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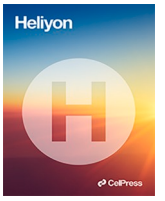

Research article

Optimization of operating parameters for 2,5-furandicarboxylic acid recovery using electrodialysis with bipolar membrane and traditional electrodialysis systems

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ABSTRACT

Electrodialysis (ED) is an eco-friendly and feasible method to separate or recover ionic compounds by electric field attraction and configuration of ion exchange membranes. Strain *Burkholderia* sp. H-2 could biotransform 5-hydroxymethylfurfural (5-HMF) into a green platform compound, 2,5-furandicarboxylic acid (FDCA), using a bioreactor system. In this study, electrodialysis with the bipolar membrane (EDBM) and traditional ED systems were applied to recover and concentrate FDCA. Artificial and real FDCA effluents of the 5-HMF biotransformation bioreactor were used as the feedstock to establish the optimal conditions for FDCA recovery. The optimal FDCA concentration and pH of the artificial FDCA effluent were 2100 mg/L and 5, respectively. The suitable current density of the EDBM was 8.93 mA/cm^2. For FDCA recovery and concentration using the ED, the feedstock volume and FDCA concentration in the concentration chamber were 1.5 L and 1000 mg/L, respectively. The FDCA recovery efficiency of the real FDCA effluent was 55.6 %. Suppose the pretreatment procedure of the real bioreactor effluent is further optimized. It is believed to benefit the enhancement of FDCA recovery efficiency and reduce energy consumption.

1. Introduction

2,5-Furan-dicarboxylic acid (FDCA) is a green platform chemical in the top-15 list of compounds provided by the US Department of Energy (DOE) [1]. Its furan ring connecting two carboxyl groups makes it versatile for polyester, medical, and fire protection industrial applications. FDCA also can be applied to textiles, carpets, food packaging, and electronic materials industries [2–4]. There are four methods to produce FDCA, which are dehydration of hexose sugar derivatives, oxidation reaction of 5-hydroxymethylfurfural (5-HMF), catalyst conversion of furan derivatives, and 5-HMF biotransformation [5–7]. Among these methods, applying 5-HMF biotransformation to produce FDCA has become an increasingly important topic because of low cost, fewer by-products, and environmental friendliness concerns. Our bacterial isolate, *Burkholderia* sp. H-2 is capable of biotransforming 5-HMF into FDCA [8,9]. After cell immobilization, this strain is used for 5-HMF biotransformation in a bioreactor for FDCA production. Therefore, FDCA recovery is

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Electrodialysis (ED) is a technology for water purification by removing ions from water [10]. The essential components of the ED system are cation exchange membranes (CEM), anion exchange membranes (AEM), and electrodes (anode and cathode set on both sides.) The staggered CEMs and AEMs stack form dilution and concentration chambers. The diluted chamber is fed with water with ions, and the concentrated chamber collects the ions. When an external voltage is applied, cations in the diluted chamber migrate to the cathode and pass through CEM but not AEM. Conversely, anions migrate to the anode through the AEM. Thus, the ion concentration in the diluted chamber gradually declines to achieve water purification [11]. The advantages of the ED system include technological compatibility and extensive types of influent treatment [12]. Electrodialysis with bipolar membrane (EDBM) is a new type of membrane technology that combines electrodialysis and bipolar membrane (BPM) and is applied for acid and base recovery. BPM is a composite membrane composed of cation exchange, anion exchange, and interfacial layers. Water is dissociated in the BPM. Then, H^+ and OH^- ions migrate to combine with the anions and the cations to achieve acid and base recovery in the acid and base chambers, respectively [13], which is more environmentally friendly than other ED technologies [14]. EDBM is widely used in the recovery of organic acids, such as lactic acid [15], succinic acid [16], and citric acid [17]. Thus, ED technology is suitable for FDCA recovery.

