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Abstract

In this work, high removal of contaminants in hospital wastewater has been achieved using an integration of electrocoagulation (EC) with ultrafiltration (UF) and reverse osmosis (RO). In EC system, Al electrodes were arranged in a monopolar-parallel and bipolar configuration. There are two parameters studied in the EC system, i.e., the configuration of electrodes ~~(2A-2C-2B~~(2A-2-C-2B and ~~4A-2C-2B)~~4A-2-C-2B) and current densities. The EC-UF system with a configuration of ~~4A-2C-2B~~4A-2 C-2B and a current density of ~~88.5~~88.5 A.m~~$^{-2}$~~$^{-2}$ resulted in high removal of TSS, TDS, BOD, and COD by ~~95.12%, 97.53%, 95.18%,~~ 95.12 %, 97.53 %, 95.18 %, and ~~97.88%,~~ 97.88 %, respectively. The effluent quality of the EC-UF was

improved by substituting UF with RO membrane. The TSS, TDS, BOD, and COD removal were enhanced to ~~97.64%, 99.85%, 97.88%,~~ 97.64 %, 99.85 %, 97.88 %, and ~~98.38%,~~ 98.38 %, respectively. The permeate flux decline in UF membrane system was ~~47.83%~~ 47.83 % during ~~60~~60 min of filtration time due to cake layer fouling on the membrane surface, while in the RO membrane system was ~~29.49%.~~ 29.49 %. Since the EC-UF and EC-RO showed high efficiency in contaminants removal, these configurations could be used as clean technology to produce clean water for water reuse purposes. At a wastewater capacity of ~~5 m~~5 m^3.day$^{~~-1~~-1}$, the operating cost for the EC-UF system was 3.92 US$.m$^{~~-3~~-3}$, while the EC-RO system was 4.02 US$.m$^{~~-3~~-3}$. The increase of wastewater capacity to ~~50 m~~50 m^3.day$^{~~-1~~-1}$ reduced the operating cost to 0.89 US$.m$^{~~-3~~-3}$ for the EC-UF system and 0.93 US$.m$^{~~-3~~-3}$ for the EC-RO system.

Keywords:

Clean technology, Electrocoagulation, Hospital wastewater, Ultrafiltration, Reuse water

Abbreviations

No keyword abbreviations are available

1 Introduction

Hospitals require a large quantity of clean water in their daily activities, particularly for medical laboratories, sanitation, gardening, kitchens, and laundries (Rani and Singh, 2021). Based on the number of beds, the daily quantity of water is estimated from 200 to ~~1200~~1200 L per bed of inpatients (Garcia-Sanz-Calcedo et al., 2017; Khan et al., 2021). Thus, a considerable effluent of wastewater should be treated before being discharged into the environment. The hospital wastewater contains an enormous diversity of chemicals, such as pharmaceuticals, radioactive elements, endocrine disruptors, detergents, heavy metals, zinc, and pathogenic microorganisms (Zhang et al., 2020). The pharmaceutical concentration in hospital wastewater was reported to be 10 – 100 times higher than in urban wastewater (Verlicchi and Zambello, 2016). Nowadays, hospital wastewater has gained serious attention due to the SARS-CoV-2 virus, which poses a danger to human health and the environment if it is not appropriately treated (Achak et al., 2021).

Various technologies have been developed as an alternative to the conventional wastewater processes, particularly to minimize the usage of chemicals, reduce the operating time, and be environmentally friendly. These technologies are advanced oxidation processes (AOPs), integrated pressure-driven membranes (ultrafiltration/UF and reverse osmosis/RO) (Ouarda et al., 2018; Siagian et al., 2021; Tiwari et al., 2021), and biological processes (activated sludge and membrane bioreactor/MBR) (Beier et al., 2012). AOPs are a technique to eliminate organic contaminants in wastewater by generating reactive oxygen species (ROS), such as hydroxyl ($OH^{\bullet}$), sulfate ($SO_4^{\bullet-}$), or chlorine ($Cl^{\bullet}$) radicals, to produce harmless contaminants (Al Mayyahi and Al-Asadi, 2018). The oxidation process is conducted through several methods, such as ozonation (Hussain et al., 2020), photocatalysis (Liu et al., 2020), UV photolysis (Chuang et al., 2017), Fenton (Liang et al., 2021), and wet air oxidation (Deshmukh and Manyar, 2021). Souza et al. (2018) found that the combination of ozonation and UV (O_3/UV) removed total organic compound (TOC) in hospital wastewater up to ~~54.7%~~ 54.7 % after ~~120~~120 min of oxidation time at a rate of ~~1.57~~1.57 g O_3.h$^{~~-1~~-1}$. In addition, the chemical oxygen demand (COD) and aromatic reduction efficiency removal could reach ~~64.05%~~ 64.05 % and ~~81%,~~ 81 %, respectively. Segura et al. (2021) reported that the Fenton oxidation provided a high pharmaceutical removal of ~~99.8%,~~ 99.8 %, greater than the catalytic wet air oxidation (CWAO) of ~~90%.~~ 90 %. Despite high removal efficiency, the Fenton oxidation process is limited by the low pH requirement, iron sludge generated, and high chemical consumption. Further research in oxidation process is continuously conducted to minimize sludge generation and chemical consumption.

Recently, more interest has been paid to the MBR process than conventional activation sludge (CAS) due to less space requirement and higher organic removal ability (Judd, 2016; Wenten et al., 2020). Ouarda et al. (2018) combined submerged MBR (s-MBR) and electrochemical oxidation (EO). The integration of MBR-EO removed pharmaceutical contaminants by ~~97%~~ 97 % after ~~40~~40 min of treatment at electrochemical oxidation's current density of ~~0.5~~0.5 A. Furthermore, the MBR-EO eliminated venlafaxine (VEN) by ~~92%,~~ 92 %, higher than MBR or EO alone by ~~30%~~ 30 % and ~~50%,~~ 50 %, respectively. High-energy consumption and complex membrane fouling become the challenges in the large-scale application of the MBR (Do and Chu, 2022). Nanofiltration and RO have also been widely used in water and wastewater treatment, including hospital wastewater. Both technologies provide excellent performance in removing contaminants and colors in the wastewater, either standing alone or combined with other processes (Lan et al., 2018; Patel et al., 2022; Rochmah and Widiasa, 2021; Tran et al., 2019). However, high water flux decline due to fouling phenomena becomes a challenge in membrane application for wastewater treatment. The fouling formation leads to higher energy consumption and chemicals for membrane cleaning, which contributes to the increase in operational costs (Zhang et al., 2019). Ultrafiltration (UF) membrane, which is viewed as an energy-efficient technology in water and wastewater treatment, is more susceptible to irreversible fouling due to its porous structure (Karimi et al., 2020). Therefore, pretreatment processes are needed prior to the membrane processes to minimize fouling formation and maintain the productivity of the membrane.

Several technologies, such as coagulation (Alibeigi-Beni et al., 2021), adsorption (Kim et al., 2022), ozonation (Zhao et al., 2019), and electrocoagulation (EC) (Tavangar et al., 2019), have been used as a pretreatment of the membrane processes. Among these technologies, EC has increasingly been used as an alternative green process instead of chemical coagulation (Hashim et al., 2019). The EC process offers some advantages, such as less sludge generated, easy operation, short operation periods, and being environmentally friendly (Shahedi et al., 2020). The EC process removes various pollutants, colloidal, and organic matters efficiently, which may minimize the irreversible fouling and prolong the life of the membrane (Lu, Jincheng et al., 2021). Several types of membranes have been combined with the EC process for water and wastewater treatment, such as microfiltration/MF (Zhen et al., 2019),UF (Sardari et al., 2018a), RO (Den and Wang, 2008; Zhao et al., 2014), membrane distillation/MD (Sardari et al., 2018b), forward osmosis/FO (Al Hawli et al., 2019; Sardari et al., 2018c), MBR (Bani-Melhem and Smith, 2012), and electrodialysis/ED (Deghles and Kurt, 2016).

The fundamental of EC process lies on the electrolysis reaction, which requires a direct electric current to generate chemical reactions at the surface of electrodes, namely anodes and cathodes (Asaithambi et al., 2021). Aluminum (Al) and iron (Fe) electrodes are generally used in EC processes due to their availability, low cost, and form amorphous metal oxides or oxyhydroxides or hydroxides matters that offer high adsorption ability to soluble contaminants (Tegladza et al., 2021). When the electric current is applied to the electrodes, the metal ions (Fe^{3+} or Al^{3+}) are released and dissolved from the sacrificial anodes into the wastewater. At the same time, the water molecules are dissociated into H^+ and OH^- at cathodes by electrochemical reaction. There are various reactions occur simultaneously in the EC reactor. The metal ions released from the anode destabilize the colloid by charge neutralizing, allowing small aggregates to form. The other metal ions react with OH- ions and undergo spontaneous hydrolysis reactions to form amorphous precipitates, such as hydroxides ($Al(OH)_3$ or $Fe(OH)_3$), oxides (such as AlO_3 or α-FeO_3), and oxyhydroxides (such as γ-AlOOH and α-FeOOH) (Tegladza et al., 2021). The amorphous compounds bind the aggregates to form larger flocs and precipitate at the bottom of the EC reactor. Oxygen evolution reaction (EOR) may occur in the anodes, simultaneously with the dissolution of metal ions.

Meanwhile, hydrogen (H_2) gas bubbles are generated simultaneously at the cathode during the electrolysis reaction, which floats the light contaminants to the top of the reactor. The flocs produced by EC process are more stable, relatively large, and slightly bound with water (Mureth et al., 2021). Thus, it can be easily separated by the filtration method. The main reactions during EC can be summarized by the following equations (Nugroho et al., 2019):

Anode reactions:

$$M_{(s)} \rightarrow M_{(aq)}^{n+} + ne^- \quad (1)$$

$$2\,H_2O_{(l)} \rightarrow 4\,H^+_{(aq)} + O_{2(g)} + 4e^- \quad (2)$$

Cathode reactions:

$$Mn_{(aq)}^{n+} + ne^- \rightarrow M_{(s)} \quad (3)$$

$$2\,H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH^- \quad (4)$$

A few studies have been conducted by using EC process for hospital wastewater treatment. Dehghani et al. (2014) studied the influence of Fe and Al electrodes in batch EC on the removal efficiency of chemical oxygen demand (COD). The removal efficiency was up to ~~87%~~ 87 % using two pairs of Fe-Fe electrodes at a reaction time of ~~60~~60 min, pH 3, and voltage of ~~30~~30 V. By changing the configuration to two pairs of Fe-Al electrodes, the COD removal efficiency was reduced to ~~75%.~~ 75 %. Furthermore, the increase of electrode distance from 2 to ~~3~~3 cm reduced the removal efficiency from 87 % to ~~68%.~~ 68 %. Esfandyari et al. (2019) found that the higher COD and turbidity removal efficiency was achieved in neutral conditions (pH 7) compared to acid (pH 4) and alkaline (pH 9) conditions. Using 3 iron pairs as electrodes, the antibiotic (cefazolin), COD, and turbidity removal at a voltage of ~~15~~15 V and reacting time of ~~30~~30 min were ~~91.92%, 87%,~~ 91.92 %, 87 %, and ~~92,16%,~~ 92,16 %, respectively. The increase of voltage improved the removal efficiency. Other studies showed that the EC process effectively removed other contaminants in hospital wastewater, such as dexamethasone (Arsand et al., 2013) and ciprofloxacin (Ahmadzadeh et al., 2017; Yoosefian et al., 2017).

Besides the type of electrodes, electrode configuration has also played an important role in EC process (Lu, Jianbo et al., 2021). The electrodes can be assembled in monopolar (either in series (MP-S) or parallel (MP-P)) and bipolar (BP). The monopolar and bipolar electrode configurations have been described in various literature (Akter et al., 2022; Al-Raad and Hanafiah, 2021; Almukdad et al., 2021). The mode of electrode connection contributes to the efficiency of contaminants removal and energy consumption (Xolov, 2021). Several studies have focused on using BP electrodes due to their simplicity, easy maintenance, less electrical connection to the electrodes, and less maintenance cost (Nippatlapalli and Philip, 2020; Qi et al., 2020; Tchamango and Darchen, 2018). In BP electrodes, the two outer electrodes are connected to the external power supply. Meanwhile, the internal sacrificial electrode, which is called BP electrodes, are not interconnected and each side of the electrodes performs simultaneously as an anode and a cathode. Compared to the monopolar configuration, bipolar electrodes showed higher removal efficiency but required higher energy consumption for the same effluent quality (Golder et al., 2007; Khaled et al., 2019). The higher energy consumption can be attributed to the longer distance between the electrodes connected to an external electric current source, resulting in higher resistance to mass transfer and lower kinetics of charge transfer (Khaled et al., 2019). Therefore, in this research, a combination of monopolar and bipolar electrodes was used to produce high removal efficiency of EC process and narrow the distance between the electrodes connected to the current source. The EC process was equipped with a low-rate agitation process to enhance the reaction rate in EC reactor during the real hospital wastewater treatment. In most studies, the monopolar electrodes consisted of one pair of anode and cathode. In this study, the number of anodes and current density were varied to investigate the influence of both operating conditions on effluent quality after the hospital wastewater treatment. In addition, the EC process was integrated with a polypropylene (PP) cartridge filter, ultrafiltration (UF), and reverse osmosis (RO) membranes to improve the effluent quality. The performances of EC-UF and EC-RO units were compared, both the effluent quality and the large-scale economic evaluation. In addition, the EC process was integrated with polypropylene (PP) cartridge filter, ultrafiltration (UF), and reverse osmosis (RO) membranes to improve the effluent quality. The performances of EC-UF and EC-RO units were compared, both the effluent quality and the large-scale economic evaluation.

2 Material and ~~Method~~method

2.1 Materials and ~~Experimental Set-Up~~experimental set-up

The wastewater was collected from one of the hospitals in Cimahi, West Java, Indonesia, without any pretreatment. The initial condition of the wastewater is shown in Table 1.

alt-text: Table 1

Table 1

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Initial condition of the hospital wastewater.

Parameter	Unit	Value
TDS	mg.L~~-1~~	974 – 995
TSS	mg.L~~-1~~	328 – 381
BOD	mg.L~~-1~~	520 – 801
COD	mg.L~~-1~~	176–281
pH	–	7–8

The EC system used in this research refers to the previous work (Nugroho et al., 2019; Nugroho et al., 2021). The cylindrical EC reactor had ~~26~~26 cm in diameter and was occupied by a six-blade turbine with a constant agitation rate of ~~70~~70 rpm. The Aluminum electrodes were used as baffles, which were arranged in 4 configurations involving monopolar and bipolar electrodes. The dimension of each electrode was ~~33~~33 cm (height) x ~~4~~4 cm (width) x ~~3~~3 mm (thickness). The wastewater flow rate to the EC reactor was set at ~~125~~125 mL/min with a residence time of ~~1~~1 h~~hour (60~~ (60 min). A 20-inch polypropylene (PP) cartridge filter ~~(5~~(5 µm), filled with ~~300 grams~~ 300 g of manganese sands in the tube side, was placed in a 20-inch blue housing.

