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Research Article

Applications of the Taguchi Method for Key Parameter Screening in Electrodialysis Reversal Used for High Salinity Wastewater

Simulation tests were employed in the treatment of wastewater from seawater that was received from a specific power plant via flue gas desulfurization (FGD) for wastewater purification in this study. After water quality and compound variation were confirmed, a simulation was conducted to investigate the efficacy of pre-processing on the elimination of the majority of pollutants, like suspended solids, calcium ions, and magnesium ions. A mini scale electrodialysis reversal (EDR) mold plant was used to eliminate target pollutants, including sulfate and chloride ions. The Taguchi method was used to determine the optimal operating conditions that applied to various operating parameters, including temperature, flow rate, voltage, and current durations of power supply, to obtain the optimal operating parameters through analysis of variance (Minitab 14.0) simulation. After the simulations, the optimal parameters in the EDR process were used to demonstrate the removal efficiency of chloride, sulfate, and conductivity following treatment by EDR. The optimal operating parameters of the Taguchi method were 1 V (voltage), 3 L min^{-1} (flow rate), 15°C (temperature), 30 min (duration of power on), and 2 min (duration of power off). Combining the Taguchi method with the EDR process on wastewater treatment not only decreased initial conductivity from 4860 to 450 μS cm^{-1}, but also alleviated concentrations of sulfate and chloride ions.

Keywords: Conductivity; Desalination; Flue gas desulfurization (FGD); Petrochemical industry wastewater; Sulfate

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1 Introduction

Water consumption has increased with industrial development. Due to the shortages of potential water source, there has been an upsurge in water recovery approaches. Many researches regarding of assessment of water quality [1–4] and environmental impacts of wastewater treatment [5–7] have been concerned nowadays to resolve the critical risk of water source shortages. The petrochemical industry, for example, uses enormous amounts of water and discharges wastewater into the environment. Some water has to be separated regularly via flue gas desulfurization (FGD) systems, termed FGD wastewater, hence wastewater contained high concentration of sulfate ions (SO_4^{2-}) can be removed by FGD systems [8].

Wastewater always includes high concentrations of SO_4^{2-}, chloride ions (Cl^-), suspended solids (SS), and metals, such as magnesium ions (Mg^{2+}) and calcium ions (Ca^{2+}) [9]. The low pH of wastewater following the FGD process indicates acidic features, making it necessary to eliminate toxic pollutants from the wastewater before discharging it into the environment. Water shortages may be avoided via desalination of seawater. Practically suitable desalination processes for the water treatment of seawater can be effective in overcoming water shortages [10]. Electrodialysis (ED) is an effective process in not only treating waste acids or alkalis, but also has been manipulated for industrial uses in different applications, like brackish water desalination [11]. It is a membrane separation technology based on the selective migration of ions in wastewater. The ion exchange membranes are installed parallel between two electrodes [12]. Ions are regenerated by the anode, while extracted metallic cations are concentrated by the cathode [13]. After this process, anions and cations are separated, and wastewater is desalinated completely.

The method of electrodialysis reversal (EDR) is amended from the ED method and alters the electrode at a set time for membrane cleaning, eliminating impurities on the surface of membrane plates to enhance the recirculation time of the plates as well as the stability of the ED system. EDR is a desalination technology that has been used for many years and is also used to treat different kinds of pollution in wastewater [14–19]. The advantage of EDR in the desalination of surface and wastewaters is the ability to work without antiscalants. EDR also has higher levels of turbidity and a higher silt density index

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Abbreviations: **ANOVA**, analysis of variance; **ED**, electrodialysis; **EDR**, ED reversal; **FGD**, flue gas desulfurization; **OA**, orthogonal array; **S/N**, signal-to-noise ratio; **SS**, suspended solid

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than other membrane desalination processes [20]. Moreover, EDR is a desalination technique commonly known to be less sensitive to inorganic scaling than other techniques [21]. Therefore, EDR has been shown to be effective as a desalination method as laboratory and commercial applications support stable long-term operation of wastewater treatment [20].

The effectiveness of EDR desalination is generally affected by ion exchange ability, concentration, membrane, voltage, and flow rate [13, 22, 23]. The aim of this study was to change different factors, such as voltage, flow rate, temperature, and duration of power supply and to apply the Taguchi method to acquire the optimal operating parameters. Those optimal parameters were then employed in the EDR system to enhance water reclamation of wastewater.