Several factors affect organic acid recovery using EDBM and ED systems. The membrane configuration of EDBM is mainly divided into three types (BPM-CEM-BPM, BPM-AEM-BPM, and BPM-AEM-CEM-BPM). Three membranes unit constructs two chambers system, and four membranes unit is three chambers system. For weak organic acid recovery, the BPM-AEM-BPM arrangement is superior to the BPM-AEM-CEM-BPM arrangement because this membrane configuration has lower internal resistance, improving the acid recovery efficiency [18]. Thus, the BPM-AEM-BPM arrangement is used for FDCA recovery in this research. The organic acid salt concentration in the feedstock also influences organic acid recovery efficiency. When the influent formate concentration increased from 0.1 mol/L to 0.4 mol/L, the EDBM efficiency increased accordingly. However, when the influent formate concentration was higher than 0.4 mol/L, formate ions diffused from the acid chamber back to the salt chamber. The formic acid recovery efficiency decreased [19]. Current density is a crucial factor in the ED and EDBM systems. High current density means high voltage and electrical driving force and results in a faster ion migration speed. However, if the EDBM system exceeds the limit current density, concentration polarization occurs, and the voltage may be too high to decrease the EDBM operation safety [20]. The dissociation state of organic acids in the salt chamber also affects the EDBM performance because only the dissociated acid can migrate under the electrical field. The dissociation constants of various organic acids are different, resulting in different dissociation states at different pH values. The highest α-ketoglutaric acid recovery concentration (2.18 g/L) was achieved at the feedstock pH of 3. When the feedstock pH increased from 6 to 10, the OH^- concentration in the feedstock significantly increased. The OH^- ions competed with α-ketoglutarate ions, deteriorating EDBM performance [21].

Based on the advantages of ED technology, EDBM and ED systems were applied to recover and concentrate FDCA in this study. To our knowledge, no literature has tried to recover FDCA using ED and EDBM systems. Establishing parameters of ED technology for FDCA recovery is beneficial to constructing the FDCA biorefinery procedures. In this study, an artificial bioreactor effluent was prepared at first, and it was used as the feedstock to establish suitable parameters for EDBM and ED system operation. For the EDBM system, the investigated factors included feedstock FDCA concentration, current density, and feedstock pH. Then, these parameters were applied to the following ED system to concentrate FDCA further. The factors studied for the ED system were feedstock volume and the FDCA concentration in the concentrated chamber. Finally, the real bioreactor effluents after different pretreatments were used as the ED system feedstock, and the ED system was operated under the optimal conditions established by the previous EDBM and ED system experiments. The FDCA recovery difference between the artificial and real bioreactor effluents was compared to evaluate the feasibility of FDCA recovery using ED and EDBM systems.

2. Materials and methods

2.1. Artificial bioreactor effluent preparation

The composition of the artificial bioreactor effluent was mainly according to the mineral salt medium (MSM) used for 5-HMF biotransformation and included 0.334 g/L NH_4Cl, 0.015 g/L $CaCl_2$, 0.2 g/L $MgSO_4{\cdot}7H_2O$, 0.5 g/L K_2HPO_4, 0.5 g/L KH_2PO_4, 300 mg/L $FeSO_4{\cdot}7H_2O$, 40 mg/L $ZnSO_4{\cdot}7H_2O$, 106 mg/L $CoCl_2{\cdot}6H_2O$, 50 mg/L $MnSO_4{\cdot}H_2O$, 34 mg/L $Na_2MoO_4{\cdot}2H_2O$, 50 mg/L $CuSO_4{\cdot}5H_2O$, and 650 mg/L Na_2SO_4. FDCA concentration was added according to the experimental design. The pH of the artificial bioreactor effluent was adjusted to 6 using 1 N NaOH and H_2SO_4.

2.2. EDBM and ED systems setup

2.2.1. EDBM system setup

The EDBM system comprised four pairs of membrane units (AEM-BPM membrane configuration) with two chambers (acid and salt chambers). The effective membrane area was 112 cm^2. The artificial bioreactor effluent was used as the feedstock of the salt chamber, and the 1 g/L H_2SO_4 was fed into the acid chamber. The anode and cathode were both titanium-based electrode plates. The electrode solution was 3 % (w/v) sodium sulfate. The volume of each chamber was 500 mL. Each chamber was equipped with a pump to supply a continuously circular flow at a flow rate of 45 L/h. The pH and conductivity meters were set to monitor the pH and conductivity variations of the acid and salt chambers in real time during the experimental period.