The UF membrane was polysulfone-based hollow fiber and supplied by GDP Filter Indonesia. The UF membrane had an outside/inside diameter of ~~2.2/1.8~~2.2/1.8 mm. A bundle of hollow fiber membranes was assembled in a 2 in ~~(0.0508~~(0.0508 m) diameter of PVC pipe with a module length of ~~30~~30 cm~~cm (0.3~~ (0.3 m). The total effective area of the UF membrane was ~~1.13 m~~1.13 m^2.module~~-1~~ $^{-1}$. Meanwhile, the spiral-wound RO (RE-2012) was provided by CSM Membrane. The RO membrane had a total membrane area of ~~1.95 m~~1.95 m^2. The schematic of EC and membrane systems is shown in Fig. 1.

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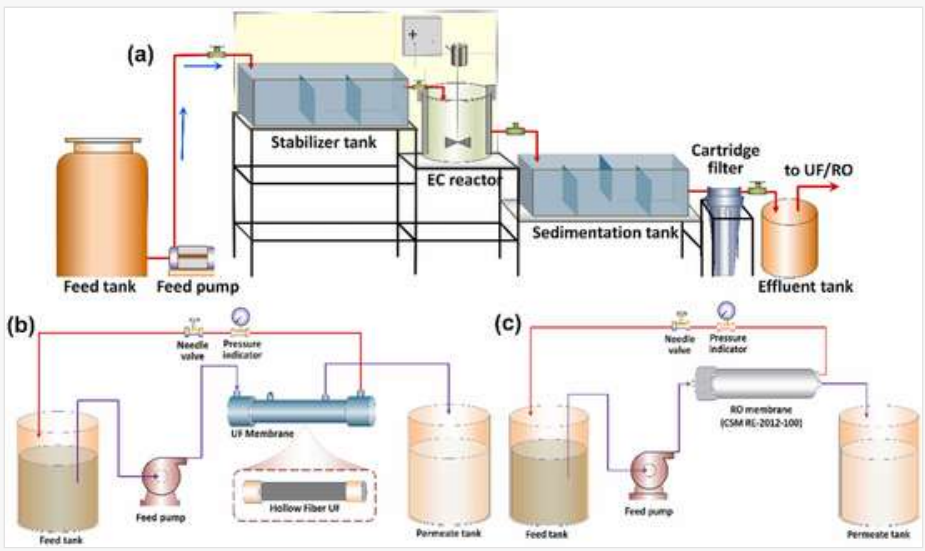
alt-text: Fig. 1

Fig. 1



Schematic of experimental apparatus for (a) electrocoagulation process, (b) ultrafiltration process, and (c) reverse osmosis process.

2.2 Hospital Wastewater Treatment by Electrocoagulation (EC) Process

The EC process treated ~~12.9~~12.9 L of wastewater per hour with 4 electrode configurations involving monopolar and bipolar configurations. The electrode configurations are 2 anodes 2 cathodes 2 bipolar ~~(2A-2C-2B)~~(2A-2-C-2B) and 4 anodes 2 cathodes 2 bipolar ~~(4A-2C-2B),~~(4A-2-C-2B), which were operated at different current densities (110.6 with code A1, 132.7 with code A2, 73.7 with code A3, and 88.5 with code A4; in A.m$^{~~-2~~-2}$). The effluent of EC process was delivered to a cartridge filter filled with manganese sand to remove the remaining particulate matter and then analyzed in terms of COD, BOD, TDS, and TSS. In laboratory experiments, the consumption of electrodes was estimated from the weight loss of the electrodes, which was determined by the difference in weight before and after the EC process. Meanwhile, the H_2 gas production during the EC process was estimated by the following Equation:

$$Q_{H2} = I \cdot A \cdot t \cdot /F.V \quad (5)$$

where Q_{H2} is the amount of H_2 gas produced by the cathode (in mole.L$^{~~-1~~-1}$), I is the applied current density (in A.m$^{~~-2~~-2}$), A is the effective surface area of cathodes that are contacted with the liquid (in m^2), t is electrolysis time (in second), F is Faraday's constant Faraday constant (96,485.33 Coulomb.mole$^{~~-1~~-1}$), and V is the treated wastewater volume (in m^3).

On the other hand, the energy consumption in EC process was by the following Equation (Hashim et al., 2019; Niazmand et al., 2019):

$$C_{power\ EC} = U.I.t/V \quad (6)$$

where $C_{power\ EC}$ is the power consumption (in W.h.m$^{~~-3~~-3}$), I is applied current (in A), U is the cell potential (in Volt), t is the operating time (in hour), and V is the volume of the wastewater treated (in m^3).

2.3 EC effluent treatment by ultrafiltration (UF) and reverse osmosis (RO)

The UF membrane was operated in crossflow mode at a constant pressure of 1 bar, while the RO membrane was performed at a constant pressure of 4 bar. The profile of permeate flux was observed during 1 h of UF and RO process by the following Equation (Aryanti et al., 2021):

$$J = V_p/A \cdot t \quad (7)$$

where J is the permeate flux (in L.m$^{~~-2~~-2}$.h$^{~~-1~~-1}$), V_p is the permeate volume (in L), A is the effective UF or RO membrane area per module (in m^2), and t is the operating time (in h). The UF and RO membranes were used to treat the hospital wastewater for 1 h (60 min). The permeate flux of the membrane was measured every 20 min, which was denoted as J_p, and then calculated using Eq. (7). The effluent was analyzed in terms of COD, BOD, TDS, and TSS. Fouling in the membrane structure was investigated by observing the change in permeate flux during 1 h of UF, which was generally expressed as normalized flux (J/J_o). The RO process was performed under the same operating condition as EC-UF, which produced the lowest quality. The operating cost of the EC-UF system, which provided a high quality of effluent, and the EC- RO system were compared.

2.4 Fouling analysis during filtration of EC effluent by UF and RO

Hermia's models were used to determine the dominant fouling in UF and RO membranes (Ariono et al., 2018; Moreira et al., 2021; Wenten et al., 2019). This model provides a correlation between permeate flux and operating time. The fouling mechanism in membrane systems is divided into 4 types, namely cake layer, intermediate blocking, standard blocking, and complete blocking, as shown in Table 2. In cake fouling, the particles or contaminants entirely cover the membrane surface. Intermediate blocking occurs when the particles are partially blocking the membrane pore. The

particles are probably deposited on the unobstructed area of the membrane surface or onto the previously deposited particles. Standard pore blocking is a fouling mechanism when the particles are adsorbed inside the membrane pore and reduce the effective diameter of the membrane pore. The last fouling mechanism is complete bocking, where the particles completely block the membrane pore. The simplex algorithm implemented in Matlab R2015b was used to determine the fouling parameter and initial flux by fitting the curve of experimental permeate flux and calculated permeate flux based on Hermia's model.

alt-text: Table 2

Table 2

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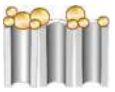
Fouling analysis equations by Hermia's model (Ariono et al., 2018).

Code	Fouling mechanisms	Figure	Linear equations	Equation no.
Model 1	Cake layer formation (code: c_f)		$J_v = \frac{J_{v,0}}{\left(2K_{cf}J_{v,o}^2 t+1\right)^{1/2}}$ $K_{cf} = kA^2$	(8)
Model 2	Intermediate blocking (code: ib)		$J_v = \frac{J_{v,o}}{K_{ib}J_{v,o}t+1}$ $K_{ib} = kA$	(9)
Model 3	Standard blocking (code: sb)		$J_v = \frac{4J_{v,o}}{\left(2K_{sb}J_{v,0}^{1/2}t+1\right)^2}$ $K_{sb} = k\,A^{1/2}$	(10)
Model 4	Complete blocking (code: cb)		$J_v = J_{v,0}\exp\left(-K_{cb}t\right)$ $K_{cb} = k$	(11)

Notes: $J_{v,o}$ is initial flux at ~~$t = 0$~~ $t = 0$ min(L.m~~$^{-2}$~~$^{-2}$.h~~$^{-1}$~~$^{-1}$), A is membrane area (m^2), t is operating time (h), and K is the fouling parameter of each fouling mechanism.

2.5 Estimation of operating cost for the EC-UF and EC-RO Process

The operating cost estimation of EC process was based on the power consumed in the electrolysis process, power for the feed pump, and electrode consumption (Kumari and Kumar, 2021), as follows:

$$Operating\ \ cost\ \ of\ \ EC = \ \ \alpha\ \ C_{power\ \ EC} + \ \ \beta\ \ W_e\ \ + \ \ \gamma\ \ C_{power\ \ pump} \tag{12}$$

where $C_{power\ EC}$ and W_e were calculated by Eqs. (1) and (2). The pump power ($C_{power\ pump}$) was calculated using Eq. (13) (Aryanti et al., 2020). The α and β were the unit price of electrical (in US$.kWh~~$^{-1}$~~$^{-1}$) and electrode (in US$.kg~~$^{-1}$~~$^{-1}$), respectively.

$$C_{power\ \ pump} = \ \ P_f\ \ x\ \ F \tag{13}$$

where P_f is the feed pump pressure (in bar) and F is the feed flow rate (in m^3.h~~$^{-1}$~~$^{-1}$). The efficiency of the feed pump was assumed to be 75 %. The operating pressure of UF and RO membranes was estimated by considering the electricity cost of the feed pump, chemicals for membrane cleaning (examples: NaOH 0.1 % and Citric acid 2 %), and membrane replacement (Aryanti et al., 2020). The recovery design of the UF membrane was 90 %, while the RO membrane was 60 %. The number of membrane modules was determined based on the permeate flux data. The membrane working live was assumed 3 years. The plant design and basic cost parameters for estimating the EC, UF, and RO membrane operating cost are detailed in Table 3.

alt-text: Table 3

Table 3

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The design and cost parameters of the EC-UF and EC-RO plants.

Parameters	Units	Values	Ref.
A. Design parameters			
1. Plant capacity	m^3.day~~$^{-1}$~~$^{-1}$	5–50	–
1. Working hour per day	h	20	–
1. UF membrane design			–
1. Permeate flux design per module	L.m^{-2}~~$^{-2}$~~.h^{-1}~~$^{-1}$~~	50	–
1. Recovery design	%	90	–
1. Effective membrane area per module	m^2	10	–
1. Operating pressure of pump (feed and backwash)	bar (kPa)	1.5 (150)	–
1. Efficiency of pump	%	75	–
1. RO membrane design			
1. Permeate flow rate per module	m^{-3}~~$^{-3}$~~.h^{-1}~~$^{-1}$~~	0.25	Engineering est.
1. Recovery design	%	50	–
1. Effective membrane area per element (length x diameter)	m^2	7.9 (40 ×4 in)	(Lenntech, 2022)
1. Operating pressure of RO pump	bar (kPa)	7 (700)	–
1. Operating pressure of CIP	Bar (kPa)	1.5 (150)	–

1. Membrane lifetime	year	2	Engineering est.
B. Operating cost parameters			
1. Al electrode	US$.kg^{-1}	1.8	(Keyikoglu and Can, 2021)
1. UF Membrane cost	US$.m^{-2}	20	Engineering est.
1. RO Membrane cost	US$.m^{-2}	40	(Choi et al., 2015)
1. Electricity	US$.kWh^{-1}	0.19	(da Silva et al., 2020)
1. Labor cost	US$.(man-month^{-1})	250	(Meratizamana and Asadib, 2020)
1. Number of labors	person	2	Engineering est.
1. Chemical cost of UF membrane	US$.m^{-3} permeate	0.01	(Bhojwani et al., 2019)
1. Chemical cost of RO membrane	US$.m^{-3} permeate	0.033	(Gökçek and Gökçek, 2016)

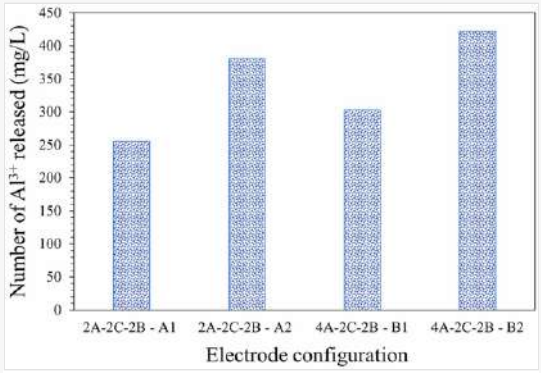
3 Results and ~~Discussions~~discussions

3.1 The amount of metal ions released from the anode and hydrogen (H_2) gas production at the cathode

The applied current density influences the metal ion released and H_2 gas production from the electrodes during ~~1~~1 h of EC process. Fig. 2 shows the influence of electrode configuration and applied current density on the theoretical number of Al^{3+} ions released from the anode. The increase of current density from 110.6 (A1) to 132.7 (A2) A.m^{-2} at the same electrode configuration of ~~2A-2C-2B~~2A-2-C-2B enhanced the number of ions released by ~~48.57%~~48.57 % (from 255.66 to ~~410.77~~410.77 mg.L^{-1}). The increase of current density improved the potential electrolysis (Galvão et al., 2020; Kamar et al., 2018), which contributed to more release of Al^{3+} ions. The highest amount of metal ions was ~~422.5~~422.5 mg.L^{-1}, which was obtained from the electrode configuration of ~~4A-2C-2B~~4A-2-C-2B at a current density of 88.5 (B2) A.m^{-2}. In addition to metal ions released by the anode, the bipolar electrode may be affected by the negative charge from the anode carried by the electrolyte molecules in the effluent. Therefore, more metal ions are released by the bipolar electrode. On the other side, H_2 gas bubbles are simultaneously generated near the cathode's surface due to water electrolysis phenomena. The amount of H_2 gas formed during the EC process is presented in Table 4, which was calculated by Eq. (5). Two parameters affected the H_2 production, i.e., the number of cathodes and applied current densities. The increase of current density at the same number of cathodes enriched the H_2 production by ~~20%~~20 % (from 168.65 to ~~202.38~~202.38 mol.L^{-1}). The increase of current density enhanced the water splitting rate near the cathode surface, and consequently, the amount of H_2 gas generated was raised.

alt-text: Fig. 2

Fig. 2



The number of Al^{3+} ions that are released from the anode.

alt-text: Table 4

Table 4

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H_2 gas production (mol.L^{-1}).