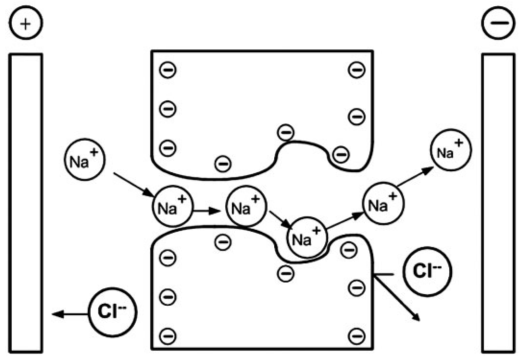

Figure 1. Migration of ions caused by an electric field [10].

2 Methods and materials

2.1 Sample sources

The wastewater samples were obtained from an FGD process that was composed of high concentrations of SO_4^{2-}, Mg^{2+}, SS, and lower concentrations of Cl^-, Ca^{2+}, and silicon (SiO_2). The composition of wastewater is illustrated in Table 1. The coagulant, calcium hydroxide ($Ca(OH)_2$), and sodium carbonate (Na_2CO_3) were added to completely eliminate Mg^{2+}, high concentration SS, and Ca^{2+} before proceeding with the EDR process.

2.2 Principles of EDR

ED is a membrane separation method where ions are transported by ion-permeable membranes and an external electrical current. The ion exchange membranes are alternately installed between two electrodes. One progressively decreases in salt concentration and is referred to as the product stream, while the other increases in concentration and is referred to as the concentration stream. The addition of acid and conditioning chemicals is required for membrane cleaning [12]. The transportation of ions under the ED process is shown in Fig. 1. The ED process deals with the problems of desalination of salted waters, wastewater minimization, ultrapure water production, concentration of dilute solutions, separation of electrolytes and non-electrolytes, and production of acids and alkalis from their salts [16].

EDR improved the ED process by reducing the conductivity of water via an uninterrupted process. A diagram of the EDR process is illustrated in Fig. 2. The electrodes of the EDR system are periodically reversed and ion transfer direction changes [24]. The periodic reverse of electrodes gives the EDR a self-cleaning mechanism that lessens the surface fouling on the ion exchange membrane. Therefore, the EDR system is able to maintain a stable flux without the effect of membrane fouling.

Table 1. Average characteristics of FGD process wastewater

	FGD process wastewater (±SD)	Unit
pH	8.5 ± 0.7	
Conductivity	4783 ± 158	$\mu S\,cm^{-1}$
Mg^{2+}	3769 ± 386	$mg\,L^{-1}$ as $CaCO_3$
Ca^{2+}	408 ± 35	$mg\,L^{-1}$ as $CaCO_3$
Total hardness	4100 ± 212	$mg\,L^{-1}$ as $CaCO_3$
Total dissolved solids	3764 ± 132	$mg\,L^{-1}$
Cl^-	40 ± 16	$mg\,L^{-1}$ as Cl^-
Total iron	0	$mg\,L^{-1}$as Fe
SO_4^{2-}	3650 ± 919	$mg\,L^{-1}$ as SO_4^{2-}
SiO_2	9.5 ± 3.5	$mg\,L^{-1}$ as SiO_2
Residual chlorine (Cl_2)	0.4 ± 0. 5	$mg\,L^{-1}$ as Cl_2
Chemical oxygen demand	8 ± 2	$mg\,L^{-1}$
SS	975 ± 389	$mg\,L^{-1}$

2.3 Taguchi experiment collocated with the EDR process

The main purpose in using the following experimental design is to provide maximum and reliable information with the fewest possible experimental runs. It uses a particular design of an orthogonal array (OA) to study the parameter space with a small number of experiments. The OA is the shortest possible matrix of combinations and all the parameters in OA are varied at the same time. Thus, their effects and performance interactions are studied simultaneously to determine which factors have the greatest effects [25, 26]. Based on the critical problem in this study, the Taguchi method developed by Dr. Taguchi [27], which is suitable in selection of influential factor to carry out the optimal parameters of a process or product of interest [28], is properly employed in resolving the problem. Taguchi's procedure used in this study is described as following steps: (i) select the critical influential factors involved in this study on both theoretical and practical aspects and define their corresponding levels, respectively; (ii) construct a proper table of OAs; (iii) execute experimental tests under various conditions listed in the orthogonal