2.2.2. The ED system setup

Traditional ED system is applied or feedstock desalination. However, from another point of view, it can be used for salts and organic acids concentration. Thus, the ED system was used for FDCA concentration in this study. The ED system comprised four pairs of membrane units (CEM-AEM membrane configuration) to form the concentrated and diluted two chambers. Each ion exchange membrane had an effective membrane area of 112 cm^2. The artificial or real bioreactor effluent was used as the feedstock for the diluted chamber, and the concentrated chamber was fed with low-concentration FDCA. The electrode and its chamber were the same as the design of the EDBM system. The electrode chamber solution was also 3 % sodium sulfate. The working volume of each chamber was 500 mL. Three pumps were set for continuous solution circulation in the concentrated, diluted, and electrode chambers at a flow rate of 45 L/h. The monitoring equipment was equipped to monitor the pH value and conductivity of each chamber in real time. Fig. 1 presents a schematic diagram of the EDBM and ED systems. The specifications of membranes are summarized in Table S1. After each set of experiments was completed, the EDBM and ED system were disassembled for cleaning. The cleaning method was to rinse the membranes (AEM, CEM, and BPM) with DI water, and then soak membranes in 3 % HCl and 3 % NaOH for one day each. Finally, the membranes were soaked in 3 % NaCl for storage.

2.3. FDCA recovery and concentration using the EDBM and ED systems

2.3.1. The operated parameters establishment of the EDBM system

There are many factors affecting the organic acids recovery using EDBM. The investigated factors included FDCA concentration in salt chamber feedstock, current density, and salt chamber feedstock pH. For the FDCA concentration in the salt chamber feedstock experiment, the artificial bioreactor effluent (pH 6) containing different FDCA concentrations (700, 1400, 2100, and 2800 mg/L) was used as the feedstock of the salt chamber. Before applying the electrical field, the solution of each chamber was circulated for 10 min for system stabilization. Then, the EDBM system was operated at the current density of 4.46 mA/cm^2. The conductivity and pH of the two chambers were automatically monitored every minute, and FDCA concentrations in the two chambers were regularly sampled. The experimental procedure for the current density experiment was the same as the one mentioned above. However, FDCA concentration in the salt chamber feedstock was designed according to the result of the FDCA concentration in the salt chamber feedstock experiment. The EDBM system was operated at various current densities (4.46, 8.93, 13.39, and 17.86 mA/cm^2). The experimental protocol followed the abovementioned protocol for the salt chamber feedstock pH experiment. FDCA concentration in the salt chamber and current density were decided according to the results of the EDBM experiments mentioned above. However, the salt chamber feedstock pH was adjusted to 5, 6, 7, and 8, respectively.

2.3.2. The operated parameters establishment of the ED system

2.3.2.1. The artificial bioreactor effluent as feedstock. To optimize the ED operation, the feedstock volume of the diluted chamber and