Configuration of electrodes	QH_2 (mole.L^{-1})	QH_2 (kg.L^{-1})
2A-2C-2B – A1	168.65	0.34
2A-2C-2B – A2	202.38	0.41
4A-2C-2B – B1	168.65	0.34
4A-2C-2B – B2	202.38	0.41

3.2 The influence of electrode configuration and applied current density on EC-cartridge/sand filter-UF and EC-cartridge/sand filter-RO effluent qualities

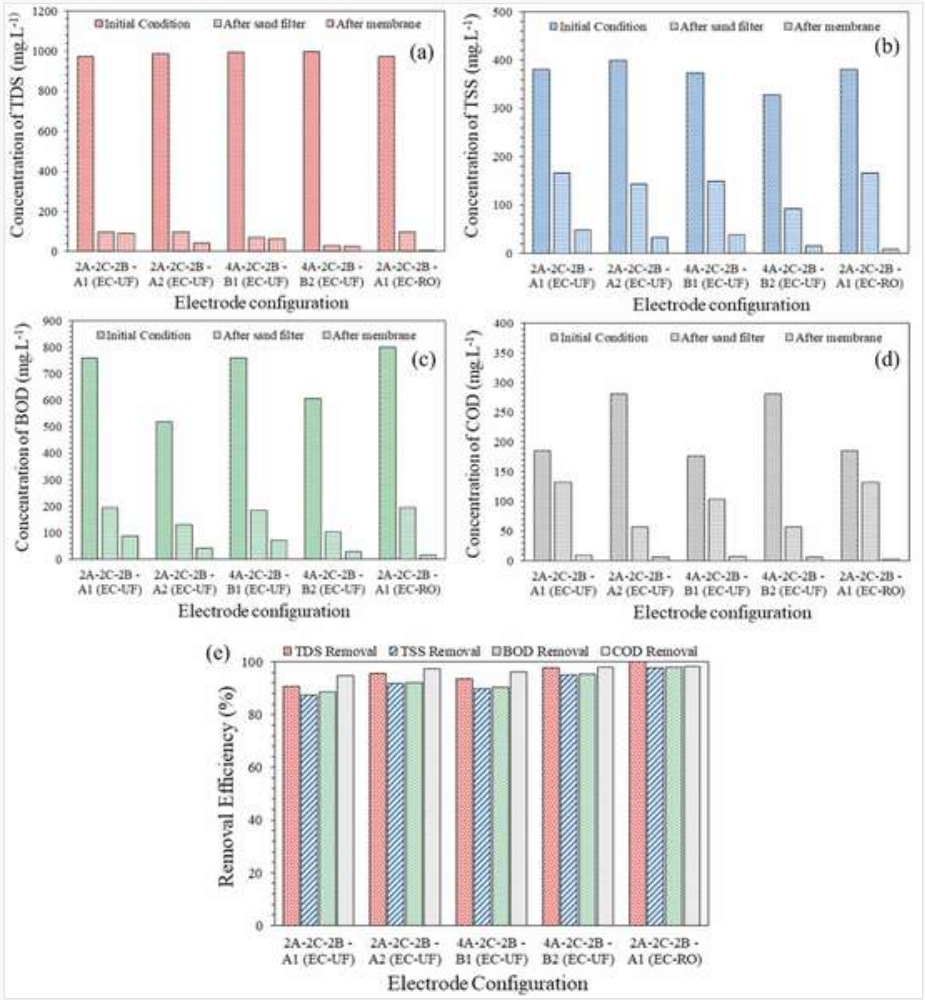
Fig. 3 shows the influence of electrode configuration and applied current density on the effluent qualities of the EC process. In this research, most of the EC processes provided high removal efficiency of contaminants removal, in terms of TDS, TSS, BOD, and COD, which was over 90%. 90 %. The lowest removal efficiency was achieved by electrode configuration of 2A-2C-2B2A-2-C-2B – A1. The TDS, TSS, BOD, and COD removals were 90.63%, 87,40%, 88.46%, 90.63 %, 87,40 %, 88.46 %, and 94.89%, 94.89 %, respectively. The change in current density from 110.6 (A1) to 132.7 (A2) A.m^{-2} at a fixed electrode configuration of 2A-2C-2B2A-2-C-2B raised the removal efficiency of the contaminants to 95.58% 95.58 % for TDS, 91.73% 91.73 % for TSS, 92.20% 92.20 % for BOD, and 97.45% 97.45 % for COD. The experimental results show that the current density plays an important role in contaminant removal. The highest removal efficiency resulted from using 4A-2C-2B4A-2-C-2B – B2. The TDS, TSS, BOD, and COD were 97.89% 97.89 % (from 996.6 to 24.5624.56 mg.L^{-1}), 95.12% 95.12 % (from 328.0 to 16.016.0 mg.L^{-1}), 95.18% 95.18 % (from 606.4 to 29.229.2 mg.L^{-1}), and 97.88% 97.88 % (from 280.6 to 5.945.94 mg.L^{-1}), respectively. Although the addition of anode number diminished the current density of electrodes, the increase in metal ions released from anodes improved the contaminant removal by the coagulation process. The experimental results

implied that the change in current density, as well as the anode number, affected the contaminant removal during the EC process.

Images are optimised for fast web viewing. Click on the image to view the original version.

alt-text: Fig. 3

Fig. 3



The concentration of contaminants before and after wastewater treatment using EC-UF and EC-RO, in terms of (a) TDS, (b) TSS, (c) BOD, (c) COD, and (d) the removal efficiency.

The contaminants removal in EC process includes a complex mechanism. It has been explained in sub-chapter 3.1 that the increase of current density enhanced the electrolysis reaction rate during the EC process. As a result, the production of Al^{3+} ions from the anodes and H_2 gas bubbles from the cathodes was raised. The Al^{3+} ions were released from the anode and dissolved into the bulk solution. A part of the ions destabilized the counter ionic species in the wastewater and led to the formation of flocs as a result of coagulation. The other Al^{3+} ions were transformed into amorphous hydroxide flocs, $Al(OH)_3$, which bound the suspended solids and organic matter in the wastewater to form larger flocs, and then settled to the bottom of the EC reactor. The increase of Al^{3+} ions in the EC process brought more contaminants removal. It has been reported in the literature that there is a competition between Al^{3+} ion dissolution and oxygen reaction evolution (EOR) during the EC process at a high applied current (Tegladza et al., 2021). The presence of EOR reaction may reduce the efficiency of Al^{3+} dissolution. In this research, the removal of the contaminant was improved when the current density was enhanced. It was suggested that the Al^{3+} dissolution offered the dominant role in agglomerating contaminants. The increase of Al^{3+} number from the bipolar electrodes could improve the number of coagulants formed in the wastewater solution. In addition, the agitation process in the EC reactor improved the distribution and mass transfer of Al^{3+} ions in the solution to destabilize and form amorphous $Al(OH)_3$. The homogenous distribution of coagulants enhanced the aggregation of the contaminants and contributed to the acceleration of coagulation. The O_2 gas oxidized organic pollutants in wastewater, improving separation efficiency.

In cathodes, the formation of H_2 gas bubbles was also influenced by the applied current, as shown in Table 4. The rise of the current increased the water electrolysis rate, which increased the bubble density in the EC reactor. It has been reported that the current density also affects the bubble size of H_2 gas (Barrera-Díaz et al., 2018). The increase of applied current reduced the H_2 bubble gas size, which facilitates high contaminants removal by H_2 gas flotation. The flotation efficiency decreases with the rise of bubble size due to less surface area and retention time of the bubbles in the solution. The bubble size distribution was not measured in the present study, but it was observed that more air bubbles are formed as the applied current increases.

The integration of EC-RO was also used to treat the hospital wastewater at an electrode configuration of ~~2A-2C-2B~~2A-2-C-2B – A1, where the UF membrane could not remove the contaminants excellently. By using RO as post-treatment instead of UF membrane, the removal efficiency of contaminants was significantly improved to ~~99.89%~~ 99.89 % for TDS, ~~97.64%~~ 97.64 % for TSS, ~~97.88%~~ 97.88 % for BOD, and ~~98.38%~~ 98.38 % for COD. The RO membrane (CSM RE-2012) had a dense layer, and consequently, high rejection of contaminants can be achieved. The performances of UF and RO membranes during the treatment of effluent from the EC process are discussed in the next sub-chapter, including the economic evaluation for both EC-UF and EC-RO configurations. The photos of EC-UF and EC-RO effluents are shown in Fig. 4. The effluents seem clear and transparent due to the high removal of TSS. The TSS of EC-UF system with electrode configuration of ~~4A-2C-2B~~4A-2-C-2B – B2 was ~~16.0~~16.0 mg.L^{+-1}. Meanwhile, the EC-RO system with electrode configuration of ~~2A-2C-2B~~2A-2-C-2B – A1 was ~~9~~9 mg.L^{+-1}. Based on the quality of the effluent produced, it can be used for sanitation and agricultural needs. The comparison of performances of several technologies for hospital wastewater treatment compared to this study is presented in Table 5. It shows that the integration of EC-UF and EC RO results in higher separation efficiency than other processes and the use of a single EC.

Images are optimised for fast web viewing. Click on the image to view the original version.

alt-text: Fig. 4

Fig. 4

Photos of (a) initial condition of hospital wastewater, (b) effluent of EC-UF ~~2A-2C-2B 12~~2A-2 C-2B 12 A, (c) effluent of EC-UF ~~4A-2C-2B 12~~4A-2 C-2B 12 A, and (d) effluent of EC-RO ~~2A-2C-2B 10~~2A-2 C-2B 10 A.

alt-text: Table 5

Table 5

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The performances of several processes for hospital wastewater treatment.

Process configuration	Operating condition	Removal efficiency (%)	Ref.
CAS bioreactor/ ultrasound	MLSS = 3000 – 8000 mg.L^{+-1} HRT = 2–8 h	COD: 92 %	(Karami et al., 2018)

	Initial condition of feed: COD: 480 – 520 mg.L^{-1} TSS: 40 – 50 mg.L^{-1} pH: 7.1 – 7.4	Effluent turbidity: 2.7 mg.L^{-1}	
Sponge-SMBR-ozonation	Membrane polymer: Membrane type: hollow fiber Membrane pore size: 0.4 μm Flux: 10–20 L.m^{-2}.h^{-1} SRT: 20 days Type of sponge: polyethylene Initial condition of feed: COD: 320 mg.L^{-1}, NOR: 16 mg.L^{-1}, CIP: 7.28 mg.L^{-1}, OFL: 21.51 mg.L^{-1}, SUL: 1.46 mg.L^{-1}	COD: > 90 % NOR: 92 % CIP: 83 % OFL: 88 % SUL: 66 %	(Vo et al., 2019)
SMBR	Membrane polymer: PVC/ZnO Permeate flux: 122.22 L,m^{-2}.h^{-1} Membrane pore size: 75–450 nm MLSS: 10,000 mg.L^{-1} SRT: 25 days	COD removal: 73.5 %	(Alsalhy et al., 2018)
Moving bed biofilm reactor (MBBR)	HRT: 24 h MLSS: 3000 mg.L^{-1} Initial feed conditions: COD: 750 – 850 mg.L^{-1} BOD: 400 – 500 mg.L^{-1} pH: 7.2 – 8.5 TSS: 300 – 400 mg.L^{-1}	BOD: 97.8 % COD: 95.6 %	(Shokoohi et al., 2017)
Supercritical water oxidation (SCWO)	Temp. in reactor: 450 °C. Pressure in reactor: 25 MPa Reaction time: 60 s Oxidant: H_2O_2 Initial condition of feed: COD: 340 – 1200 mg.L^{-1} BOD5: 300 – 570 mg.L^{-1} TOC: 64.9 – 260.3 mg.L^{-1}	COD, BOD, TOC, TN, and SS: > 90 %	(Top et al., 2020)
Coagulation. UV/H_2O_2, and activated sludge	Initial Feed condition PhACs: 0.108 mg.L^{-1}	PhACs: 83 %	(Mir-Tutusaus et al., 2021)
Coagulation. UV/H_2O_2, and fungal treatment	Initial Feed condition PhACs: 0.108 mg.L^{-1} COD: 174 mg.L^{-1} TSS: 108 mg.L^{-1}	PhACs: 94 % COD: 87 mg.L^{-1} TSS: 16 mg.L^{-1}	(Mir-Tutusaus et al., 2021)
Catalytic wet air oxidation	Catalyst: Pt supported multi-walled carbon nanotubes (Pt/CNT) Initial feed conditions: COD: 332 – 650 mg.L^{-1} TSS: 126 – 733 mg.L^{-1} pH: 6.8 – 8.7 mg.L^{-1}		(Segura et al., 2021)
Homogenous Fenton	Catalyst: dissolved $Fe(NO_3)_3$ 98 % Fe^{3+} concentration: 25 mg.L^{-1} H_2O_2 concentration: 2.123 g.g^{-1} COD Operating temperature: 70 °C Reaction time: 240 min Initial feed conditions: COD: 332 – 650 mg.L^{-1} TSS: 126 – 733 mg.L^{-1} pH: 6.8 – 8.7 mg.L^{-1}	COD removal: 70 % TOC: 50 % PhACs (130 oC): 90 %	(Segura et al., 2021)
Heterogeneous Photo-Fenton	Catalyst: Fe-BTC (Basolite F300-like semi-amorphous) H_2O_2 concentration: 1.125 g.g^{-1} COD Reaction time: 120 min Initial feed conditions:	COD: 94.5 % PhACs: 90 %	(Segura et al., 2021)

	COD: 332 – 650 mg.L^{-1} TSS: 126 – 733 mg.L^{-1} pH: 6.8 – 8.7 mg.L^{-1}		
EC	Electrodes: Fe-Al Retention time: 15 min Potential: 40 V Initial condition: COD: 502.8 mg.L^{-1} BOD_5: 136.4 mg.L^{-1} Phenols: 2.8 mg.L^{-1} TSS: 158.6 mg.L^{-1}	COD: 75.5 % BOD_5: 59.2 % Phenols: 80.7 % TSS: 75.6 %	(Yánes et al., 2021)
EC	Electrode: Fe Electrolysis time: 41 min Applied current: 2.64 A Effective area of electrode: 50 cm^2 Initial condition: COD: 377.5 mg.L^{-1} Chloride: 11.5 mg.L^{-1} pH: 7.41	COD: 54.98 % Chloride: 66.79 %	(Bajpai and Katoch, 2020)
EC-UF	Electrode: Al (combination of monopolar and bipolar) Electrolysis time: 60 min EC-UF process: Electrode configuration: 4A-2 C-2B Applied current: 12 A Current density: 88.5 A.m^{-2} Operating pressure of UF: 1 bar EC-RO process: Electrode configuration: 2A-2 C-2B Applied current: 10 A Current density: 110.6 A.m^{-2} Operating pressure of RO: 4 bar Initial conditions: TDS: 974–995 mg.L^{-1} TSS: 328 – 381 mg.L^{-1} BOD: 520 – 801 mg.L^{-1} COD: 176 – 281 mg.L^{-1}	TSS: 95.12 % TDS: 97.53 % BOD: 95.18 % COD: 97.88 %	This study
EC-RO		TSS: 97.64 % TDS: 99.85 % BOD: 97.88 % COD: 98.38 %	

Note: CAS ~~= conventional~~ = conventional activated sludge, MLSS ~~= mixed~~ = mixed liquor suspended solids, HRT ~~= hydraulic~~ = hydraulic retention time, COD ~~= chemical~~ = chemical oxygen demand, ~~SMBR= submerged~~ SMBR= submerged membrane bioreactor, ~~NOR= norfloxacin,~~ NOR= norfloxacin, CIP ~~= ciprofloxacin,~~ = ciprofloxacin, OFL ~~= Ofloxacin,~~ = Ofloxacin, SUL ~~= sulfamethoxazole,~~ = sulfamethoxazole, PVC ~~= polyvinyl~~ = polyvinyl chloride, ZnO ~~= zinc~~ = zinc oxide, SRT ~~= solid~~ = solid retention time, BOD ~~= biological~~ = biological oxygen demand, TSS ~~= total~~ = total suspended solids, TN ~~= total~~ = total nitrogen, TOC ~~= total~~ = total organic compounds, PhACs ~~= pharmaceutically~~ = pharmaceutically active compounds, EC ~~= electrocoagulation,~~ = electrocoagulation, UF ~~= ultrafiltration,~~ = ultrafiltration, RO ~~= reverse~~ = reverse osmosis