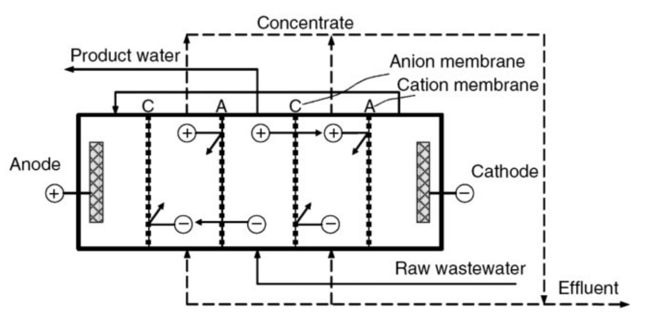

Figure 2. Schematic diagram of an EDR system (A: Anion membrane; C: Cation membrane) [10].

table and compute the relevant values; (iv) summarize the optimal conditions of test that carry out the optimum removal efficiency of contaminants in wastewater, and (v) justify the above optimal levels by levels by comparing all the signal-to-noise ratio (S/N) values resulting from various levels associated with the orthogonal table ($S/N = 10\log(1/\sigma^2)$), where S/N is signal-to-noise ratio and σ is standard deviation]. The above efforts can maximize the S/N value and hence minimize the variance of removal efficiency of contaminants data [29–31].

To analyze the significance of five factors (voltage, flow rate, temperature, duration for power on, and duration for power off) at four different levels, a full factorial experimentation would require 4^5 experiments to find the influential parameters, while the Taguchi design involves sixteen experiments using an OA L_{16} (4^5). An orthogonal array can demonstrate the definite sequence of each test, and the parameter design is the main purpose to determine a combination set of optimal factors. This set of factors appears to be less sensitive to noise factors, thus having the best stability.

The Taguchi method generally covers two parts, the S/N ratio and the analysis of variance (ANOVA) table, where the S/N ratio determines the level setting of the factors affecting quality variations. The S/N ratio depicts the quality stability, which means higher S/N ratio lesser loss.

The S/N ratio equation for three characteristics of quality (mapping) is listed below, where the number of test runs is depicted as n, and the value of one test is shown as y_i:

Nominal-the-best type I, the quality characteristic variance and theaverage bias should be concurrently indicated.

$$\frac{S}{N} = -10\log\frac{\sum_{i=1}^{n}(y_i - m)^2}{n} = -10\log\left[(\bar{y} - m)^2 + S_n^2\right] \quad (1)$$

The median of experimental results for each test is defined as m; the number of runs of each experimental group is denoted as n; the standard deviation is represented as S_n.

$$S_n = \sqrt{\frac{\sum_{i=1}^{n}(y_i - \bar{y})^2}{n}} \quad (2)$$

The mean of the experimental results of each experimental group is depicted as y.

Nominal-the-best type II is used in cases with one or more adjustable factors, where the mean can be adjusted to the target value without introducing bias, considers only the quality characteristic variance, and is actually applied to cases where the target value is equal to zero.

$$\frac{S}{N} = -10\log\frac{\sum_{i=1}^{n}(y_i - \bar{y})^2}{n} = -10\log(S_n^2) \quad (3)$$

Nominal-the-best type III is employed in cases with one or more adjustable factors, where only the quality characteristic variance is considered, and the mean can be adjusted to the target value without incurring bias.

$$\frac{S}{N} = -10\log\left(\frac{S^2}{y^2}\right) \quad (4)$$

Smaller-the-better:

$$\frac{S}{N} = -10\log\frac{\sum_{i=1}^{n} y_i^2}{n} = -10\log(\bar{y}2 + S_n^2) \quad (5)$$

Larger-the-better:

$$\frac{S}{N} = -10\log\frac{\sum_{i=1}^{n}(1/y_i^2)}{n} \quad (6)$$

After S/N ratios are calculated, S/N ratio curves can be plotted according to the level of each controllable factor in order to represent which level combination has the optimal effect [31–33].

As the results of the Taguchi method, as indicated in Eq. (6), the larger of S/N is considered optimal in this study. Therefore, an L_{16} comparison list could acquire optimal parameters through ANOVA (Minitab 14.0) simulation and then utilize the best parameters in the EDR process to demonstrate the removal efficiency of Cl^-, SO_4^{2-}, and conductivity after EDR process treatment. Experimental parameters of the L_{16} comparison list include voltage, flow rate, temperature, and duration of power supply. The data obtained from the experiments could next be analyzed for further test of contaminants elimination.