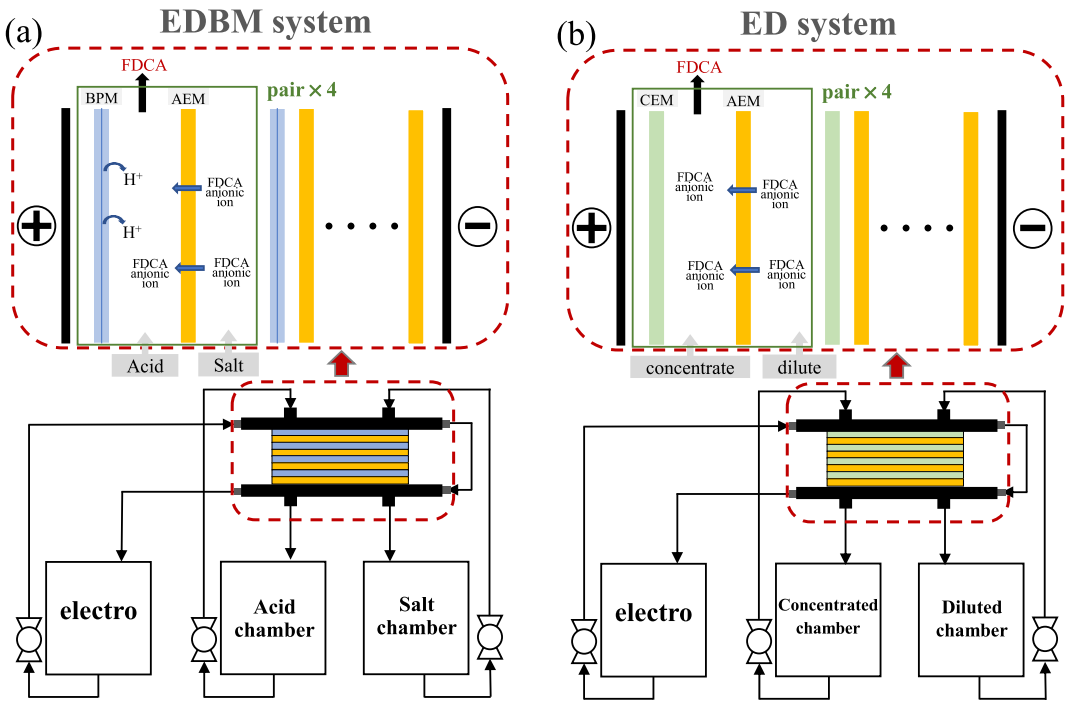

Fig. 1. The construction of EDBM (a) and ED (b) systems.

FDCA concentration in the concentrated chamber were investigated. For the feedstock volume of the diluted chamber experiment, the whole protocol was the same as 2.3.1. The FDCA concentration and pH of the feedstock for the diluted chamber and current density were decided based on the results of the EDBM system. The FDCA concentration of the concentrated chamber was 1.0 g/L. However, the feedstock volume of the diluted chamber was adjusted to 0.5, 1.0, and 1.5 L, respectively. In the FDCA concentration in the concentrated chamber experiment, the FDCA concentration was adjusted to 500, 1000, 1500, and 2100 mg/L, respectively. The experimental procedure followed the procedure mentioned in 2.3.1.

2.3.2.2. The real bioreactor effluent as feedstock. After establishing the operated parameters for FDCA recovery from the artificial bioreactor effluent using the EDBM and ED systems, the real bioreactor effluent replaced the artificial bioreactor effluent as the feedstock. Three batch bioreactor effluents after different treatments were used so the effect of effluent treatment on FDCA recovery could also be studied. Various treatment of three real bioreactor effluents is presented in Table 1. The experimental procedure was the same as the procedure mentioned in 2.3.1. However, the feedstock pH, current density, feedstock volume, and FDCA concentration in the concentrated chamber were set according to the results of 2.3.1 and 2.3.2.1.

2.4. Analysis methods

The pH value was measured with a pH meter (Mettler Toledo-20). Conductivity was measured with a conductivity meter (HACH/sensIONTM + EC7). FDCA was analyzed using high-performance liquid chromatography (HPLC) equipped with a UV detector. The column was Mightysil RP-18 GP (5 μm, 250 × 4.6 mm). After proper dilution and filtration with a 0.22 μm filter, a 20 μL sample was injected into the HPLC system. The detection wavelength of FDCA was 245 nm. The mobile phase was acetic acid: water (4:6) with a 0.6 mL/min flow rate.

2.5. Calculations

FDCA recovery efficiency (R), current efficiency (CE), energy consumption (E), and FDCA flux (J) are important parameters for evaluating electrodialysis performance [14,22], and the relevant calculation equations are as follows.

(1) FDCA recovery efficiency (R)

$$R(\%) = \frac{\Delta C_{FDCA} \text{ in the acid (concentrated) chamber (mg/L)} \times V \text{ (L)}}{\Delta C_{FDCA} \text{ in the salt (diluted) chamber (mg/L)} \times V \text{ (L)}} \times 100\% \quad (1)$$

ΔC_{FDCA} is the FDCA concentration difference before and after the experiment; V is the working volume (L) in the chamber.

(2) Current efficiency (CE)

$$CE(\%) = \frac{F \times z \times V \times \Delta C_{FDCA}}{N \times I \times \Delta T} \times 100\% \quad (2)$$

ΔC_{FDCA} is the FDCA concentration difference before and after the experiment; z is the ion valence; V is the working volume (L) in the chamber; F is Faraday's constant (96485 C/mol); N is the number of electrodialysis units (N = 4); I is current (A); Δt is experimental time (s).

(3) Energy consumption (E)

$$E(kWh/kg) = \int \frac{U \times I}{\Delta m} dt \quad (3)$$

U is the voltage drop across the membrane (V); I is the current (A); Δm is the FDCA mass difference before and after the experiment (g); t is the experimental time (h).

(4) FDCA flux (J)

$$J\left(mol/m^2h\right) = \frac{n}{N \times A \times t} \quad (4)$$

Table 1
Various treatments of the real bioreactor effluents.

