

## Integrated Electrocoagulation and Tight Ultrafiltration Membrane for Wastewater Reclamation and Reuse

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### Abstract

*Wastewater reclamation and reuse have become an alternative to save operational costs while reducing the impact of waste pollution. In this paper, integration of electrocoagulation (EC) and polysulfone-based ultrafiltration (UF-PSf) membrane were used for the mentioned purpose. The EC unit equipped with 7 (seven) pieces of E-shaped Al electrodes, which operated at a current of 3 (three) Amperes and a residence time of 2 (two) hours. The waste samples were obtained from textile and oil palm industries. The experimental results were compared based on product quality and economic feasibility. When used for textile waste treatment, the integrated EC-UF units reduced TDS, TSS, BOD, and COD by 77%, 95%, 70 -80%, and 60-70%, respectively. While in palm oil waste treatment, the TDS and TSS reduced by 92% and 98%. The electrode loss rate in palm oil waste treatment was 2 (two) times greater than textile waste. By assuming that the waste production capacity of both industries was 400 m<sup>3</sup>/day, the water production cost in textile waste treatment was Rp. 4,000/m<sup>3</sup>, while in the palm oil waste treatment was Rp. 6,000/m<sup>3</sup>. These results showed that the EC-UF unit could be used as an economical and environmentally friendly alternative process for reclamation of industrial wastewater that meets the clean water quality standards.*

**Keywords:** *electrocoagulation, industrial wastewater, ultrafiltration, reclamation, reuse water*

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### INTRODUCTION

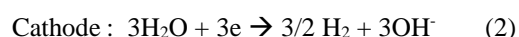
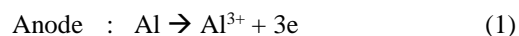
The need for clean water along with the increasing rate of population and industries impact on the increase of water production cost. This problem is getting worse by the decrease of clean water amount due to the pollution of industrial waste into the surface water source that is not appropriately handled or disposed directly into the environment without treatment. In carrying out their processing activities,

several industries use a huge amount of fresh water and even becomes the main component in producing good quality products. Indeed, high operational cost is required concerning the provision of clean water for the processes. Reclamation of the wastewater becomes one of the efforts to overcome this problem. The large amounts of wastewater can be processed into clean water and reused as process water. This concept is known as "re-use" or reuse of water concept.

Several technologies have been used for wastewater treatment, such as adsorption, oxidation, biological processing, coagulation, and filtration (Lazic *et al.*, 2017; Fatta-Kassinos *et al.*, 2016). However, these conventional waste treatments are not effective when they are applied individually (Mohan *et al.*, 2007). Biological processing is considered an efficient and economical technique in reducing organic compounds in liquid waste. Nevertheless, this method requires large operational costs and long operating time (Sarria *et al.*, 2002; Grandclement *et al.*, 2017; Stamatelatos, 2017). Meanwhile, the adsorption process is not effective to be applied in a large-scale owing to a large amount of adsorbent required as well as the complexity and cost of chemicals for the regeneration process (Robinson *et al.*, 2001; Makertihartha *et al.*, 2017; Siddique *et al.*, 2017). These problems encourage further research to find alternative technologies that are effective in dealing with industrial wastewater.

Electrocoagulation (EC) is one of the technologies used to treat industrial and domestic wastewater (Kobya *et al.*, 2007). This method has been developed as an alternative to the conventional coagulation process to minimize the use of chemicals (such as alum and ferric chloride) and overcome the problem of sludge (Un *et al.*, 2009). In addition, the EC is able to accelerate the process of charge neutralization of contaminants through the electrolysis process, and therefore, the time required for the agglomeration and settling of the particles can be shortened (Moussa *et al.*, 2017). In some cases of waste treatment with high COD and BOD value, the EC process is able to reduce COD and BOD contents up to 90% (Sridhar *et al.*, 2014). Therefore, the research on processing raw water and liquid waste based on EC is growing rapidly.

Some operating parameters play an important role in the precipitation process in EC unit, i.e., current strength, surface area of the electrode, and distance between electrodes (Susetyaningsih *et al.*, 2008). The increase of current strength in the EC process results in higher current flowed to the electrode, which enhances the number of metal decay ( $M^{n+}$ ) into the solution and subsequently converted into coagulant through chemical reaction. The reaction in EC reactor when using Al as an electrode is as follows (El-Ashtoukhy *et al.*, 2016):



The formed ions ( $\text{Al}^{3+}$  and  $\text{OH}^-$ ) react to form different types of monomeric compounds and then converted into  $\text{Al}(\text{OH})_3$  (s) as a coagulant. The coagulants ( $\text{Al}(\text{OH})_3$ ) built larger floc by binding the contaminants. The flocs are lifted to the EC reactor surface by  $\text{H}_2$  gas bubbles, which is produced in the second reaction, and then removed from the EC reactor (Bassyouni *et al.*, 2017).