2.4 Temperature parameter

Temperature rise can increase the ion activity in water, so it increases the water reclamation rate of the EDR system by altering a solution's viscosity. From the open literature, many studies have found that temperature change in the EDR system not only varied the solution's viscosity and density, but also influenced the migration speed of ions and ion diffusion [34–36].

2.5 Flow rate parameter

The EDR process leads wastewater into a central compartment and selectively moves aqueous ions through ion exchange membranes, with outside electric fields as the driving force. A lower flow rate during the EDR process leads to increased desalination. However, a very low flow rate will result in membrane fouling and affect ion migration between the two electrodes, causing a decrease in desalination efficiency.

2.6 Voltage parameter

The applied cell voltage is an operating condition in the ED process, as the voltage determines the current in the cell and hence the desalination efficiency and energy consumption [37].

2.7 Duration of power supply

The EDR system should be turned off during desalination, fouling on the ion exchange membrane should be eliminated, and the optimal time of power supply duration must be determined.

2.8 Analysis of component elements

An ion chromatograph (Dionex, DX-120) and an inductively coupled plasma atomic emission spectrometer (Optima, 5100DV) were employed to determine the ion concentration and the metal ion concentration in the aqueous phase, respectively. The analysis for ion concentration is adopted by the method (W415.52B), and the analysis for metal ion concentration is chosen by the method (M105.00B) from Environmental Analysis Laboratory, Environmental Protection Administration, Executive Yuan, Taiwan, ROC [38].

3 Results and discussion

3.1 Taguchi's experimental results

To obtain better responses, the following experiments were applied to the S/N calculation. The S/N ratios for every factor at four levels are presented in Fig. 3a–e and the larger result of S/N is considered best. Figure 3a indicates that the S/N ratios are 38.73, 35.99, 37.44, and 34.45 at a temperature setting of 15, 20, 30, and 40°C, respectively. The greatest efficiency of wastewater treatment under EDR process occurs at 15°C. Figure 3b depicts that for 15, 20, 30, and 40°C the S/N is 35.35, 36.88, 37.51, and 36.88 at a power duration of 10, 20, 30, and 40 min, respectively. The removal efficiency of pollutants increases with increases in power duration. The duration for power on will affect the efficiency of treatment under the EDR process, but the most efficient time of tolerance of the EDR membrane in this study is 30 min. Figure 3c indicates the EDR process was affected by flow rates. With flow rates of 3, 4, 5, and 6 L min^{-1}, S/N is 38.08, 36.37, 36.11, and 36.06, respectively. The hydraulic retention time was increased by a low flow rate. It increased the residence time for wastewater in the electrode. The benefit of Taguchi's experimental results was to enhance the removal efficiency for high conductivity.

Furthermore, according to Fig. 3d, at a voltage of 1 V, ions of the solution can be attracted, with the phenomenon of electron transmission caused by the electrical charge of ions, the anode ions transported to the cathode and the cathode ions transported to the anode. The transportation velocity of ions is related to the enforced increase of electric film and the transportation velocity is elevated when the voltage of anode and cathode electrodes is increased. From Fig. 3e, the duration for power off for 2 min is optimal, so that fouling on the ion exchange membrane can be eliminated.

3.2 EDR desalination rate

The plot of average conductivity of raw wastewater is shown in Fig. 4a, in which the average conductivity of raw wastewater is 4748 μS cm^{-1}. After applying experimental parameters, which are shown in Table 2, to ANOVA simulation software, the results under ANOVA simulation are displayed in Table 3. We adopted the optimal operating conditions (1 V, 3 L per min, 15°C, duration for power on: 30 min, duration for power off: 2 min) in the EDR system to process desalination. Figure 4b shows the result of the EDR desalination process where the initial conductivity decreased from 4860 to