Code number	FDCA conc. (mg/L)	Centrifugation (speed, time)	Filter pore size (μm)
I	1573	8000 rpm, 10 min	70
II	2671	10000 rpm, 15 min	70
III	2701	10000 rpm, 15 min	0.22

n is the FDCA moles transferred from the diluted chamber to the concentrated chamber (mol); N is the number of electrodialysis units (N = 4); A is the effective area of the ion exchange membrane (m^2); t is the experimental time (h).

3. Results and discussion

3.1. FDCA recovery using the EDBM system with the artificial bioreactor effluent as feedstock

3.1.1. The effect of FDCA concentration in the feedstock

Increasing the organic acid salt concentration in the feedstock is beneficial for organic acid recovery. However, a high concentration of organic acid recovered in the acid chamber might lead to the natural diffusion of organic acids back to the salt chamber to lower acid recovery efficiency [19], Fig. 2 presents the variation of the voltage and FDCA recovery efficiency of the EDBM system at different FDCA concentrations in the feedstocks. The variation of the FDCA concentrations in the salt and acid chamber is shown in Fig. S3. The trend of the voltage curve in each group was similar. The voltage of each group was 8 V at the beginning of the experiment. With the system operation, the voltage of each group decreased to 7 V, except for the group of 700 mg/L FDCA in the feedstock. The FDCA recovery efficiencies of 700, 1400, and 2100 mg/L FDCA were 62.2 %, 64.0 %, and 65.6 %, respectively, indicating that high influent FDCA concentration was accompanied by high FDCA recovery efficiency. When further increasing FDCA concentration to 2800 mg/L, the FDCA recovery efficiency within the first 10 min was slightly higher than those of other groups. The accumulated FDCA concentration of 2800 mg/L FDCA group was the highest in the acid chamber, compared to other groups (Fig. S3). However, the FDCA recovery efficiency began to decrease after 50 min operation. The recovery efficiency of 2800 mg/L FDCA at the end of the experiment was only 40.5 %. Due to the high organic acid concentration gradient between the salt and the acid chambers, the organic acid ions diffused from the acid chamber back to the salt chamber. Thus, the organic acid recovery efficiency declined [23]. Our result was consistent with the results of the published reference. Based on the above results, 2100 mg/L FDCA was chosen for the following experiments.

3.1.2. The effect of current density

Fig. 3 displays the voltage and FDCA recovery efficiency variations of the EDBM system at different current densities. The high current density caused the high initial voltage. At the beginning of the experiment, due to the significant conductivity difference between the salt and acid chambers, the system resistance was relatively large, resulting in a higher voltage. As the system operated, the voltage of each current density gradually decreased within 30 min. The reason might be that the conductivity difference between the salt and acid chambers decreased, which reduced the system resistance and voltage. The voltages of the current densities in the order of low to high declined from 9, 11, 15, and 16 V to 8, 9, 10, and 11 V, respectively. The FDCA recovery efficiency increased with the increase of current density when the current density was between 4.46 and 8.93 mA/cm^2. At the end of the experiment, the highest FDCA recovery efficiencies of the current density 4.46 and 8.93 mA/cm^2, were 63.2 % and 68.5 %, respectively. When further increasing current density to 13.39 and 17.86 mA/cm^2, the FDCA recovery efficiency significantly increased within 10 min. The variations of FDCA concentrations at the current density of 13.39 and 17.86 mA/cm^2 in the acid and salt chambers were more significant than those at the current density of 4.46 and 8.93 mA/cm^2 (Fig. S4). The best FDCA recovery efficiencies of current density 13.39 and 17.86 mA/cm^2 were obtained at 20 and 10 min, respectively, and were 53.2 % and 51.0 %, respectively. However, with continuous operation, the FDCA recovery efficiencies of the two current densities remained stable and could not increase further. High current density resulted in a faster migration rate of organic acid ions and improved the efficiency of ions passing through the AEM. However, increasing the current density also resulted in a higher voltage, which promoted water dissociation in the BPM and might cause concentration polarization. Therefore, organic acid recovery efficiency declined [24]. The current density of 8.93 mA/cm^2 had the highest FDCA recovery efficiency, so this current density was applied for the following experiments.

3.1.3. The effect of feedstock pH

The variations of the voltage and FDCA recovery efficiency of the EDBM using the feedstocks with different pHs are shown in Fig. 4.