The number of electrodes also affects the electrocoagulation process. The increase of electrode plate number enhances the contact between the liquid waste and the surface of electrode plate. This condition impacts the formation of more coagulants to reduce the concentration of pollutants in effluents. In addition, further developments are focused on the design or configuration of EC units. Among the development is integrating the EC and membrane-based processes (Vinduja *et al.*, 2013; Nguyen *et al.*, 2014; Zhao *et al.*, 2014). Membrane is classified as a physical separation process, which offers high selectivity at low operating pressure (Khoiruddin, *et al.*, 2017; Qu *et al.*, 2013; Wardani, *et al.*, 2017; Wenten *et al.*, 2016; Wenten *et al.*, 2017). With its high selectivity, membrane technology is able to eliminate flocs formed during the electrocoagulation process. Ben-Sasson *et al.* (2013) found that the integrated EC and membrane process was able to produce better effluent quality than used as a single unit. In addition, the use of EC before the membrane unit minimizes the formation of fouling on the membrane system. Consequently, the productivity of the membrane is kept high and extend the membrane lifetime (Wang *et al.*, 2008). The integrated EC-membrane process has been applied to treat wastewater from the textile industry. Auoni *et al.* (2009) found that a combination of EC and nanofiltration (EC-NF) process removed color by 99%, while total turbidity, COD, sodium, magnesium, potassium, chloride, and sulfate could be eliminated to 92%.

In this study, the integrated EC-UF was used to treat wastewater from textile and palm oil industries. The EC process was conducted in batch mode with a reaction time of 120 minutes. The UF membrane had a tight structure, which was prepared by dissolving polysulfone (PSf) in acetone and N-dimethyl acetamide (DMAc).

The UF membrane was prepared based on membrane formulations in the previous study (Aryanti *et al.*, 2015). As the previous study result, this UF membrane had a tight surface structure with higher selectivity to humic substances. The influences of operating parameters, such as electrolysis time and current density on effluent quality were investigated. This reclamation system is expected to produce clean water that meets the water quality standard according to Government Regulation of the Republic of Indonesia No. 82 of 2011, where the maximum BOD content of 50 mg / L, COD of 100 mg / L, and TDS of 1000 mg / L.

## MATERIALS AND METHODS

### Materials

Two samples wastewater was used in this study. The first sample was obtained from a river contaminated with textile industry waste at Cimindi Area, Cimahi, West Java, Indonesia. The contaminated river water samples obtained from three sampling points. The first point (point-1) was the

shortest distance from the output of factory waste into the river, while the second (point-2) and third (point-3) points were 50 and 100 meters from the output point, respectively. The second sample was liquid waste (sludge) from the palm oil industry in Bengkulu, Sumatra, Indonesia.

### Preparation of Flat-sheet UF membrane

The flat-sheet UF membrane was prepared by dissolving 20% of PSf in 4 % wt of acetone, 51 % wt of N-dimethyl acetamide (DMAC), and 25 % wt of PEG400. The solution was stirred until homogenous and then casted on a glass plate with a thickness of 200  $\mu\text{m}$ . The nascent flat-sheet membrane was immediately immersed in demineralized water for 14 hours to form the UF membrane structure.

### Wastewater treatment by Integrated EC-UF Unit

The schematic of the integrated EC-UF unit is shown in Figure 1 with specifications as described in Table 1. The EC process was conducted at reactor which had a volume of 15 L. The EC reactor equipped with 7 (seven) E-shaped Aluminum (Al) electrode plates (3 cathodes and 4 anodes), with the electrode distance of 4.5 cm.

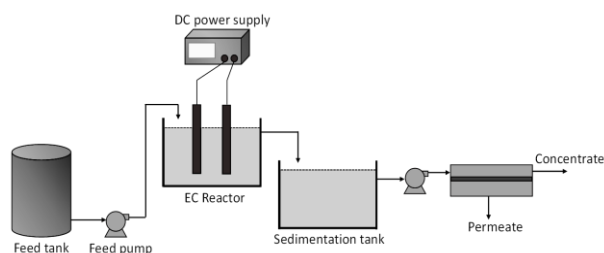


Figure 1. Schematic of the waste water treatment using integrated EC-UF system.