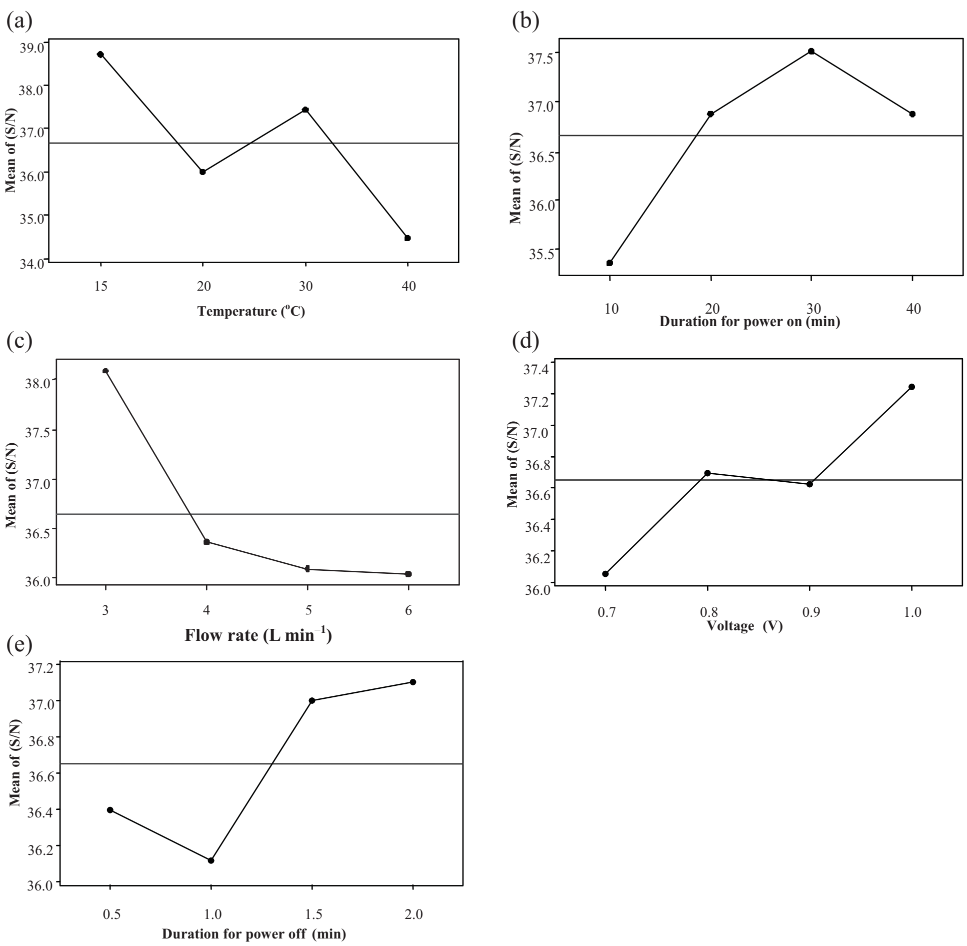

Figure 3. (a) (S/N) versus temperature, (b) (S/N) versus duration for power on, (c) (S/N) versus flow rate, (d) (S/N) versus voltage, and (e) (S/N) versus duration for power off.

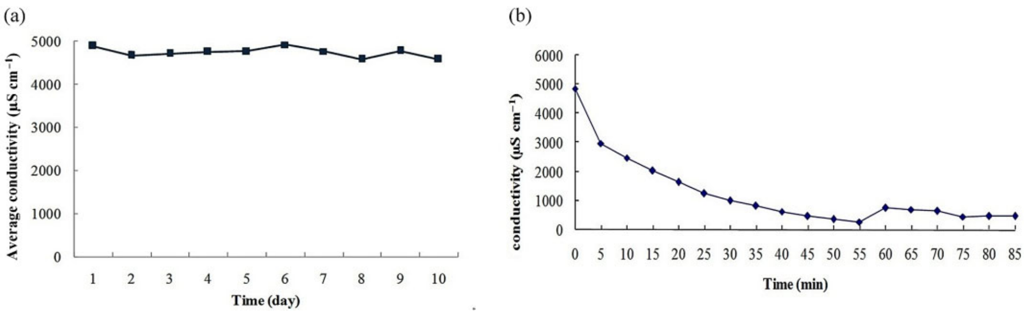

Figure 4. (a) Average conductivity versus time and (b) conductivity versus time.