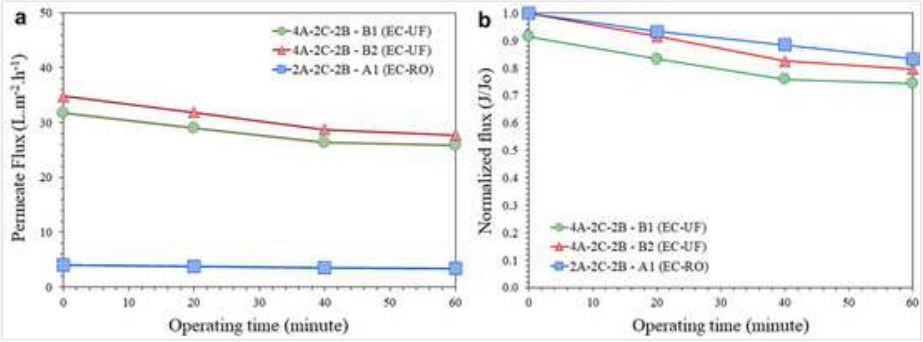
3.3 Profile of permeate flux and fouling analysis during filtration of EC effluent

Fig. 5**a** presents the profile of permeate flux during ~~60~~60 min of filtration process by UF and RO membranes. The permeate flux decline during the filtration process was evaluated through normalized flux (J/Jo) (Fig. 5**b**). Prior to the membrane, the effluent of EC was filtered by a cartridge filter to remove large particulates and reduce membrane fouling. The permeate flux was significantly reduced up to ~~20~~20 min of filtration by UF membrane, either effluent of EC with a configuration of ~~4A-2C-2B~~4A-2-C-2B – B1 and ~~4A-2C-2B~~4A-2-C-2B – B2. The flux reduction was attributed to the accumulation of contaminants on the membrane structure, which is known as the fouling phenomenon. ~~4A-2C-2B~~4A-2-C-2B – B2 (EC-UF) configuration provided higher permeate flux than ~~4A-2C-2B~~4A-2-C-2B – B1 (EC-UF). It has been explained in Section 3.2 that the configuration of ~~4A-2C-2B~~4A-2-C-2B – B2 (EC-UF) offered higher contaminant removal. Consequently, fouling resistance on or in the UF membrane structure could be minimized. After ~~60~~60 min of filtration process, the configuration of ~~4A-2C-2B~~4A-2-C-2B – B2 (EC-UF) resulted in lower flux decline of ~~47.83%~~ 47.83 % (from 53.10 to ~~27.70~~27.70 L.m^{-2}.h^{-1}) compared to ~~51.24%~~ 51.24 % (from 53.10 to ~~25.89~~25.89 L.m^{-2}.h^{-1}) for the ~~4A-2C-2B~~4A-2-C-2B – B1 (EC-UF). Meanwhile, the RO permeate flux ~~(2A-2C-2B~~(2A-2-C-2B – A1) seemed stable up to ~~60~~60 min of the filtration process compared to the ultrafiltration process. At

an operating pressure of ~~4~~4 bar, a low permeate flux was obtained at the beginning of operating time, which was equal to ~~4.76~~4.76 L.m$^{~~-2~~-2}$.h$^{~~-1~~-1}$. The flux reduction was ~~29.49%~~ 29.49 % after ~~60~~60 min of filtration.

alt-text: Fig. 5

Fig. 5



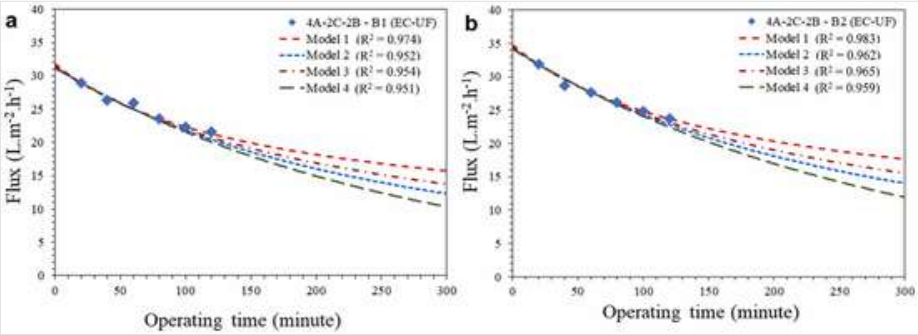
(a) Profile of permeate flux of membrane systems and (b) normalized permeate flux (J/Jo).

The difference profile of permeate flux decline between UF and RO membrane was attributed to the pore structure and type of the membrane module. The feed channel spacer inside the spiral-wound membrane RO generated local mixing flow near the membrane surface, which enhanced the mass transfer and minimized fouling on the membrane surface (Lin et al., 2020). In addition, the non-porous structure of RO could minimize the irreversible fouling formation inside the membrane structure during ~~60~~60 min of filtration. In long-term application, fouling inside the RO membrane could be formed and entrapped inside the spiral-wound system (Matin et al., 2021). Therefore, sufficient pretreatment systems are required to ensure the provision of good quality RO feedwater resulting in a stable performance for long-term application. While in hollow fiber UF membrane, a significant flux decline was found in the first ~~20~~20 min of filtration. The flux decline was attributed to the rapid accumulation of mainly organic and soluble contaminants onto the UF membrane surface. Drag permeation from feed side to the permeate side could accelerate the adsorption of contaminants to the membrane surface. Contaminants that are smaller than the membrane pores entered the membrane pores, which contributed to irreversible fouling. In addition, the entrapped contaminants between the fibers might deteriorate the flux decline in the membrane system. Periodically cleaning of the membrane, such as backwash or air scouring, is recommended to maintain membrane performances.

Hermia's model was used to predict the dominant fouling mechanisms in the membrane system. As shown in Table 1, there are 4 types of fouling mechanisms, namely cake fouling layer (model 1), intermediate blocking (model 2), standard blocking (model 3), and complete blocking (model 4). Fig. 6 shows the fouling mechanisms during ultrafiltration of EC effluent with a configuration of ~~4A-2C-2B~~4A-2:C-2B at an applied current density of 73.7 (B1) and ~~88.5~~88.5 A.m$^{~~-2~~-2}$ (B2). In both configurations, cake layer fouling (model 1) became the dominant fouling mechanism during ~~60~~60 min of the filtration process, followed by intermediate blocking, standard blocking, and complete blocking. The rapid accumulation of contaminants on the membrane surface could cover the membrane surface, particularly in at the beginning of the ultrafiltration process. The cake layer formation reduced the permeate flux significantly. While in the RO membrane system, the 4 types of fouling might occur simultaneously, which was indicated by the high value of R^2 in the 4 fouling models (Fig. 7).

alt-text: Fig. 6

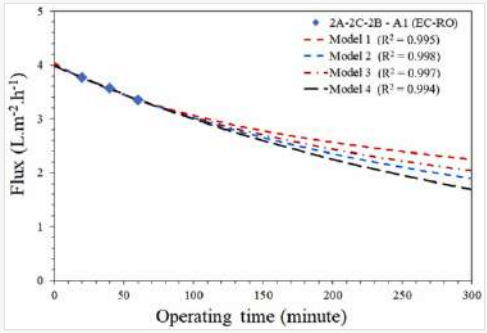
Fig. 6



Fouling mechanisms in UF membrane according to Hermia's model for difference electrode configuration di EC process: (a) ~~4A-2C-2B~~4A-2-C-2B – B1 and (b) ~~4A-2C-2B~~4A-2-C-2B – B2.

alt-text: Fig. 7

Fig. 7



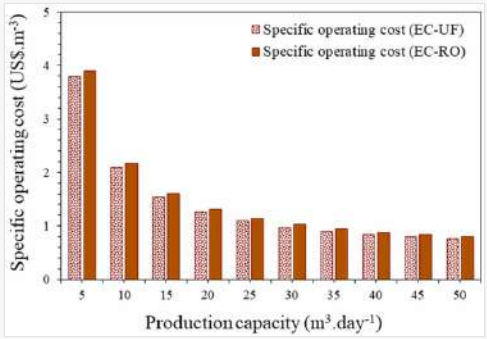
Fouling mechanisms in RO membrane according to Hermia's model.

3.4 Operating cost of EC-UF and EC-RO system

The operating costs of EC-UF and EC-RO systems were calculated by equations and design parameters in sub-chapter 2.5. The operating costs of the two hospital waste treatment designs are shown in Fig. 8. Based on the calculation results, there are slightly different between the operational cost of UF and RO membranes. It shows that the increase of production capacity from 5 to ~~50 m~~50 m^3 reduced the operating cost from 3.92 to 0.89 US$.~~m-3~~$m^{-3}$ for EC-UF system and from 4.02 to 0.93 US$.~~m-3~~$m^{-3}$ for EC-RO system. By excluding labor costs, the operating cost was from 0.56 to 0.58 US$.~~m-3~~$m^{-3}$ for EC-UF system and from 0.6 ~~-~~to 0.69 US$.~~m-3~~$m^{-3}$ for EC-RO system. The operating cost was determined by dividing the total operating cost with the production capacity. Therefore, the scaling up production capacity resulted in economics that led to a lower specific operating cost per volume of treated wastewater. Table 6 compares the operational costs of various EC processes in different wastewater treatments with this study. Most of the operating cost was calculated by Eq. (12). In this research, the operational cost calculation involved the annual cost of the energy required in EC system and pump units, electrodes, membrane replacement for UF and RO, chemicals for UF and RO membrane, and labor. The detailed cost has been presented in Table 2. Compared to other studies, the integration of EC and UF or RO can be used as an alternative process to hospital wastewater treatment, which provides high removal efficiency of contaminants. Process optimization was further required to minimize the energy costs in the EC process, such as reducing the spaces between the electrodes, varying the geometry of the electrodes, adjusting the conductivity of the electrodes, etc. (Mollah et al., 2004; Naje et al., 2017~~)~~).

alt-text: Fig. 8

Fig. 8



Operating cost of EC-UF and EC-RO systems at different production capacity.

alt-text: Table 6

Table 6

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Economic of various electrocoagulation plants for wastewater treatment and its comparison with this study.

Application	Mode and Electrode	Removal efficiency	Current density (A.m^{-2})	Operating Cost (US$.m^{-3})	Reference
Textile Wastewater	Mode: Batch Electrode: Al	Dyes: 98.6 % COD: 84 %	Voltage: 20 V Average current: 0.26 A	0.256	(Dalvand et al., 2011)
Poultry slaughterhouse wastewater	Mode: Batch Electrode: Fe	COD: 93 %	150	0.3 – 0.4	(Bayramoglu et al., 2006)
Coal mine drainage wastewater	Mode: Batch Electrode: Fe	Heavy metal: 28.7–99.96 %	200–500	1.09–2.184	(Oncel et al., 2013)
Textile wastewater	Mode: batch Electrode: Al (monopolar)	COD: 15–62 %	50–200	0.32–0.58 US$.kg^{-1} COD	(Bayramoglu et al., 2004)
Chromium removal	Mode: Batch Electrode: Fe (monopolar)	Chromium: 99.64 %	89.45	0.207	(Patel and Parikh, 2021)
Olive oil wastewater	Mode: Batch Electrode: Al	COD: 78.51 % Turbidity: 97.92 %	150	0.12	(Niazmand et al., 2019)
Hospital wastewater (EC+UF)	Mode: Continue Electrode: Al (monopolar + bipolar)	TSS: 95.12 % TDS: 97.53 % BOD: 95.18 % COD: 97.88 %	66	3.92 (5 m^3 wastewater.day^{-1}, including labor cost) 0.89 (50 m^3 wastewater.day^{-1}, including labor cost)	This Study
Hospital wastewater (EC+RO)	Mode: Continue Electrode: Al (monopolar + bipolar)	TSS: 97.64 % TDS: 99.85 % BOD: 97.88 % COD: 98.38 %	77	4.02 (5 m^3 wastewater. day^{-1}, including labor cost) 0.93	This Study

				(50 m^3 wastewater. day^{-1} =1, including labor cost)	

4 Conclusion

In this work, the electrocoagulation (EC) process with membranes was integrated with membranes to treat real hospital wastewater. Two types of membranes were used, namely ultrafiltration (UF) and reverse osmosis (RO). In EC systems, Al electrodes were used, which were arranged in monopolar-parallel and bipolar configurations. There are two parameters studied in the EC system: the configuration of electrodes and applied currents. Two types of membranes were used for treating the effluent of EC process, namely ultrafiltration (UF) and reverse osmosis (RO). The effluent qualities of the two configurations of wastewater systems (EC-UF and EC-RO) were investigated towards the total suspended solids (TSS), chemical oxygen demand (COD), biological oxygen demand (BOD), and total dissolved solids (TDS). It was found that the integration of EC-UF with a configuration electrode of 4 anodes 2 cathodes 2 bipolar (4A-2C-2B)(4A-2-C-2B) at a current density of 88.588.5 $A.m^{-2}$ =2 resulted in high removal of TSS, TDS, BOD, and COD by 95.12%, 97.53%, 95.18%, 95.12 %, 97.53 %, 95.18 %, and 97.88%, 97.88 %, respectively. The effluent quality was decreased by reducing the number of anodes from 4 to 2 pcs. The configuration of 2A-2C-2B2A-2-C-2B and current density of 110.6110.6 $A.m^{-2}$ =2 resulted in TSS, TDS, BOD, and COD removal of 90.63%, 94.89%, 88.46%, 90.63 %, 94.89 %, 88.46 %, and 90.63%, 90.63 %, respectively. The lower quality of EC-UF system could be improved by substituting UF with RO membrane. The TSS, TDS, BOD, and COD removal were enhanced to 97.64%, 99.85%, 97.88%, 97.64 %, 99.85 %, 97.88 %, and 98.38%, 98.38 %, respectively. The UF membrane was more severe to fouling compared to the RO membrane, which was attributed to the formation of cake layer fouling on the membrane surface. The permeate flux decline in UF membrane system was 47.83% 47.83 % during 6060 min of filtration time, while in the RO membrane system was 29.49. When the wastewater capacity was increased from 5 to 50 m50 $m^3.day^{-1}$ =1, the operating cost for the EC-UF system was decreased from 3.92 to 0.89 US$.$m^{-3}$ =3, while the EC-RO system was decreased from 4.02 to 0.93 US$.$m^{-3}$ =3. Since the EC-UF and EC-RO showed high efficiency of contaminants removal, these configurations could be used as alternative clean technology to produce clean water for water reuse purposes from hospital wastewater. Process optimization is further required to minimize the energy costs in the EC process, which has an impact on reducing operating costs.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Process Safety and Environmental Protection

High-Efficiency Contaminant Removal from Hospital Wastewater by Integrated Electrocoagulation-Membrane Process

--Manuscript Draft--

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COVER LETTER

Dear Editor,

We are pleased to submit a manuscript as a research article entitled "High-Efficiency Contaminant Removal in Hospital Wastewater by Electrocoagulation Process Integrated with Membranes" to be considered for publication in the Journal of Process Safety and Environmental Protection. It has not been published elsewhere and submitted simultaneously for publication elsewhere.