Table 1. Specification of laboratory EC-UF unit

Parameters	Units	Value
<b>Electrocoagulation Unit</b>		
Reactor volume	L	15
Feed flow-rate	mL/min	
Product flow-rate	mL/min	
Feed temperature	$^{\circ}\text{C}$	25
Type and shape of electrodes	-	E-shaped Aluminum
Number of electrodes	pcs	7
The total surface area of the electrode	$\text{m}^2$	0,02454
Electrode distance	cm	4,5
Applied current	Ampere	3
Residence time	minute	120
<b>Ultrafiltration Unit</b>		
<b>Membrane materials</b>		
	-	Polysulfone (PSf)
Surface area	$\text{cm}^2$	45
Feed flow rate	L/h	58
Operating pressure	bar	1
Pure water flux	$\text{L}/\text{m}^2.\text{h}$	120
Rejection of humic compounds	-	>80%

To prevent the rapid electrode decay, the applied current used in this study was low, i.e. 3 Ampere. The wastewater was treated for 120 minutes, where sampling was carried out every 20 minutes to observe the effluent characteristics. After 120 minutes, the effluent sent to the settling tank and then pumped to the UF membrane module. The membrane system was operated at constant pressure (1 bar) for 120 minutes. Prior to use, the membrane was washed with demineralized water by flowing the water into membrane module in a cross-flow mode for 30 minutes. The total dissolved solids (TSS) and turbidity of effluent were measured after the ultrafiltration finished.

### Measurement of total suspended solids (TSS), pH, and total dissolved solids (TDS)

The total suspended solids (TSS) was measured by turbidity meter (Turbi Check, Levibond). Before the sample test, the turbidity meter was calibrated using the standard solution provided by the supplier (0.02 NTU, 20.0 NTU, 100 NTU, dan 800 NTU (polymer). Meanwhile, the solution pH was measured using pH meter (Hanna Instruments HI 98107). The TDS was measured by TDS meter (TDS-3, HM Digital).

### Analysis of biological oxygen demand (BOD)

The BOD value was determined by measuring oxygen consumed in a 5-d (5-d BOD or BOD5) according to AWWA 5210-B standard method (Mecha et al., 2016). The dissolved oxygen concentration was measured by redox titration method using thiosulfate as titrant. The BOD value was calculated from the difference between the initial and final value of dissolved oxygen.

### Analysis of chemical oxygen demand (COD)

The COD value was determined using closed reflux spectrophotometry according to SNI 6989.2:2009 standard method (BSN, 2009). The sample was oxidized by  $\text{Cr}_2\text{O}_7^{2-}$  in closed reflux. The amount of oxidant demand was expressed in oxygen equivalents ( $\text{O}_2$  mg/L), which measured by UV-Vis spectrophotometry. The  $\text{Cr}_2\text{O}_7^{2-}$  was measured at a wavelength of 400 nm, while the  $\text{Cr}^{3+}$  was at 600 nm.

### Electrical power (P) calculation.

The calculation of electrical power (P) was carried out based on the following equation:

$$P = \frac{V.I.t}{1000} \quad (1)$$

where V is readable current-voltage (Volt), I is applied current (Ampere), t is operating time (seconds). In the laboratory-scale experiment, the current voltage is 22.1 Volt. In addition, the electrode loos per hour during the water treatment process was also calculated as the basis for economic evaluation.

**RESULTS AND DISCUSSION**

**The performance of EC units for waste water treatment**

The total suspended solids (TSS) value on effluent during 120 minutes of EC process is shown in Figure 3. During the wastewater treatment, the EC process reduced the turbidity of solution by 90% in textile waste and 92% in POME waste. It is suggested that the destabilization colloidal particles by  $Al^{3+}$  ions allow the coagulants ( $Al(OH)_3$ ) to bind contaminants and form larger aggregates (flocs). Thus, these flocs settle to the bottom of the EC tank and produce clear effluent at the top side.

Figure 2 presents the TDS value of wastewater during 120 minutes of EC process. It shows that the TDS value decreases with the increase of operating time. It is attributed to the coagulation process by  $Al(OH)_3$  generated from the electrolysis process. The coagulants bind the dissolved ions in solution and then settle to the bottom of the EC reactor. As shown in Figure 2 that the effluent of textile waste had a greater TDS value compared to palm oil waste because the higher content of dissolved ions (synthetic dyes) in textile waste.