Table 2. Experimental parameters used in this study

Class	Voltage (V)	Flow rate (L min^{-1})	Temperature (°C)	Duration for power on (min)	Duration for power off (min)
1	0.7	3	40	20	1.5
2	0.7	4	30	10	2
3	0.7	5	20	40	0.5
4	0.7	6	15	30	1
5	0.8	3	30	40	1
6	0.8	4	40	30	0.5
7	0.8	5	15	20	2
8	0.8	6	20	10	1.5
9	0.9	3	20	30	2
10	0.9	4	15	40	1.5
11	0.9	5	40	10	1
12	0.9	6	30	20	0.5
13	1	3	15	10	0.5
14	1	4	20	20	1
15	1	5	30	30	1.5
16	1	6	40	40	2

Table 3. Results under ANOVA simulations

Voltage (V)	S/N	Flow rate (L min^{-1})	S/N	Temperature (°C)	S/N	Duration for power on (min)	S/N	Duration for power off (min)	S/N
0.7	36.05	3	38.08	15	38.73	10	35.35	0.5	36.39
0.8	36.69	4	36.37	20	35.99	20	36.88	1.0	36.11
0.9	36.62	5	36.11	30	37.44	30	37.51	1.5	37.00
1.0	37.24	6	36.06	40	34.45	40	36.88	2.0	37.10

450 μS cm^{-1} (final conductivity), and the desalination efficiency was 91%.

3.3 Cl^- and SO_4^{2-} removal

Cl^- variations in raw wastewater are recorded in Fig. 5, and Cl^- concentrations in raw wastewater varied between 250 and 350 mg L^{-1}, with an average concentration of 303 mg L^{-1}. The removal efficiency of chloride desalination is shown in Fig. 6, where the initial concentration of Cl^- (303 mg L^{-1}) decreased to a final concentration (29 mg L^{-1}) at the 75th min and the removal efficiency was 90%.

SO_4^{2-} variations in raw wastewater are illustrated in Fig. 7, with an average concentration of 3326 mg L^{-1}. After the EDR desalination process, the original concentration of SO_4^{2-} (3150 mg L^{-1}) was significantly reduced to the final concentration (47 mg L^{-1}) at the 75th min and the removal efficiency of desalination was 98% (Fig. 8). With decreasing conductivity, the concentration of Cl^- and SO_4^{2-} also relatively decreased.

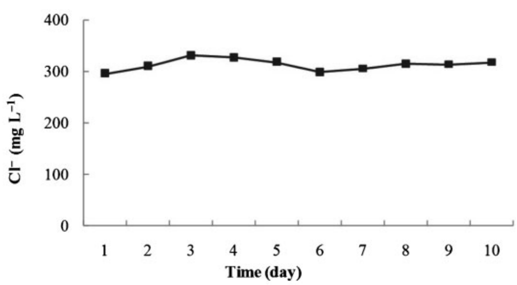

Figure 5. Cl^- concentration variations in raw wastewater.

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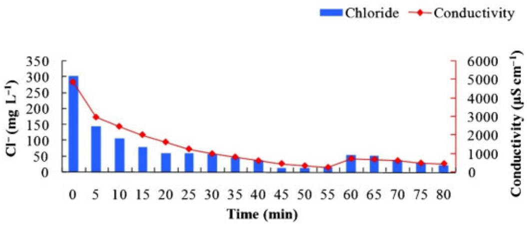

Figure 6. Removal efficiency of Cl^- desalination and conductivity.

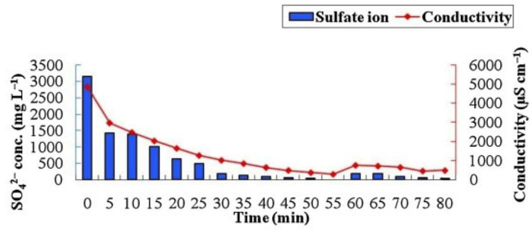

Figure 8. Removal efficiency of SO_4^{2-} desalination and conductivity.

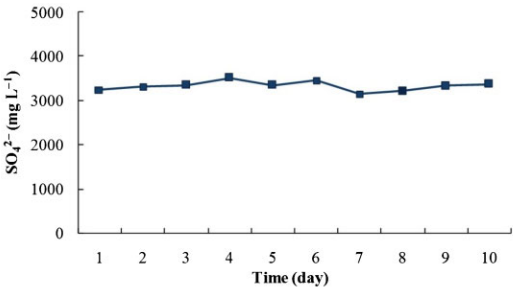

Figure 7. SO_4^{2-} concentration variations in raw wastewater.