This paper focuses on real hospital wastewater treatment. The hospital wastewater contains an enormous diversity of chemicals, such as pharmaceuticals, radioactive elements, endocrine disruptors, detergents, heavy metals, zinc, and pathogenic microorganisms. Various technologies have been proposed as an alternative to the conventional wastewater processes, particularly to minimize the usage of chemicals, reduce the operating time, and be environmentally friendly. In this paper, hospital wastewater was treated by integration of electrocoagulation (EC) with ultrafiltration (EC) and reverse osmosis (RO) membranes. In the EC system, Al electrodes were arranged in a monopolar-parallel and bipolar configuration. There are two parameters studied in the EC system, i.e., the configuration of electrodes (2A-2C-2B and 4A-2C-2B) and applied currents (10 A and 12A). The EC process significantly reduced the suspended solids in wastewater but not on chemical and organic compounds. Since the EC process removed the suspended solids and most of the chemical or organic compounds, the application of membranes as a post-treatment became feasible. The effluent qualities of the EC-UF and EC-RO systems were analyzed in terms of total suspended solids (TSS), total dissolved solids (TDS), biological oxygen demand (BOD), and chemical oxygen demand (COD). The EC-UF system with a configuration of 4A-2C-2B 12A resulted in high removal of TSS, TDS, BOD, and COD by 95.12%, 97.53%, 95.18%, and 97.88%, respectively. The effluent quality of the EC-UF was improved by substituting UF with RO membrane. The TSS, TDS, BOD, and COD removal were enhanced to 97.64%, 99.85%, 97.88%, and 98.38%. Since the EC-UF and EC-RO showed high efficiency in contaminants removal, these configurations could be used as an alternative clean technology to produce clean water for water reuse purposes. Finally, the EC-UF and EC-RO operating costs were calculated at various wastewater capacities. The calculation of operating cost involved the annual cost of the energy required in EC system and pump units, electrodes, membrane replacement for UF and RO, chemicals for UF and RO membrane, and labor. At a wastewater capacity of 5 $m^3.h^{-1}$, the operating cost for the EC-UF system was 3.92 US$.$m^{-3,}$ while the EC-RO system was 4.02 US$.$m^{-3}$. The increase of wastewater capacity to 50 $m^3.day^{-1}$ reduced the operating cost to 0.89 US$.$m^{-3}$ for the EC-UF system and 0.93 US$.$m^{-3}$ for the EC-RO system.

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Thank You.
Best regards,

Putu Teta P. Aryanti
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High-Efficiency Contaminant Removal from Hospital Wastewater by Integrated Electrocoagulation-Membrane Process

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Abstract

In this work, high removal of contaminants in hospital wastewater has been achieved using an integration of electrocoagulation (EC) with ultrafiltration (UF) and reverse osmosis (RO). In EC system, Al electrodes were arranged in a monopolar-parallel and bipolar configuration. There are two parameters studied in the EC system, i.e., the configuration of electrodes (2A-2C-2B and 4A-2C-2B) and applied currents (10 A and 12A). The EC-UF system with a configuration of 4A-2C-2B 12A resulted in high removal of TSS, TDS, BOD, and COD by 95.12%, 97.53%, 95.18%, and 97.88%, respectively. The effluent quality of the EC-UF was improved by substituting UF with RO membrane. The TSS, TDS, BOD, and COD removal were enhanced to 97.64%, 99.85%, 97.88%, and 98.38%. The permeate flux decline in UF membrane system was 47.83% during 60 minutes of filtration time due to cake layer fouling on the membrane surface, while in the RO membrane system was 29.49%. Since the EC-UF and EC-RO showed high efficiency in contaminants removal, these configurations could be used as clean technology to produce clean water for water reuse purposes. At a wastewater capacity of 5 $m^3.h^{-1}$, the operating cost for the EC-UF system was 3.92 US$.$m^{-3,}$ while the EC-RO system was 4.02 US$.$m^{-3}$. The increase of wastewater capacity to 50 $m^3.day^{-1}$ reduced the operating cost to 0.89 US$.$m^{-3}$ for the EC-UF system and 0.93 US$.$m^{-3}$ for the EC-RO system.

Keywords: clean technology, electrocoagulation, hospital wastewater, ultrafiltration, reuse water

1. Introduction

Hospitals require a large quantity of clean water in their daily activities, particularly for medical laboratories, sanitation, gardening, kitchens, and laundries (Rani and Singh, 2021). Based on the number of beds, the daily quantity of water is estimated from 200 to 1200 L per bed of inpatients (Garcia-Sanz-Calcedo et al., 2017; Khan et al., 2021). Thus, a considerable effluent of wastewater should be treated before being discharged into the environment. The hospital wastewater contains an enormous diversity of chemicals, such as pharmaceuticals, radioactive elements, endocrine disruptors, detergents, heavy metals, zinc, and pathogenic microorganisms (Zhang et al., 2020). The pharmaceutical concentration in hospital wastewater was reported to be 10 – 100 times higher than the urban wastewater (Verlicchi and Zambello, 2016). Nowadays, hospital wastewater has gained serious attention due to the SARS-CoV-2 virus, which poses a danger to human health and the environment if it is not appropriately treated (Achak et al., 2021).

Various technologies have been developed as an alternative to the conventional wastewater processes, particularly to minimize the usage of chemicals, reduce the operating time, and be environmentally friendly. These technologies are advanced oxidation processes (AOPs), integrated pressure-driven membranes (ultrafiltration/UF and reverse osmosis/RO) (Ouarda et al., 2018; Siagian et al., 2021; Tiwari et al., 2021), and biological processes (activated sludge and membrane bioreactor/MBR) (Beier et al., 2012). AOPs is a technique to eliminate organic contaminants in wastewater by generating reactive oxygen species (ROS), such as hydroxyl (OH*), sulfate (SO_4^{*-}), or chlorine (Cl*) radicals to produce harmless contaminants (Al Mayyahi and Al-Asadi, 2018). The oxidation process is conducted through several methods, such as ozonation (Hussain et al., 2020), photocatalysis (Liu et al., 2020), UV photolysis (Chuang et al., 2017), Fenton (Liang et al., 2021), and wet air oxidation (Deshmukh and Manyar, 2021). Souza et al. (2018) found that the combination of ozonation and UV (O_3/UV) removed total organic compound (TOC) in hospital wastewater up to 54.7% after 120 minutes of oxidation time at a rate of 1.57 g $O_3.h^{-1}$. In addition, the chemical oxygen demand (COD) and aromatic reduction efficiency removal could reach 64.05% and 81%, respectively. Segura et al. (2021) reported that the Fenton oxidation provided a high pharmaceutical removal of 99.8%, greater than the catalytic wet air oxidation (CWAO) of 90%. Despite high removal efficiency, the Fenton oxidation process is limited by the low pH requirement, iron sludge generated, and high chemical consumption. Further research in oxidation process is continuously conducted to minimize sludge generation and chemical consumption.

Recently, more interest has been paid to the MBR process than conventional activation sludge (CAS) due to less space requirement and higher organic removal ability (Judd, 2016; Wenten et al., 2020). Ouarda et al. (2018) combined submerged MBR (s-MBR) and electrochemical oxidation (EO). The integration of MBR-EO removed pharmaceutical contaminants by 97% after 40 minutes of treatment at electrochemical oxidation's current density of 0.5 A. Furthermore, the MBR-EO eliminated venlafaxine (VEN) by 92%, higher than MBR or EO alone by 30% and 50%, respectively. High-energy consumption and complex membrane fouling become the challenges in the large-scale application of the MBR (Do and Chu, 2022). Nanofiltration and RO have also been widely used in water and wastewater treatment, including hospital wastewater. Both technologies provide excellent performance in removing contaminants and colors in the wastewater, either standing alone or combined with other processes (Lan et al., 2018; Patel et al., 2022; Rochmah and Widiasa, 2021; Tran et al., 2019). However, high water flux decline due to fouling phenomena becomes a challenge in membrane application for wastewater treatment. The fouling formation leads to higher energy consumption and chemicals for membrane cleaning, which contributes to the increase in operational costs (Zhang et al., 2019). Ultrafiltration (UF) membrane, which is viewed as an energy-efficient technology in water and wastewater treatment, is more susceptible to irreversible fouling due to its porous structure (Karimi et al., 2020). Therefore, pretreatment processes are needed prior to the membrane processes to minimize fouling formation and maintain the productivity of the membrane.

Several technologies, such as coagulation (Alibeigi-Beni et al., 2021), adsorption (Kim et al., 2022), ozonation (Zhao et al., 2019), and electrocoagulation (EC) (Tavangar et al., 2019), have been used as a pretreatment of membrane processes. Among these technologies, EC has increasingly been used as an alternative green process instead of chemical coagulation (Hashim et al., 2019). The EC process offers some advantages, such as less sludge generated, easy operation, short operation periods, and being environmentally friendly (Shahedi et al., 2020). The EC process removes various pollutants, colloidal, and organic matters efficiently, which may minimize the irreversible fouling and prolong the life of the membrane (Lu, Jincheng et al., 2021). The fundamental of EC process lies on the electrolysis reaction, which requires a direct electric current to generate chemical reactions at the surface of electrodes, namely anodes and cathodes (Asaithambi et al., 2021). Aluminum (Al) and iron (Fe) electrodes are the two commonly used electrodes in EC processes due to their availability, low cost, and form amorphous metal oxides or oxyhydroxides or hydroxides matters that offer high adsorption ability to soluble contaminants (Tegladza et al., 2021).

When the electric current is applied to the electrodes, the metal ions (Fe^{3+} or Al^{3+}) are released and dissolved from the sacrificial anodes to the wastewater in EC reactor. At the same time, the water molecules are dissociated into H^+ and OH^- at cathodes by electrochemical reaction. There are various reactions occur simultaneously in the EC reactor. The metal ions released from the anode destabilize the colloid by charge neutralizing, allowing small aggregates to form. The other metal ions react with OH- ions and undergo spontaneous hydrolysis reactions to form amorphous precipitates, such as hydroxides ($Al(OH)_3$ or $Fe(OH)_3$), oxides (such as AlO_3 or α-FeO_3), and oxyhydroxides (such as γ-AlOOH and α-FeOOH) (Tegladza et al., 2021). The amorphous compounds bind the aggregates to form larger flocs and precipitate to the bottom of the EC reactor. Oxygen evolution reaction (EOR) may occur in the anodes, simultaneously with the dissolution of metal ions.

Meanwhile, hydrogen (H_2) gas bubbles are generated simultaneously at the cathode during the electrolysis reaction, which floats the light contaminants to the top of the reactor. The flocs produced by EC process are more stable, relatively large, and slightly bound with water (Mureth et al., 2021). Thus, it can be easily separated by the filtration method. The main reactions during EC can be summarized by the following equations (Nugroho et al., 2019):

Anode reactions:

$$M_{(s)} \rightarrow M_{(aq)}^{n+} + ne^- \quad (1)$$

$$2H_2O_{(l)} \rightarrow 4H^+_{(aq)} + O_{2(g)} + 4e^- \quad (2)$$

Cathode reactions:

$$Mn_{(aq)}^{n+} + ne^- \rightarrow M_{(s)} \quad (3)$$

$$2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH^- \quad (4)$$

A few studies have been conducted by using EC process for hospital wastewater treatment. Dehghani et al. (2014) studied the influence of Fe and Al electrodes in batch EC on the removal efficiency of chemical oxygen demand (COD). The removal efficiency was up to 87% using two pairs of Fe-Fe electrodes at a reaction time of 60 min, pH 3, and voltage of 30 V. By changing the configuration to two pairs of Fe-Al electrodes, the COD removal efficiency was reduced to 75%. Furthermore, the increase of electrode distance from 2 to 3 cm reduced the removal efficiency from 87 to 68%. Esfandyari et al. (2019) found that the higher COD and turbidity removal efficiency was achieved in neutral conditions (pH 7) compared to acid (pH 4) and alkaline (pH 9) conditions. Using 3 iron pairs as electrodes, the antibiotic (cefazolin), COD, and turbidity removal at a voltage of 15 V and reacting time of 30 min were 91.92%, 87%, and 92,16%, respectively. The increase of voltage improved the removal efficiency. Other studies showed that the EC process effectively removed other contaminants in hospital wastewater, such as dexamethasone (Arsand et al., 2013) and ciprofloxacin (Ahmadzadeh et al., 2017; Yoosefian et al., 2017).

Besides the type of electrodes, electrode configuration has also played an important role in EC process (Lu, Jianbo et al., 2021). The electrodes can be assembled in monopolar (either in series (MP-S) or parallel (MP-P)) and bipolar (BP). The monopolar and bipolar electrode configurations have been described in various literature (Akter et al., 2022; Al-Raad and Hanafiah, 2021; Almukdad et al., 2021). The mode of electrode connection contributes to the efficiency of contaminants removal and energy consumption (Xolov, 2021). Several studies have focused on using BP electrodes due to their simplicity, easy maintenance, less electrical connection to the electrodes, and less maintenance cost (Nippatlapalli and Philip, 2020; Qi et al., 2020; Tchamango and Darchen, 2018). In BP electrodes, the two outer electrodes are connected to the external power supply. Meanwhile, the internal sacrificial electrode, which is called BP electrodes, are not interconnected and each side of the electrodes performs simultaneously as an anode and a cathode. Compared to the monopolar configuration, bipolar electrodes showed higher removal efficiency but required higher energy consumption for the same effluent quality (Golder et al., 2007; Khaled et al., 2019). The higher energy consumption can be attributed to the longer distance between the electrodes connected to an external electric current source, resulting in higher resistance to mass transfer and lower kinetics of charge transfer (Khaled et al., 2019). Therefore, in this research, a combination of monopolar and bipolar electrodes was used to produce high removal efficiency of EC process and narrow the distance between the electrodes connected to the current source. The EC process was equipped with a low rate agitation process to enhance the reaction rate in EC reactor during the real hospital wastewater treatment. In addition, the EC process was integrated with polypropylene (PP) cartridge filter, ultrafiltration (UF), and reverse osmosis (RO) membranes to improve the effluent quality. The performances of EC-UF and EC-RO units were compared, both the effluent quality and the large-scale economic evaluation.

2. Material and Method

2.1. Materials and Experimental Set-Up

The wastewater was collected from one of the hospitals in Cimahi, West Java, Indonesia, without any pretreatment. The initial condition of the wastewater is shown in **Table 1**.

Table 1. Initial condition of the hospital wastewater

Parameter	Unit	Value
TDS	$mg.L^{-1}$	974 – 995
TSS	$mg.L^{-1}$	328 – 381
BOD	$mg.L^{-1}$	520 – 801
COD	$mg.L^{-1}$	176 - 281
pH	-	7-8

The EC system used in this research refers to the previous work (Nugroho et al., 2019; Nugroho et al., 2021). The cylindrical EC reactor had 26 cm in diameter and was occupied by a six-blade turbine with a constant agitation rate of 70 rpm. The Aluminum electrodes were used as baffles, which were arranged in 4 configurations involving monopolar and bipolar electrodes. The dimension of each electrode was 33 cm (height) x 4 cm (width) x 3 mm (thickness). The wastewater flow rate to the EC reactor was set at 125 mL/min with a residence time of 1 hour (60 minutes). A 20-inch polypropylene (PP) cartridge filter (5 µm), filled with 300 grams of manganese sands in the tube side, was placed in a 20-inch blue housing.