The performance of EC units to reduce BOD and COD value in textile industry wastewater by EC process is presented in Figure 3. When the EC unit was used to treat the textile waste sample at point-1, the BOD value of the water sample can be reduced from 82.8 mg/L to 50.4 mg/L (or by 39%), while the COD reduced from 183.2 mg/L to 108.5 mg/L (or by 41%). The coagulants formed in situ in the EC reactor adsorbed the organic molecules in the waste solution. Thus the value of BOD and COD decreased. The same trend also obtained when the EC unit was used to treat textile waste samples at point-2 and point-3. At point-2, the BOD and COD values were decreased by 55% (from 85.1 to 38.4 mg/L) and 25% (from 108.5 to 81.41 mg/L), respectively. Meanwhile, at point-3 the BOD and COD value values were reduced by 52% (from 69.6 to 33.6 mg/L) and 47% (from 149.3 to 79.62 mg/L). These results show that the EC unit is able to produce clean water that meets the water quality standards.

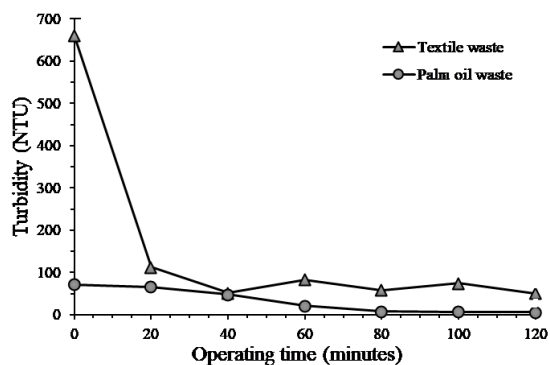


Figure 2. Profile of turbidity of waste water during 120 minutes of EC process

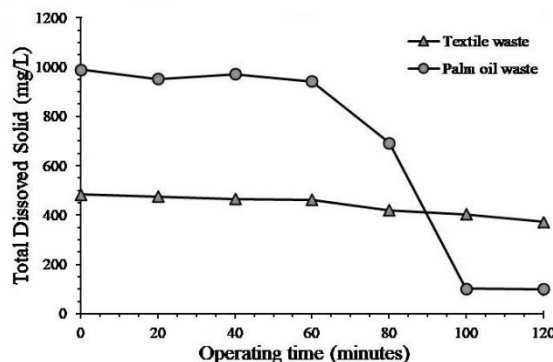


Figure 3. Profile of total dissolved solids of wastewater during 120 minutes of EC process

**The performance of EC-UF unit for wastewater treatment**

The effluent from the EC unit was further treated by UF membrane unit, which was operated at a constant pressure of 1 bar. The UF membrane unit can completely rejected the flocs formed during the EC process as well as microorganisms. The performances of the EC dan UF membrane unit in reducing TDS and TSS are shown in Figure 5 and 6. As shown in Figure 6A that the integrated EC-UF reduced the TSS value of both wastes significantly. During the textile waste treatment, the TSS value reduced by 99.7% (from 661 NTU to 1.07 NTU). A high reduction of TSS also achieved when used the integrated EC-UF unit for treating palm oil waste, i.e., by 98,5% (from 71.4 to 1.07 NTU). Meanwhile, the TDS value can be reduced by 77% in textile wastewater treatment and 92% in palm oil wastewater treatment (Figure 6B). High rejection of UF membrane during palm oil waste treatment attributed to polarization concentration phenomenon on the membrane surface.

Furthermore, the integrated UF-EC was able to reduce 70-80% BOD and 60-70% BOD levels (Figure 6). The range of BOD and COD levels was obtained based on sampling at several points. Based on these results, the UF membranes can be considered as post treatment in wastewater treatment to improve the quality of EC effluents.

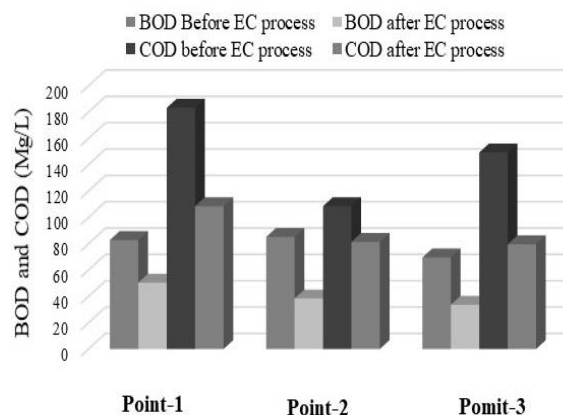


Figure 4. BOD and COD value during textile industry wastewater treatment by EC process