3.4 Overall removal efficiency

The average removal efficiencies of each monitored item are summarized in Table 4. When operating at the optimal conditions, raw wastewater with a conductivity of 4860 $\mu S\,cm^{-1}$, a Cl^- concentration between 250 and 350 $mg\,L^{-1}$, and a SO_4^{2-} average concentration of 3326 $mg\,L^{-1}$, the EDR system has an average desalination efficiency of 91%, a Cl^- removal efficiency of 90%, and a SO_4^{2-} removal efficiency of 99%. The final conductivity was 450 $\mu S/cm$ and the final concentrations of Cl^- and SO_4^{2-} were 29 and 47 $mg\,L^{-1}$, respectively.

Table 4. Average removal efficiencies of each monitored item

Average concentration	Conductivity ($\mu S\,cm^{-1}$)	SO_4^{2-} ($mg\,L^{-1}$)	Cl^- ($mg\,L^{-1}$)
Initial concentration	4860	3150	303
Final concentration	450	47	29
Removal efficiency (%)	91	99	90

4 Concluding remarks

Wastewater treatment has become a critical global issue due to the fast-developing industry. Researchers from many different countries have studied this issue for the past several decades and achieved some practical and excellent results to desalinate wastewater from the industrial process or seawater [10, 12, 33, 39]. As previous studies demonstrated, the most common pollutants, such as suspended solids, calcium ions, magnesium ions, chemical oxygen demand, and conductivity, exist in wastewater from industrial processes, so the most efficient way, the ED process combined with ANOVA or EDR process, is employed to eliminate those most common pollutants efficiently. With the rapid development of Taiwan's industry over the past three decades, the consequent outcome, wastewater, has resulted in environmental problems in Taiwan. As the resolution of this consequent outcome, the process of high removal efficiency of

Table 5. Experiments with different operating parameters

Class	Voltage (V)	Flow rate ($L\,min^{-1}$)	Temperature (°C)	Duration for power on (min)	Duration for power off (min)	Removal efficiency (%)
1	37.24	38.08	38.73	35.35	36.39	90
2	37.24	36.37	35.99	36.88	36.11	63
3	37.24	36.11	37.44	37.51	37.00	86
4	37.24	36.06	34.45	36.88	37.10	57
5	36.62	38.08	35.99	37.51	37.10	86
6	36.62	36.37	38.73	36.88	37.00	89
7	36.62	36.11	34.45	35.35	36.11	40
8	36.62	36.06	37.44	36.88	36.39	69
9	36.69	38.08	37.44	36.88	36.11	85
10	36.69	36.37	34.45	37.51	36.39	55
11	36.69	36.11	38.73	36.88	37.10	88
12	36.69	36.06	35.99	35.35	37.00	53
13	36.05	38.08	34.45	36.88	37.00	62
14	36.05	36.37	37.44	35.35	37.10	61
15	36.05	36.11	35.99	36.88	36.39	55
16	36.05	36.06	38.73	37.51	36.11	78
Maximum optimal	37.24	38.08	38.73	38.73	37.10	–
operating conditions (compared with Tab. 3)	1 V (37.24)	3 $L\,min^{-1}$ (38.08)	15°C (38.73)	30 min (38.73)	2 min (37.10)	91

pollutants can be adopted to desalinate wastewater. The desalination process of this study is as follows.

A coagulation–flocculation process is applied to completely eliminate Mg^{2+}, SS, and Ca^{2+} by adding $Ca(OH)_2$ and Na_2CO_3 to wastewater, and then Taguchi's experimental method combined with the EDR process is adopted to remove SO_4^{2-} and Cl^-, resulting in water reclamation. Through experiments with different operating parameters, as shown in Table 5, we found that parameters in the L_{16} comparison list would best influence the EDR process treatment. The effective factors of S/N include temperature, flow rate, and duration of power on or off. Other factors, such as lower flow rate and longer duration of power on, can also enhance the efficiency of wastewater treatment. Moreover, adopting the optimal parameters in the EDR system not only significantly reduced conductivity from 4860 to 450 $\mu S\,cm^{-1}$, but also decreased concentrations of SO_4^{2-} and Cl^- in the treatment of wastewater containing high concentrations of SO_4^{2-} and Cl^-. Therefore, the result of this study also demonstrates a good desalination process of wastewater that is as good as the previous study's results.

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