ which is previously developed by Van Leeuwen *et al.* in 2009 (Van Leeuwen *et al.* 2009; Mussared *et al.* 2014). The WTC-Ecoag developed through the assumption of the similarity of the performances in enhanced coagulation using alum (chemical) coagulant and En-EC using aluminium electrodes. The consumed coulombs were monitored during the process to achieve the desired coagulant dose using a developed coulomb meter. The C_{E} was monitored and recorded during the process using a developed precise electricity meter.

DOC measurements were conducted by a TOC analyser (Model 820, Sievers Instruments, USA). A spectrophotometer (Model UV-120, Miostech, Australia) was used to measure UV absorbance at 254nm, A_{254} (m^{-1}) and true colour (HU) at 456nm. A YSI EXO2 sonde equipped with an fDOM probe (Xylem instructions, USA)

¹WTC-Coag software (an updated version of mEnCo software) is a model-based decision support system for coagulant dose prediction in conventional water treatment process based on raw water quality parameters of A_{254} , colour, and turbidity.

²The constant coagulant dose equal to almost 50% of the enhanced coagulation dose predicted by WTC-Ecoag was selected for I_{EC} and pH_{EC} experiments. The 50% value was

was used for fDOM analysis. Samples were filtered before DOC, UV absorbance, and true-colour analyses, using 0.45µm polyether-sulfone membrane filters (Sartorius, Germany).

Experiments were conducted using 2 litres of samples and a jar testing procedure. The process comprised four stages being electro-coagulation, flocculation, settling, and filtration. The electro-coagulation (first stage) process time was in the range of 10-150 minutes depending on the selected I_{EC} and required coagulant dose used in the experiment. The processing times were 14 and 15 minutes for the flocculation and settling processes, respectively. The filtration stage was then simulated using a Whatman® Grade 1 filter paper. The pH was monitored during the process using a multi-meter (MultiLab 4010-3W, YSI Inc., USA) equipped with a pH probe. HCl (0.1 M) and NaOH (0.1 M) solutions were used to adjust the pH to a target value during the EC process. The R% was calculated through A_{254} removal during the treatment process.

The I_{EC} impacts on the process have investigated at the range of 25 to 750 mA in constant coagulant dose² and pH^3 conditions. The pH_{EC} impact on the process was investigated in the range of 5-9 for each water sample at the constant dose and I_{EC} conditions. The coagulant dose influences on the process were investigated for each sample in the range of 25% to 250% of the enhanced coagulation dose which was estimated by WTC-ECoag based on the required water quality parameters as the inputs for the software to predict the coagulant dose (i.e., turbidity, colour, and A_{254} values) (Table 1).

Table 1. Water quality of raw samples

	Murray River	Myponga River
Cond. ($\mu\text{S cm}^{-1}$)	260	550
Turbidity (NTU)	45.7	12.1
DOC (mg L^{-1})	4.8	17.4
A_{254} (m^{-1})	13.7	90.3
True colour (HU)	14	170
SUVA ₂₅₄ *	2.9	5.2
fDOM (RFU)**	17.8	112.6

*DOC-specific A_{254} ($\text{L mg}^{-1} \text{m}^{-1}$)

**Fluorescent Dissolved Organic Matter (fDOM) content in relative fluorescence unit (RFU).

3. Results and Discussion

Analyses of the water samples confirmed the high contrast between them, for organic concentration and character, and turbidity (Table 1). The sample from the Myponga River, with higher SUVA₂₅₄ and fDOM value, contained a higher ratio of coagulable organic matter.

The required coagulant doses for the En-EC process were calculated using WTC-ECoag software as 60.1 Coulomb

selected arbitrary and due to its more probable sensitivity for DOC removal in comparison to higher coagulant dose where the major portion of coagulable DOC content will be removed.

³The constant pH value of 6 was selected for I_{EC} experiments which is accepted as optimum pH value for alum coagulant in the enhanced coagulation process.

liter⁻¹ (C L⁻¹) i.e., equivalent to 5.6 mg L⁻¹ Al; and 176 C L⁻¹ i.e., equivalent to 16.4 mg L⁻¹ Al, for Murray River and Myponga River samples, respectively.