The UF membrane was polysulfone-based hollow fiber and supplied by GDP Filter Indonesia. The UF membrane had an outside/inside diameter of 2.2/1.8 mm. A bundle of hollow fiber membranes was assembled in a 2 in (0.0508 m) diameter of PVC pipe with a module length of 30 cm (0.3 m). The total effective area of the UF membrane was 1.13 m^2/module. Meanwhile, the spiral-wound RO (RE-2012)

was provided by CSM Membrane. The RO membrane had a total membrane area of 1.95 m^2. The schematic of EC and membrane systems is shown in **Figure 1**.

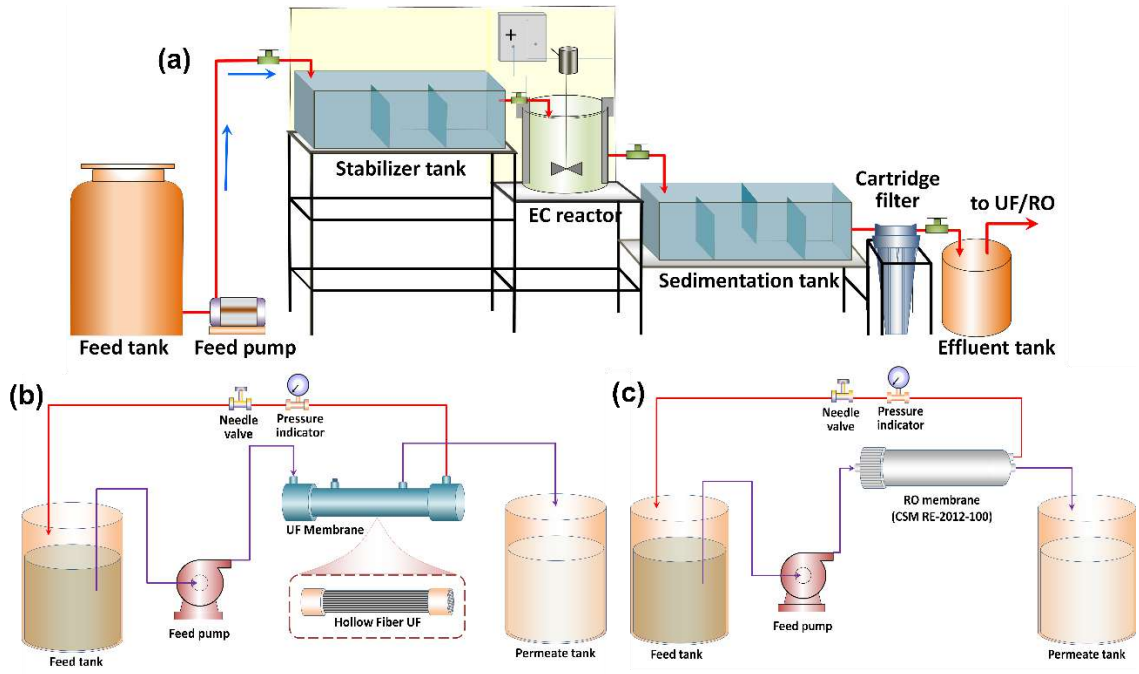


Figure 1. Schematic of experimental apparatus for (a) electrocoagulation process, (b) ultrafiltration process, and (c) reverse osmosis process

2.2. Hospital Wastewater Treatment by Electrocoagulation (EC) Process

The EC process treated 12.9 L of wastewater per hour with 4 electrode configurations involving monopolar and bipolar configurations. The electrode configurations are 2 anodes 2 cathodes 2 bipolar (2A-2C-2B) and 4 anodes 2 cathodes 2 bipolar (4A-2C-2B), which were operated at two different applied currents (10 and 12 A). The effluent of EC process was delivered to a cartridge filter filled with manganese sand to remove the remaining particulate matter and then analyzed in terms of COD, BOD, TDS, and TSS. In laboratory experiments, the consumption of electrodes was estimated from the weight loss of the electrodes, which was determined from the difference in weight before and after the EC process. Meanwhile, the H_2 gas production during the EC process was estimated by the following Equation:

$$Q_{H2} = {}^{I.A.t}/_{F.V} \quad (5)$$

where Q_{H2} is the number of H_2 gas produced by the cathode (in mole.L^{-1}), I is the applied current (in A.cm^{-2}), A is the effective surface area of cathodes that are contacted with the liquid (in cm^2), t is electrolysis time (in second), and V is the treated wastewater volume (in m^3).

On the other hand, the energy consumption in EC process was by the following Equation (Hashim et al., 2019; Niazmand et al., 2019):

$$C_{power\ EC} = {}^{U.I.t}/_{V} \quad (6)$$

where $C_{power\ EC}$ is the power consumption (in W.h.m^{-3}), I is applied current (in A), U is the potential of cell (in Volt), t is the operating time (in hour), and V is the volume of the wastewater treated (in m^3).

2.3. EC effluent treatment by ultrafiltration (UF) and reverse osmosis (RO)

The UF membrane was operated in crossflow mode at a constant pressure of 1 bar, while the RO membrane was performed at a constant pressure of 4 bar. The profile of permeate flux was observed during 1 hour of UF and RO process by the following Equation (Aryanti et al., 2021):

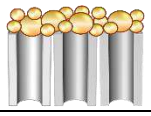
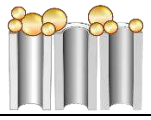
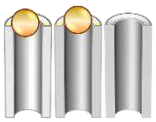
$$J = {}^{V_p}/_{A\,.\,t} \quad (7)$$

where J is the permeate flux (in L.m^{-2}.h^{-1}), V_p is the permeate volume (in L), A is the effective UF or RO membrane area per module (in m^2), and t is the operating time (in h). The UF and RO membranes were used to treat the hospital wastewater for 1 hour (60 minutes). The permeate flux of the membrane was measured every 20 minutes, which was denoted as J_p, and then calculated using Equation (4). The effluent was analyzed in terms of COD, BOD, TDS, and TSS. Fouling in the membrane structure was investigated by observing the change in permeate flux during 1 hour of UF, which was generally expressed as normalized flux (J/J_o). The RO process was performed under the same operating condition as EC-UF, which produced the lowest quality. The operating cost of the EC-UF system with the highest quality and the EC- RO system will be compared.

2.4. Fouling analysis during filtration of EC effluent by UF and RO

Hermia's models were used to analyze the dominant fouling in UF and RO membranes (Ariono et al., 2018; Moreira et al., 2021; Wenten et al., 2019). The fouling mechanism in UF and RO membrane systems is divided into 4 types: cake layer, intermediate blocking, standard blocking, and complete blocking, as shown in **Table 2**. Intermediate blocking occurs when the particles are partially blocking the membrane pore. The particles are probably deposited on the unobstructed area of the membrane surface or onto the previously deposited particles. Standard pore blocking is a fouling mechanism when the particles are adsorbed inside the membrane pore and reduce the effective diameter of the membrane pore. The last fouling mechanism is complete bocking, where the particles completely block the membrane pore. In cake fouling, the particles or contaminants are entirely cover the membrane surface. This model provides a correlation between permeate flux and operating time. A simple plot in a J vs. t was used to define the most dominant fouling in the membrane system.

Table 2. Linear equations for fouling analysis by Hermia's model.

Code	Fouling mechanisms	Figure	Linear equations	Equation no.
Model 1	Cake layer formation (code: c_f)		$\frac{1}{J^2} = \frac{1}{J_o^2} + K_{cf}t$	(8)
Model 2	Intermediate blocking (code: ib)		$\frac{1}{J} = \frac{1}{J_o} + K_{ib}A\,t$	(9)
Model 3	Standard blocking (code: sb)		$\frac{1}{J^{1/2}} = \frac{1}{J^{1/2}} + K_{sb}t$	(10)
Model 4	Complete blocking (code: cb)		$\ln(J) = \ln(J_o) + K_{cb}t$	(11)
Notes: Jo is initial flux at t = 0 min.(L.m^{-2}.h^{-1}), A is membrane area (m^2), t is operating time (h), and K is the fouling parameter of each fouling mechanism.				

2.5. Estimation of operating cost for the EC-UF and EC-RO Process

The operating cost estimation of EC process was based on the power consumed in the electrolysis process, power for the feed pump, and electrode consumption (Kumari and Kumar, 2021), as follows:

$$Operating\ cost\ of\ EC = \alpha\, C_{power\ EC} + \beta\, W_e + \gamma\, C_{power\ pump} \quad (12)$$

where W_e and $C_{power\ EC}$ were calculated by Equations (1) and (2). The α and β were the unit price of electrical (in US$/kWh) and electrode (in US$/kg), respectively. The pump power ($C_{power\ pump}$) was calculated using Equation (13) (Aryanti et al., 2020).

$$C_{power\ pump} = P_f\ x\ F \quad (13)$$

where P_f is the feed pump pressure (in bar) and F is the feed flow rate (in $m^3.h^{-1}$). The efficiency of the feed pump was assumed to be 75%. The operating pressure of UF and RO membranes were estimated by considering the electricity cost of feed pump, chemicals for membrane cleaning (examples: NaOH 0.1% and Citric acid 2%), and membrane replacement (Aryanti et al., 2020). The recovery design of the UF membrane was 90%, while the RO membrane was 60%. The number of membrane modules was determined based on the permeate flux data. The membrane working live was assumed 3 years. The plant design and basic cost parameters for estimating the EC, UF, and RO membrane operating cost are detailed in **Table 3**.

Table 3. The design and cost parameters of the EC-UF and EC-RO plants.

Parameters	Units	Values	Ref.
A. Design parameters			
- Plant capacity	$m^3.h^{-1}$	5 - 50	-
- Working hour per day	h	20	-
- UF membrane design			-
- Permeate flux design per module	$L.m^{-2}.h^{-1}$	50	-
- Recovery design	%	90	-
- Effective membrane area per module	m^2	10	-
- Operating pressure of pump (feed and backwash)	bar (kPa)	1.5 (150)	-
- Efficiency of pump	%	75	-
- RO membrane design			
- Permeate flow rate per module	$m^{-3}.h^{-1}$	0.25	Engineering est.
- Recovery design	%	50	-
- Effective membrane area per element (length x diameter)	m^2	7.9 (40 x 4 in)	(Lenntech, 2022)
- Operating pressure of RO pump	bar (kPa)	7 (700)	-
- Operating pressure of CIP	Bar (kPa)	1.5 (150)	-
- Membrane lifetime	year	2	Engineering est.
B. Operating cost parameters			
- Al electrode	$US\$.kg^{-1}$	1.8	(Keyikoglu and Can, 2021)
- UF Membrane cost	$US\$.m^{-2}$	20	Engineering est.
- RO Membrane cost	$US\$.m^{-2}$	40	(Choi et al., 2015)
- Electricity	$US\$.kWh^{-1}$	0.19	(da Silva et al., 2020)

- Labor cost	US$.(man-month^{-1})	250	(Meratizamana and Asadib, 2020)
- Number of labors	person	2	Engineering est.
- Chemical cost of UF membrane	US$.m^{-3} permeate	0.01	(Bhojwani et al., 2019)
- Chemical cost of RO membrane	US$.m^{-3} permeate	0.033	(Gökçek and Gökçek, 2016)

3. Results and Discussions

3.1. The number of hydrogen (H_2) gas production at the cathode and metal ions released from the anode

The applied current density influences the H2 gas production and metal ion released from the electrodes during 1 hour of EC process. **Figure 2** shows the influence of electrode configuration and applied current on the theoretical number of Al^{3+} ions released from the anode. The increase of applied current from 10 to 12 A with an electrode configuration of 2A-2C-2B enhanced the number of ions released by 48.57% (from 255.66 to 410.77 mg/L). While in the 4A-2C-2B configuration, the number of ions released was enhanced by 39.75% (from 302.33 to 422.48 mg/L). The increase of applied current improved the potential electrolysis (Galvão et al., 2020; Kamar et al., 2018), which contributed to more release of Al^{3+} ions. The number of ions was further raised by increasing the number of anodes in the EC reactor. On the other side, H_2 gas bubbles are simultaneously generated near the cathode's surface due to water electrolysis phenomena. The amount of H_2 gas formed during the EC process is presented in **Table 4,** which was calculated by Equation (5). Two parameters affected the H_2 production, i.e., the number of cathodes and applied currents. The increase of applied current from 10 to 12 A at the same number of cathodes enriched the H_2 production by 20% (from 168.65 to 202.38 mol.L^{-1}). The increase of applied current enhanced the water splitting rate near the cathode surface, and consequently, the number of H_2 gas generated was raised.

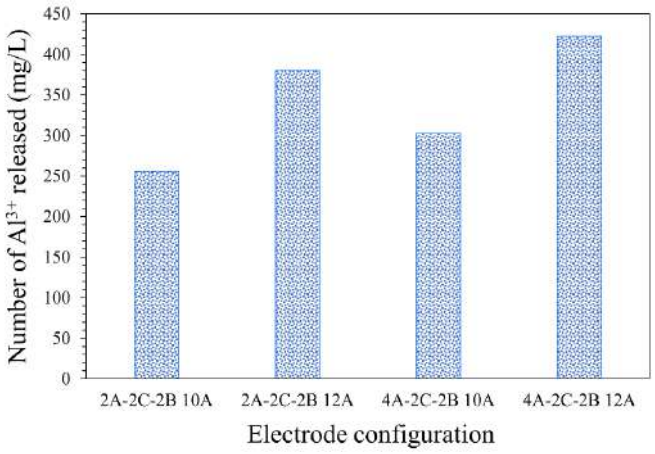


Figure 2. The number of Al^{3+} ions that are released from the anode.

Table 4. H_2 gas production (mol.L^{-1})

Configuration of electrodes	QH_2 (mole.L^{-1})	QH_2 (kg.L^{-1})
2A-2C-2B 10 A	168.65	0.34

2A-2C-2B 12 A	202.38	0.41
4A-2C-2B 10 A	168.65	0.34
4A-2C-2B 12 A	202.38	0.41

3.2. The influence of electrode configuration and applied current on EC-cartridge/sand filter-UF and EC-cartridge/sand filter-RO effluent qualities

Figure 3 shows the influence of electrode configuration and applied current on the effluent qualities of the EC process. In this research, most of the EC process provided high removal efficiency of contaminants removal, in terms of TDS, TSS, BOD, and COD, which was over 90%. The lowest removal efficiency was achieved by electrode configuration of 2A-2C-2B at an applied current of 10 A. The TDS, TSS, BOD, and COD removals were 90.63%, 87,40%, 88.46%, and 94.89%, respectively. When the anode number was increased from 2 to 4 pieces (4A-2C-2B 10A), the removal of the contaminants was slightly improved to 93.63% for TDS, 89.84% for TSS, 90.37% for BOD, and 96.15% for COD. The change of applied current from 10 A to 12 A at a fixed electrode configuration of 2A-2C-2B raised the removal efficiency of the contaminants to 95.58% for TDS, 91.73% for TSS, 92.20% for BOD, and 97.45% for COD. These results implied that the change in applied current gave a more significant effect to the contaminant removal compared to the addition of electrodes in the EC reactor. The highest removal efficiency was resulted by using 4A-2C-2B with a current density of 12A. The TDS, TSS, BOD, and COD were 97.89% (from 996.6 to 24.56 mg/L), 95.12% (from 328.0 to 16.0 mg/L), 95.18% (from 606.4 to 29.2 mg/L), and 97.88% (from 280.6 to 5.94 mg/L), respectively.