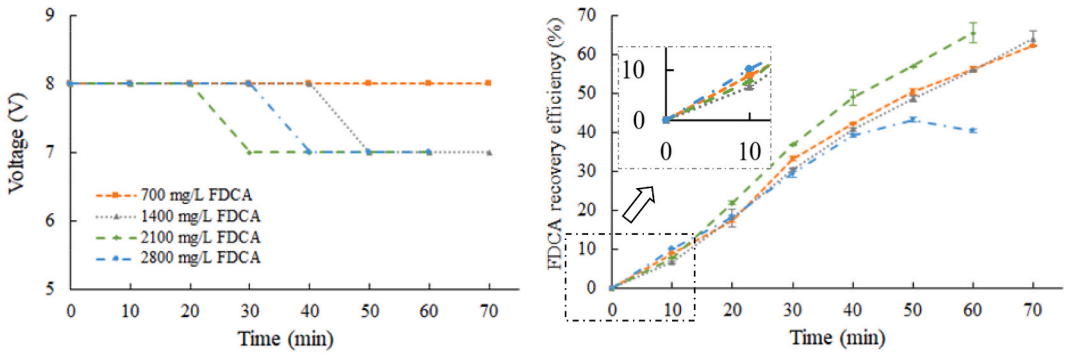

Fig. 2. The variation of the voltage and FDCA recovery efficiency of the EDBM system at different FDCA concentrations in the feedstocks.

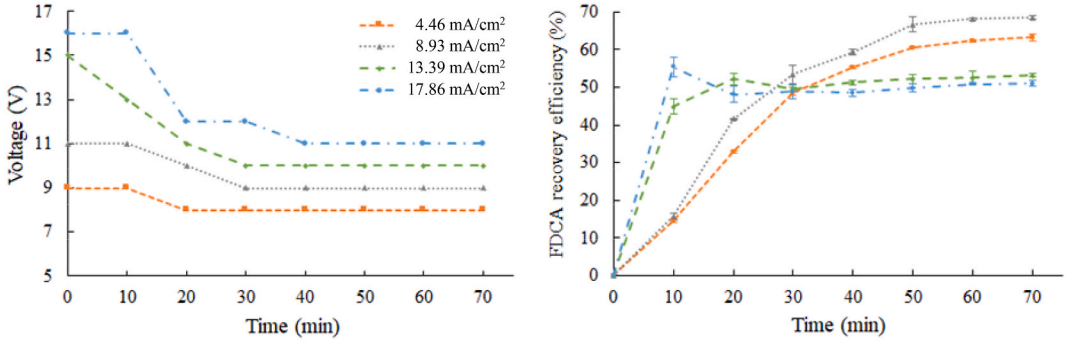

Fig. 3. The variations of voltage and FDCA recovery efficiency of the EDBM system at different current densities.

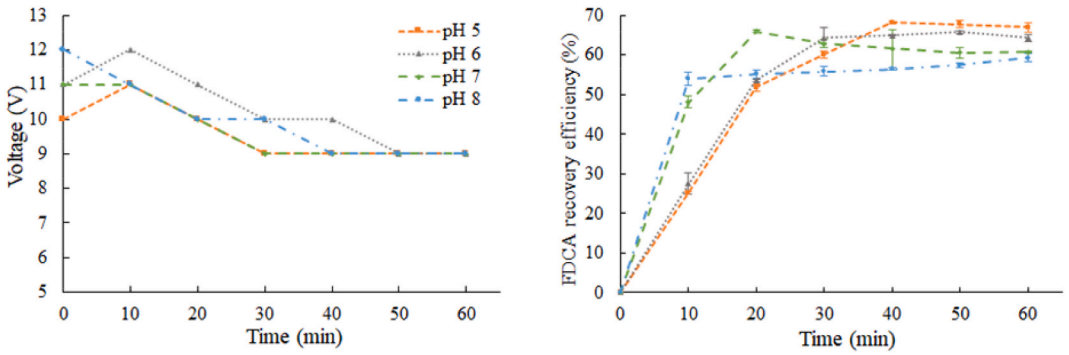

Fig. 4. The variations of the voltage and FDCA recovery efficiency of the EDBM using the feedstocks with different pHs.