The results of the investigation of the I_{EC} impacts on R% and C_E during the En-EC process are presented in figures 1a and 1b, respectively. For the I_{EC} range studied, the I_{EC} has an insignificant⁴ impact on the R% (Figure 1a) whilst the higher I_{EC} can cause significantly higher C_E during the process (Figure 1b). This indicates that the coagulant dose is the determinant parameter for R% during the En-EC process in comparison to the electrical potential applied on the electrodes or the speed of getting the required coagulant dose measured by I_{EC}. However, to achieve a faster process (less residence time), higher applied voltage (electric potential) and energy consumption will be required which can be exacerbated by higher faradaic voltage loss in higher electric current density (Gu *et al.* 2009). The optimum I_{EC} value for both residence time and energy consumption are different for the two studied samples (Figure 1b). A higher optimum I_{EC} value of around 0.1 (A) for the Myponga River sample in comparison with 0.05 (A) for the Murray River sample can be explained by almost two times higher water conductivity of the Myponga River sample that allowed it to achieve higher I_{EC} values at the same time of lower voltage and C_E.

Figures 2a and 2b show the results of the investigation on the impacts of pH_{EC} on R% and C_E during the En-EC process. The results indicate the effect of pH_{EC} on the C_E is insignificant⁵ for the pH range studied, in the case of Myponga River. A slight decrease in C_E by decrease in the pH_{EC} could be due to the greater impact of pH on the increase in the electric conductivity in the Murray River sample with a lower level of initial conductivity. The results shows that a pH_{EC} of more than 6 can significantly decrease R% during En-EC. Similar results and operational optimum value of pH around 6 has been previously reported for an aluminium based chemical coagulant (alum) in organic matter removal during enhanced coagulation process (Van Leeuwen *et al.* 2009; Matilainen *et al.* 2010).

The results of the investigation on coagulant dose impact on the R% and C_E are presented in Figures 3a and 3b for Murray River and Myponga River, respectively. The results indicate a higher C_E is associated with higher dose

production during the EC process which in turn indicates the necessity of the optimum dose control system for conserving both energy and sacrificial electrode mass during the En-EC process. In the case of organic carbon removals, the maxima R% levels of 73% and 92% were achieved for Murray River and Myponga River, respectively. The different removal efficiencies can be attributed to the different organic concentrations or characteristics of the two samples studied.

The WTC-ECoag predictions for optimum enhanced coagulation dose for the two water samples studied (i.e., 5.6 and 16.4 mg L⁻¹ Al) (Figure 3) were found to be practically close to the doses determined from the experimental data (4.7 and 14.1 mg L⁻¹) obtained by first derivative analyses of DOC vs dose using TableCurve 2D (v. 5.01, Systat Software, USA).

4. Conclusion

The findings of this study show potential of the proposed En-EC process for maximizing the removal of DOC from surface drinking water sources. Also, WTC-ECoag showed potential to be used for optimum coagulant dose prediction for the En-EC process. The insignificant impact of the En-EC on sample pH is a potential advantage over the use of acidic chemical coagulants, which may need pH control procedures. Future work will further investigate the impact of En-EC on organic pool characteristics in comparison with the enhanced coagulation process, as well as the capability of WTC-ECoag for waters with different organic matter and turbidity characteristics.

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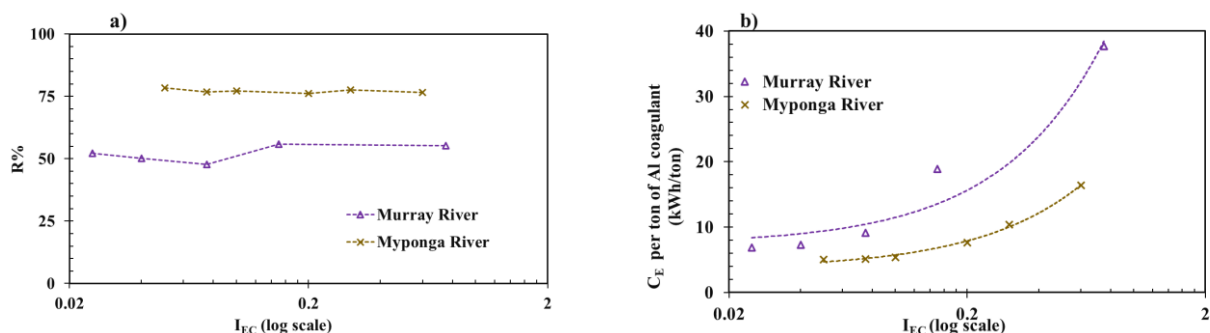


Figure 1. The influence of I_{EC} on R% (a) and C_E (b) during En-EC (pH=6 and coagulant dose of 21.5 C L⁻¹ [i.e. equivalent to 2 mg L⁻¹ Al] for Murray River sample and pH=6 and coagulant dose of 90 C L⁻¹ [i.e. equivalent to 8.4 mg L⁻¹ Al] for Myponga River sample).