The contaminants removal in EC process includes a complex mechanism. It has been explained in sub-chapter 3.1 that the increase of applied current enhanced the electrolysis reaction rate during the EC process. As a result, the production of Al^{3+} ions from the anodes and H_2 gas bubbles from the cathodes was raised. The Al^{3+} ions were released from the anode and dissolved into the bulk solution. A part of the ions destabilized the counter ionic species in the wastewater and led to the formation of flocs as a result of coagulation. The other Al^{3+} ions were transformed into amorphous hydroxide flocs, $Al(OH)_3$, which bound the suspended solids and organic matter in the wastewater to form larger flocs, and then settled to the bottom of the EC reactor. The increase of Al^{3+} ions in the EC process brought more contaminants removal. It has been reported in the literature that there is a competition between Al^{3+} ion dissolution and oxygen reaction evolution (EOR) during the EC process at a high applied current (Tegladza et al., 2021). The presence of EOR reaction may reduce the efficiency of Al^{3+} dissolution. In this research, the removal of the contaminant was increased when the applied current was increased from 10 to 12 A. It was suggested that the Al^{3+} dissolution was still giving a dominant role in agglomerating the contaminants. The addition of Al^{3+} number from the bipolar electrodes could enhance the number of coagulants formed in the wastewater solution. In addition, the agitation process in the EC reactor improved the distribution and mass transfer of Al^{3+} ions in the solution to destabilize and form amorphous $Al(OH)_3$. The homogenous distribution of coagulants enhanced the aggregation of the contaminants and contributed to the acceleration of coagulation. The O_2 gas oxidized organic pollutants in wastewater, improving separation efficiency.

In cathodes, the formation of H_2 gas bubbles was also influenced by the applied current, as shown in **Table 4**. The rise of the applied current increased the water electrolysis rate, which increased the bubble density in the EC reactor. It has been reported that the applied current also affects the bubble size of H_2 gas (Barrera-Díaz et al., 2018). The increase of applied current reduced the H_2 bubble gas size, which facilitates high contaminants removal by H_2 gas flotation. The flotation efficiency decreases with the rise of bubble size due to less surface area and retention time of the bubbles in the solution. The bubble size distribution was not measured in the present study, but it was observed that more air bubbles are formed as the applied current increases.

The integration of EC-RO was also used to treat the hospital wastewater at an electrode configuration of 2A-2C-2B and an applied current of 10 A, where the UF membrane could not remove the contaminants excellently. By using RO as post-treatment instead of UF membrane, the removal efficiency of contaminants was significantly improved to 99.89% for TDS, 97.64% for TSS, 97.88% for BOD, and 98.38% for COD. The RO membrane (CSM RE-2012) had a dense layer, and consequently, high rejection of contaminants can be achieved. The performances of UF and RO membranes during the treatment of effluent from the EC process will be discussed in the next sub-chapter, including the economic evaluation for both EC-UF and EC-RO configurations. The photos of EC-UF and EC-RO effluents are shown in **Figure 4,** where the effluent quality seems clear. Based on the quality of the effluent produced, it can be used for sanitation and agricultural needs. The comparison of performances of several technologies for hospital wastewater treatment compared to this study is presented in **Table 5**.

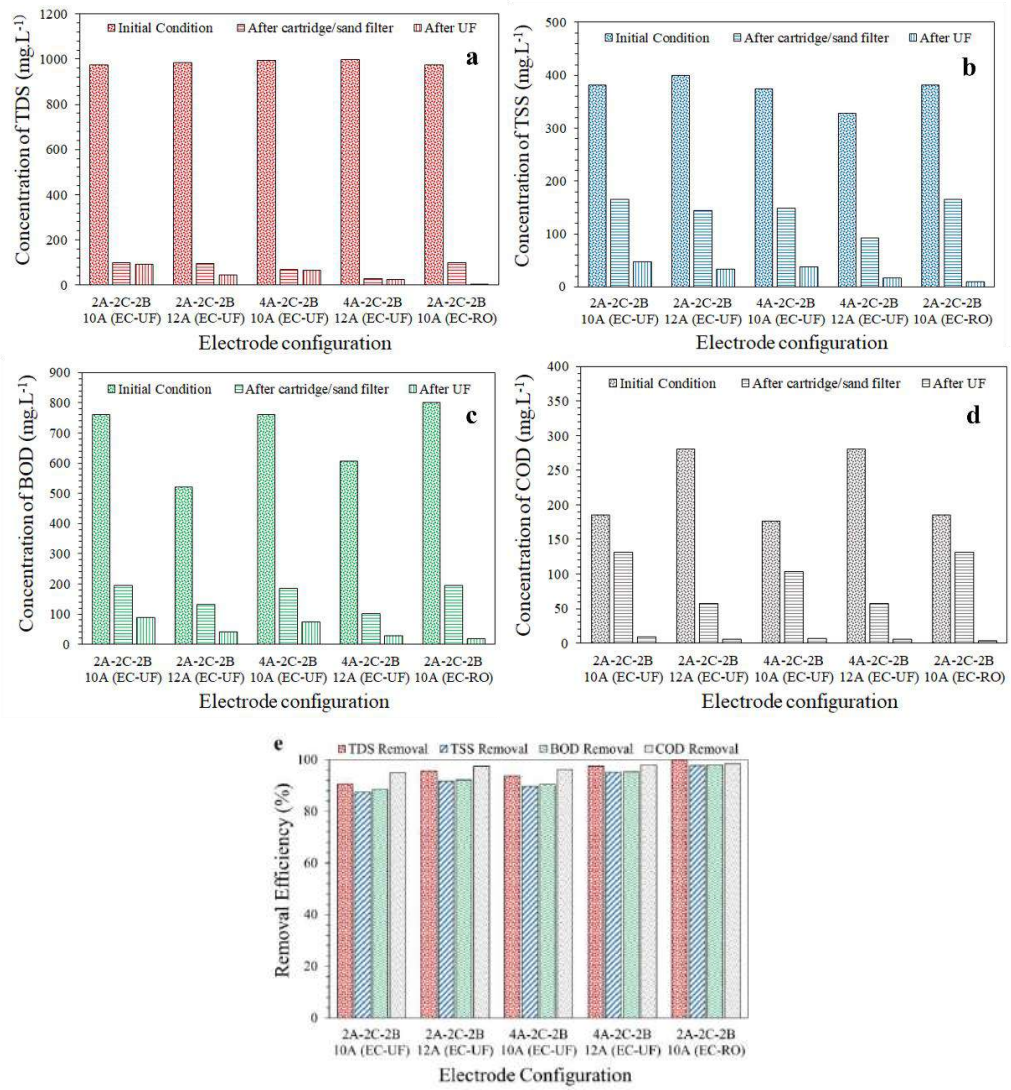


Figure 3. The concentration of contaminants before and after wastewater treatment using EC-UF and EC-RO, in terms of (a) TDS, (b) TSS, (c) BOD, (c) COD, and (d) the removal efficiency

(a) (b) (c) (d)

Figure 4. Photos of (a) initial condition of hospital wastewater, (b) effluent of EC-UF 2A-2C-2B 12A, (c) effluent of EC-UF 4A-2C-2B 12A, and (d) effluent of EC-RO 2A-2C-2B 10A

Table 5. The performances of several processes for hospital wastewater treatment

Process configuration	Operating condition	Removal efficiency (%)	Ref.
CAS bioreactor/ ultrasound	MLSS = 3000 – 8000 mg.L-1 HRT = 2-8 hours Initial condition of feed: COD: 480 – 520 $mg.L^{-1}$ TSS: 40 – 50 $mg.L^{-1}$ pH: 7.1 – 7.4	COD: 92% Effluent turbidity: 2.7 $mg.L^{-1}$	(Karami et al., 2018)
Sponge-SMBR-ozonation	Membrane polymer: Membrane type: hollow fiber Membrane pore size: 0.4 µm Flux: 10-20 $L.m^{-2}.h^{-1}$ SRT : 20 days Type of sponge: polyethylene Initial condition of feed: COD: 320 $mg.L^{-1}$, NOR: 16 $mg.L^{-1}$, CIP: 7.28 $mg.L^{-1}$, OFL: 21.51 $mg.L^{-1}$, SUL: 1.46 $mg.L^{-1}$	COD: >90% NOR: 92% CIP: 83% OFL: 88% SUL: 66%	(Vo et al., 2019)
SMBR	Membrane polymer: PVC/ZnO Permeate flux: 122.22 $L,m^{-2}.h^{-1}$ Membrane pore size: 75-450 nm MLSS: 10.000 mg/L SRT: 25 days	COD removal: 73.5%	(Alsalhy et al., 2018)
Moving bed biofilm reactor (MBBR)	HRT: 24 hours MLSS: 3000 $mg.L^{-1}$ Initial feed conditions: COD: 750 – 850 $mg.L^{-1}$	BOD: 97.8% COD: 95.6%	(Shokoohi et al., 2017)

	BOD: 400 – 500 mg.L^{-1} pH: 7.2 – 8.5 TSS: 300 – 400 mg.L^{-1}		
Supercritical water oxidation (SCWO)	Temp. in reactor: 450 °C. Pressure in reactor: 25 MPa Reaction time: 60 seconds Oxidant: H_2O_2 Initial condition of feed COD: 340 – 1200 mg.L-1 BOD5: 300 – 570 mg.L-1 TOC: 64.9 – 260.3 mg.L-1	COD, BOD, TOC, TN, and SS: >90%	(Top et al., 2020)
Coagulation. UV/H_2O_2, and activated sludge	Initial Feed condition PhACs: 0.108 mg.L^{-1}	PhACs: 83%	(Mir-Tutusaus et al., 2021)
Coagulation. UV/H_2O_2, and fungal treatment	Initial Feed condition PhACs: 0.108 mg.L^{-1} COD: 174 mg.L^{-1} TSS: 108 mg.L^{-1}	PhACs: 94% COD: 87 mg.L^{-1} TSS: 16 mg.L^{-1}	(Mir-Tutusaus et al., 2021)
Catalytic wet air oxidation	Catalyst: Pt supported multi-walled carbon nanotubes (Pt/CNT) Initial feed conditions: COD: 332 – 650 mg.L^{-1} TSS: 126 – 733 mg.L^{-1} pH: 6.8 – 8.7 mg.L^{-1}		(Segura et al., 2021)
Homogenous Fenton	Catalyst: dissolved $Fe(NO_3)_3$ 98% Fe^{3+} concentration: 25 mg.L^{-1}	Removal efficiency: COD removal: 70%	(Segura et al., 2021)

	H2O2 concentration: 2.123g.g^{-1} COD Operating temperature: 70 ^{o}C Reaction time: 240 minutes Initial feed conditions: COD: 332 – 650 mg.L^{-1} TSS: 126 – 733 mg.L^{-1} pH: 6.8 – 8.7 mg.L^{-1}	TOC: 50% PhACs (130oC): 90%	
Heterogeneous Photo-Fenton	Catalyst: Fe-BTC (Basolite F300-like semi-amorphous) H_2O_2 concentration: 1.125g.g^{-1} COD Reaction time: 120 minutes Initial feed conditions: COD: 332 – 650 mg.L^{-1} TSS: 126 – 733 mg.L^{-1} pH: 6.8 – 8.7 mg.L^{-1}	COD: 94.5% PhACs: 90%	(Segura et al., 2021)
EC	Electrodes: Fe-Al Retention time: 15 minutes Potential: 40 V Initial condition: COD: 502.8 mg.L^{-1} BOD_5: 136.4 mg.L^{-1} Phenols: 2.8 mg.L^{-1} TSS: 158.6 mg.L^{-1}	COD: 75.5% BOD_5: 59.2% Phenols: 80.7% TSS: 75.6%	(Yánes et al., 2021)
EC	Electrode: Fe Electrolysis time: 41 minutes Applied current: 2.64 A Effective area of electrode: 50 cm^2 Initial condition:	COD: 54.98% Chloride: 66.79%	(Bajpai and Katoch, 2020)

	COD: 377.5 mg.L^{-1} Chloride: 11.5 mg.L^{-1} pH: 7.41		
EC-UF	Electrode: Al (combination of monopolar and bipolar) Electrolysis time: 60 minutes EC-UF process: Electrode configuration: 4A-2C-2B Applied current: 12 A Operating pressure of UF: 1 bar	TSS: 95.12% TDS: 97.53% BOD: 95.18% COD: 97.88%	This study
EC-RO	EC-RO process: Electrode configuration: 2A-2C-2B Applied current: 10 A Operating pressure of RO: 4 bar Initial conditions: TDS: 974-995 mg.L^{-1} TSS: 328 – 381 mg.L^{-1} BOD: 520 – 801 mg.L^{-1} COD: 176 – 281 mg.L^{-1}	TSS: 97.64% TDS: 99.85% BOD: 97.88% COD: 98.38%	

Note: CAS = conventional activated sludge, MLSS = mixed liquor suspended solids, HRT = hydraulic retention time, COD = chemical oxygen demand, SMBR= submerged membrane bioreactor, NOR= norfloxacin, CIP = ciprofloxacin, OFL = Ofloxacin, SUL = sulfamethoxazole, PVC = polyvinyl chloride, ZnO = zinc oxide, SRT = solid retention time, BOD = biological oxygen demand, TSS = total suspended solids, TN = total nitrogen, TOC = total organic compounds, PhACs = pharmaceutically active compounds, EC = electrocoagulation, UF = ultrafiltration, RO = reverse osmosis

3.3. Profile of permeate flux and fouling analysis during filtration of EC effluent

Figure 5 presents the profile of permeate flux during 60 minutes of filtration process by UF and RO membranes. Prior to the membrane, the effluent of EC was filtered by a cartridge filter to remove large particulates and reduce membrane fouling. The permeate flux was significantly reduced up to 20 minutes of filtration by UF membrane, either effluent of EC with a configuration of 4A-2C-2B 10A and 4A-2C-2B 12 A. The flux reduction was attributed to the accumulation of contaminants on the membrane structure, which is known as fouling phenomenon. 4A-2C-2B 12A (EC-UF) configuration provided higher permeate flux than 4A-2C-2B 10A (EC-UF). It has been explained in section 3.2 that the configuration of 4A-2C-2B 12A (EC-UF) offered higher contaminant removal. Consequently, fouling resistance on or in the UF membrane structure could be minimized. After 60 minutes of filtration process, the configuration of 4A-2C-2B 12A (EC-UF) resulted in lower flux decline of 47.83% (from 53.10 to 27.70 $L.m^{-2}.h^{-1}$) compared to 51.24% (from 53.10 to 25.89 $L.m^{-2}.h^{-1}$) for the 4A-2C-2B 10A (EC-UF). Meanwhile, the RO permeate flux was seemed stable up to 60 minutes of the filtration process. At an operating pressure of 4 bar, a low permeate flux was obtained at the beginning of operating time, which was equal to 4.76 $L.m^{-2}.h^{-1}$. The flux reduction was 29.49% after 60 minutes of filtration.