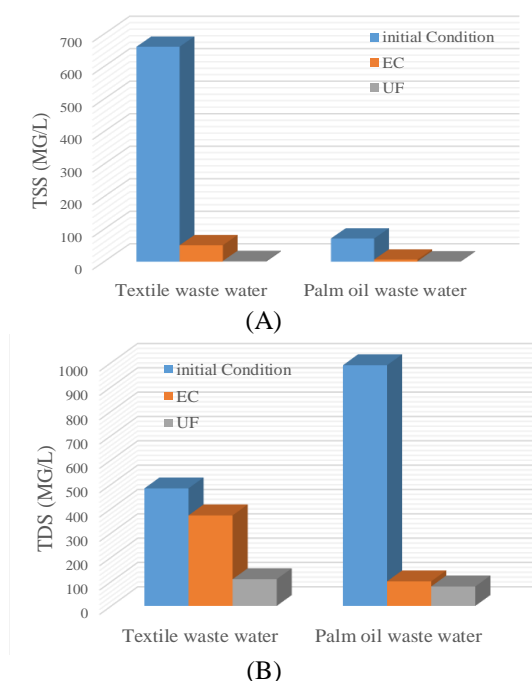


Figure 5. (A) TSS and (B) TDS values of effluent from EC and UF process

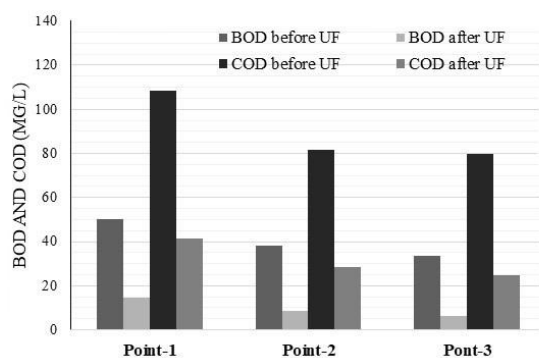


Figure 6. BOD and COD value during textile industry wastewater treatment by EC process

#### Economic Evaluation of Water Production Costs

The economic evaluation of the wastewater treatment using integrated EC-UF units has been calculated with a waste production capacity of 400 m<sup>3</sup>/day. This base was chosen based on waste production data at the smallest capacity of the oil palm industry at 30 palm fruit bunch (PFB)/hour (Abrar, 2016). Some assumptions were used in the calculations, as shown in Table 2.

Table 2. Economic evaluation base

Parameters	Assumptions
Wastewater capacity	400 m <sup>3</sup> /day
Operating time (day/hour per year)	288 day/year or 20 hour/day
Power cost (per kWh)	IDR. 1.500,00,-
Electrode cost (per kg)	IDR. 50.000,-
Total investment and operating cost	IDR. 200.000.000,-

The economic evaluation was calculated by involving capital and operational costs per year. The capital costs included equipment costs (EC units, UF-PSf, pump prices, piping and control systems). Meanwhile, the operational costs included membrane replacement costs, electricity requirements, and electrode costs. The electrode costs were calculated based on the loss of the electrode mass during the two hours of waste treatment in the EC unit. Based on the experimental results, the electrode loss during the textile waste treatment process was 0.75 grams/hour, while in POME waste treatment was 1.9 grams/hour. When it was assumed that the waste production capacity was 400 m<sup>3</sup>/day, the economic calculation showed that the production cost of water in the textile waste treatment process was IDR 4,000/m<sup>3</sup>. Meanwhile in POME waste treatment, the price of water production was IDR 6,000/m<sup>3</sup>. It has been reported that the clean water cost in Indonesia for industrial needs above 20 m<sup>3</sup>/month is IDR 12,550/m<sup>3</sup> (Pam Jaya, 2018). Hence, the integrated EC-UF unit can reduce the operational cost related to the clean water supply. Therefore, the integrated EC-UF unit can be considered as an alternative process for industrial water reclamation that meets quality standards.

#### CONCLUSIONS

A combination of electrocoagulation (EC) and ultrafiltration (UF) membrane has been used for reclamation of industrial wastewater and reused as process water as well as reducing the impact of waste pollution to the environment. During two (2) hours of textile wastewater treatment, the integrated EC-UF process reduces TDS, TSS, BOD, and COD by 77%, 95%, 70 -80%, and 60-70%, respectively. In palm oil waste treatment, the TDS and TSS can be reduced by 92% and 98%. Based on the economic evaluation of the waste treatment by integrated EC-UF at a capacity of 400 m<sup>3</sup> / hour, the price of water production in the textile industry waste treatment is IDR. 4,000/m<sup>3</sup>, while in POME waste is IDR 6,000/m<sup>3</sup>, which are lower than the price of commercial clean water. Therefore, the integrated EC-UF unit can be considered a competitive and economical technological alternative for reclamation of industrial wastewater. This unit may produce clean water that meets process water standards with low operational costs.

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