⁴The average \pm SD of R% equal to 43.5% \pm 3.4% and 77.2% \pm 0.8% for Murray River and Myponga River, respectively.

⁵ The average \pm SD of C_E equal to 6.06 \pm 0.29 kWh ton⁻¹ and 5.58 \pm 0.02 kWh ton⁻¹ for Murray River and Myponga River, respectively.

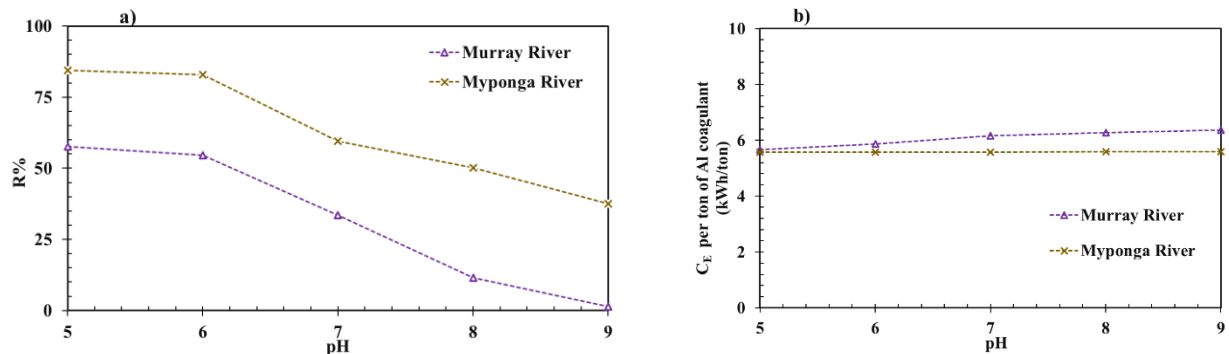


Figure 2. The influence of pH_{EC} on $R\%$ (a) and C_E (b) during En-EC ($I_{\text{EC}}=0.05$ A and coagulant dose of 21.5 C L^{-1} [i.e. equivalent to 2 mg L^{-1} Al] for Murray River sample and $I_{\text{EC}}=0.1$ A and coagulant dose of 90 C L^{-1} [i.e. equivalent to 8.4 mg L^{-1} Al] for Myponga River sample).

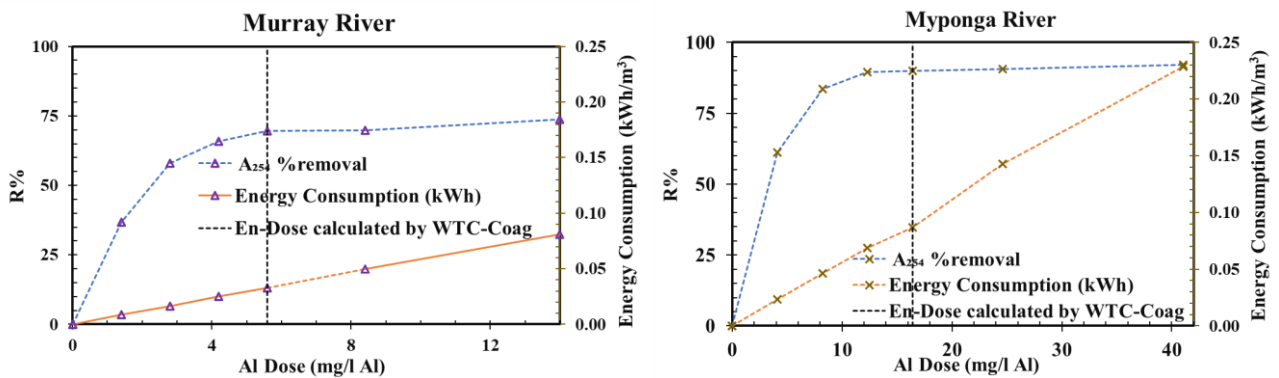


Figure 3. The influence of coagulant dose on $R\%$ and C_E during En-EC ($\text{pH}=6$ and $I_{\text{EC}}=0.05$ A for Murray River sample (a) and $\text{pH}=6$ and $I_{\text{EC}}=0.1$ A for Myponga River sample (b)).

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