The permeate flux decline was evaluated through normalized flux (J/Jo). The difference profile of permeate flux decline between UF and RO membrane was attributed to the pore structure and type of the membrane module. The feed channel spacer inside the spiral-wound membrane RO generated local mixing flow near the membrane surface, which enhanced the mass transfer and minimized fouling on the membrane surface (Lin et al., 2020). In addition, the non-porous structure of RO could minimize the irreversible fouling formation inside the membrane structure during 60 minutes of filtration. In long-term application, fouling inside the RO membrane could be formed and entrapped inside the spiral-wound system (Matin et al., 2021). Therefore, sufficient pretreatment systems are required to ensure the provision of good quality RO feedwater resulting in a stable performance for long-term application. While in hollow fiber UF membrane, a significant flux decline was found in the first 20 minutes of filtration. The flux decline was attributed to the rapid accumulation of mainly organic and soluble contaminants onto the UF membrane surface. Drag permeation from feed side to the permeate side could accelerate the adsorption of contaminants to the membrane surface. Contaminants that are smaller than the membrane pores entered the membrane pores, which contributed to irreversible fouling. In addition, the entrapped contaminants between the fibers might deteriorate the flux decline in the membrane system. Periodically cleaning of the membrane, such as backwash or air scouring, is recommended to maintain membrane performances.

Hermia's model was used to predict the dominant fouling mechanisms that occurred in the membrane system. As shown in Table 1, there are 4 types of fouling mechanisms, namely cake fouling layer (model 1), intermediate blocking (model 2), standard blocking (model 3), and complete blocking (model 4) according to Hermia's model. **Figure 6** shows the fouling mechanisms during ultrafiltration of EC effluent with a configuration of 4A-2C-2B at an applied current of 10A and 12A. In both configurations, cake layer fouling (model 1) became the dominant fouling mechanism during 60 minutes of the filtration process. It has been explained that the rapid accumulation of contaminants on the membrane surface could cover the membrane surface, particularly in the first 20 minutes of filtration time. The cake layer formation reduced the permeate flux significantly. While in the RO membrane system, the 4 types of fouling might occur simultaneously, which was indicated by the high value of R^2 in the 4 fouling models (**Figure 7**).

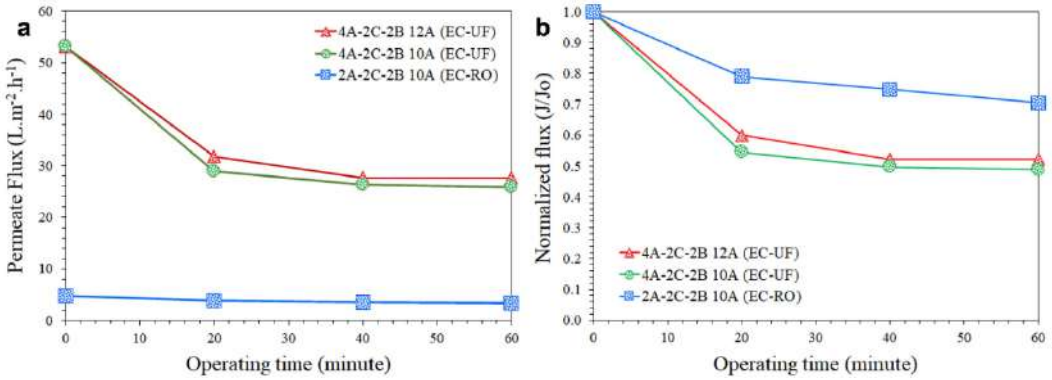


Figure 5. (a) Profile of permeate flux of membrane systems and (b) normalized permeate flux

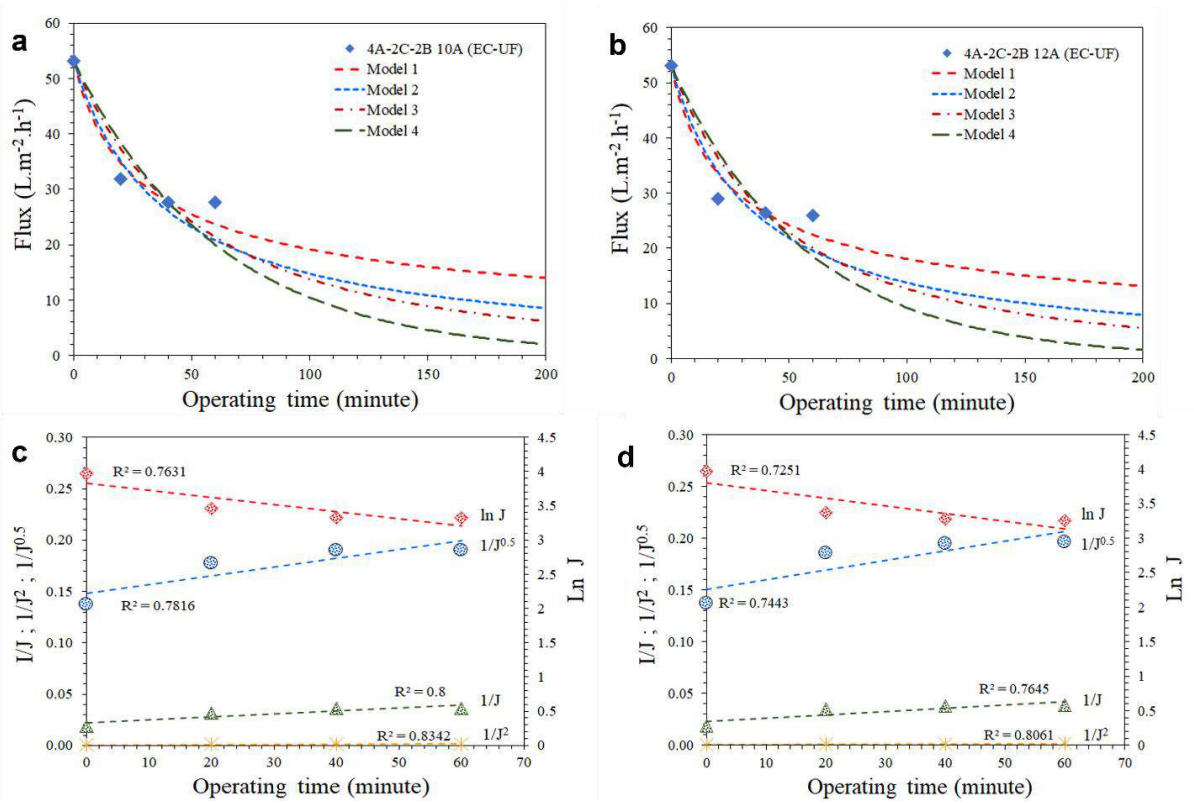


Figure 6. Fouling mechanisms in UF membrane according to Hermia's model

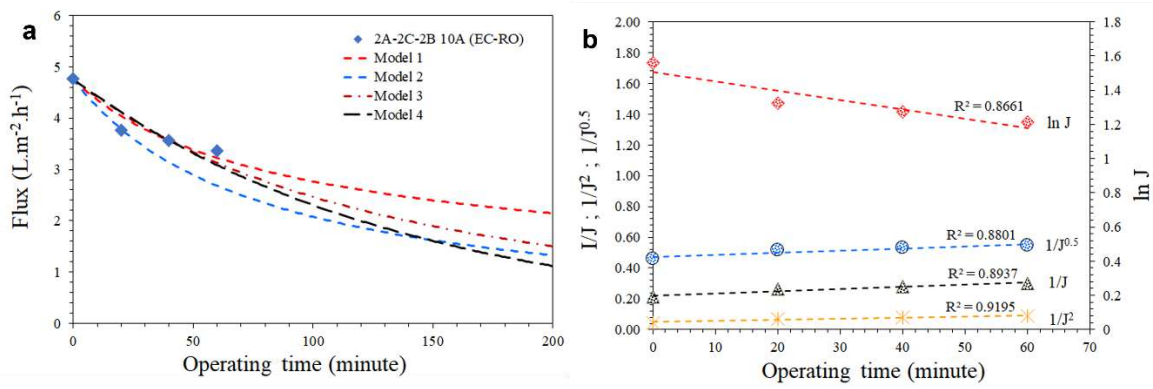


Figure 7. Fouling mechanisms in RO membrane according to Hermia's model

3.4. Operating cost of EC-UF and EC-RO system

The operating costs of EC-UF and EC-RO systems were calculated by equations and design parameters in sub-chapter 2.5. The operating costs of the two hospital waste treatment designs are shown in **Figure 8**. Based on the calculation results, there are slightly different between the operational cost of UF and RO membranes. It shows that the increase of production capacity from 5 to 50 m^3 reduced the operating cost from 3.92 to 0.89 US$.$m^{-3}$ for EC-UF system and from 4.02 to 0.93 US$.$m^{-3}$ for EC-RO system. By excluding labor costs, the operating cost was from 0.56 to 0.58 US$/m3 for EC-UF system and from 0.6 – 0.69 US$/m3 for EC-RO system. **Table 6** compares the operating costs of various EC processes in different wastewater treatments with this study. Most of the operating cost was calculated by Equation (12). In this research, the operating cost calculation involved the annual cost of the energy required in EC system and pump units, electrodes, membrane replacement for UF and RO, chemicals for UF and RO membrane, and labor. The detailed cost has been presented in Table 2. Compared to other studies, the integration of EC and UF or RO can be used as an alternative process to hospital wastewater treatment, which provides high removal efficiency of contaminants. Process optimization was further required to minimize the energy costs in the EC process, such as such minimized the spaces between the electrodes, varying the geometry of the electrodes, adjusting the conductivity of the electrodes, etc. (Mollah et al., 2004; Naje et al., 2017)

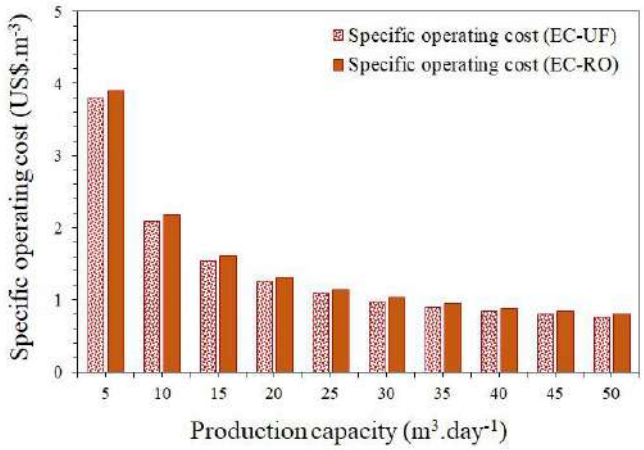


Figure 8. Operating cost of EC-UF and EC-RO systems at different production capacity

Table 6. Economic of various electrocoagulation plants for wastewater treatment and its comparison with this study.

Application	Mode and Electrode	Removal efficiency	Current density ($A.m^{-2}$)	Operating Cost (US.m^{-3}$)	Reference
Textile Wastewater	Mode: Batch Electrode: Al	Dyes: 98.6% COD: 84%	Voltage: 20 V Average current: 0.26 A	0.256	(Dalvand et al., 2011)
Poultry slaughterhouse wastewater	Mode: Batch Electrode: Fe	COD: 93%	150	0.3 – 0.4	(Bayramoglu et al., 2006)

Coal mine drainage wastewater	Mode: Batch Electrode: Fe	Heavy metal: 28.7–99.96 %	200 - 500	1.09 - 2.184	(Oncel et al., 2013)
Textile wastewater	Mode: batch Electrode: Al (monopolar)	COD: 15–62%	50–200	0.32–0.58 US$/kg COD	(Bayramoglu et al., 2004)
Chromium removal	Mode: Batch Electrode: Fe (monopolar)	Chromium: 99.64 %	89.45	0.207	(Patel and Parikh, 2021)
Olive oil wastewater	Mode: Batch Electrode: Al	COD: 78.51% Turbidity: 97.92%	150	0.12	(Niazmand et al., 2019)
Hospital wastewater (EC+UF)	Mode: Continue Electrode: Al (monopolar + bipolar)	TSS: 95.12% TDS: 97.53% BOD: 95.18% COD: 97.88%	66	3.92 (5 m^3 wastewater/ day, including labor cost) 0.89 (50 m^3 wastewater/ day, including labor cost)	This Study
Hospital wastewater (EC+RO)	Mode: Continue Electrode: Al (monopolar + bipolar)	TSS: 97.64% TDS: 99.85% BOD: 97.88% COD: 98.38%	77	4.02 (5 m^3 wastewater/ day, including labor cost) 0.93 (50 m^3 wastewater/ day, including labor cost)	This Study

4. Conclusion

In this work, the electrocoagulation (EC) process with membranes was integrated with membranes to treat real hospital wastewater. Two types of membranes were used, namely ultrafiltration (UF) and reverse osmosis (RO). In EC systems, Al electrodes were used, which were arranged in monopolar-parallel and bipolar configurations. There are two parameters studied in the EC system: the configuration of electrodes and applied currents. Two types of membranes were used for treating the effluent of EC process, namely ultrafiltration (UF) and reverse osmosis (RO). The effluent qualities of the two configurations of wastewater systems (EC-UF and EC-RO) were investigated towards the total suspended solids (TSS), chemical oxygen demand (COD), biological oxygen demand (BOD), and total dissolved solids (TDS). It was found that the integration of EC-UF with a configuration electrode of 4

anodes 2 cathodes 2 bipolar (4A-2C-2B) at an applied current of 12 A resulted in high removal of TSS, TDS, BOD, and COD by 95.12%, 97.53%, 95.18%, and 97.88%, respectively. The effluent quality was decreased by reducing the number of anodes from 4 to 2 pcs and applied current from 12 to 10 A. The configuration of 2A-2C-2B 10A resulted in TSS, TDS, BOD, and COD removal of 90.63%, 94.89%, 88.46%, and 90.63%. The lower quality of EC-UF system could be improved by substituting UF to RO membrane. The TSS, TDS, BOD, and COD removal were enhanced to 97.64%, 99.85%, 97.88%, and 98.38%. The UF membrane was more severe to fouling compared to the RO membrane, which was attributed to the formation of cake layer fouling on the membrane surface. The permeate flux decline in UF membrane system was 47.83% during 60 minutes of filtration time, while in the RO membrane system was 29.49. When the wastewater capacity was increased from 5 to 50 $m^3.h^{-1}$, the operating cost for the EC-UF system was decreased from 3.92 to 0.89 US$.$m^{-3}$, while the EC-RO system was decreased from 4.02 to 0.93 US$.$m^{-3}$. Since the EC-UF and EC-RO showed high efficiency of contaminants removal, these configurations could be used as alternative clean technology to produce clean water for water reuse purposes from hospital wastewater.

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☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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