Within the first 10 min, the voltage of pH 5 and 6 groups slightly increased. It was speculated that 1 N sulfuric acid was added to lower the pH value, which resulted in a significant conductivity difference between the salt and acid chambers. Therefore, the voltage increased in the beginning. The voltage of the pH 7 group remained stable within the first 10 min, which was also the same as the last experimental result at a current density of 8.93 mA/cm^2. This meant experiments had reproducibility. The voltage of the pH 8 group decreased. As the experiment progressed, the voltage of each group dropped and remained at 9 V until the end of the experiment. The FDCA recovery efficiencies of the pH 5 and pH 6 groups were lower than those of pH 7 and 8 within the first 10 min. The FDCA concentration variation in both chambers was fitted with the trend of FDCA recovery efficiencies (Fig. S5). The pKa1 and pKa2 of FDCA were 2.1 and 3.4, respectively. FDCA dissociation ratio reached 0.95 when the pH was higher than 4.75 [25]. Thus, the influence of FDCA dissociation extent could be eliminated. The reason might be that the sulfate concentration in these two feedstocks was high, which competed with FDCA ion to pass through the AEM. As the experiment progressed, the FDCA recovery efficiencies of pH 5 and pH 6 groups increased significantly and were 67.0 % and 64.4 %, respectively, at the end of the experiment. The FDCA recovery efficiencies of the pH 7 and 8 groups at the end of the experiment were 60.8 % and 59.2 %, respectively. Feeding the feedstock at a higher pH could shorten the time required for FDCA recovery, but the OH^- also competed with FDCA ions to reduce the EDBM system

Table 2
The optimal conditions for FDCA recovery using the EDBM system.

Parameter	Influent FDCA conc. (mg/L)	Current density (mA/cm^2)	Feedstock pH	FDCA recovery efficiency (%)	Current efficiency (%)	Energy consumption (kWh/kg)
Influent FDCA conc.	700	4.46	6	62.2	3.06	52.6
	1400	4.46	6	64.0	6.28	30.6
	2100	4.46	6	65.6	9.78	18.2
	2800	4.46	6	40.5	7.70	13.8
Current density	2100	4.46	6	63.2	8.53	14.1
	2100	8.93	6	68.5	4.47	30.6
	2100	13.29	6	53.2	2.73	35.1
	2100	17.86	6	51.0	2.16	48.3
Feedstock pH	2100	8.93	5	67.0	4.80	23.1
	2100	8.93	6	64.4	4.88	22.7
	2100	8.93	7	60.8	4.72	17.8
	2100	8.93	8	59.2	4.63	18.9

performance. Szczygiełda et al. explored the influence of different pHs on the α-ketoglutaric acid recovery. The results indicated that OH^- competed with α-ketoglutarate anions, which reduced the α-ketoglutaric acid recovery efficiency of the EDBM system [16].

The optimal conditions for FDCA recovery using the EDBM system are summarized in Table 2. When the influent FDCA concentration increased from 700 mg/L to 2100 mg/L, the FDCA recovery efficiency and current efficiency significantly improved, and the energy consumption was reduced. Due to the increase in current density, the current efficiency decreased, and the energy consumption increased. When the current density increased to 8.93 mA/cm^2, FDCA recovery efficiency slightly improved and was 68.5 %. The current efficiency dropped to 4.6 %, and the energy consumption enhanced to 30.6 kWh/kg. Increasing feedstock pH led to declining FDCA recovery efficiency but did not affect current efficiency. Acidic feedstock had higher energy consumption than neutral and alkaline feedstock. Compared to the published research, the current efficiency obtained in this study was quite low. There are three possible reasons. First, the biotransformed FDCA concentration in this study was much lower than the published fermented organic acid concentrations, such as lactic acid [26] and malic acid [27]. The second reason was speculated to be the complex composition of the artificial FDCA effluent. Except for FDCA, the effluent also contained several mineral salts mentioned in section 2.1. These salts dissociated into cations and anions. Liu et al. investigated the effects of inorganic ions on the transfer of weak organic acids and their salts in an electrodialysis system. They found the dehydration of organics by inorganic ions played an important role in the electrodialysis process. The migration flux of organics with inorganic ions follows the order of Mg^{2+}>Ca^{2+}>K^+>Na^+, SO_4^{2-}> Cl^- [28]. This implied that the inorganic ions (such as Na_2SO_4, KH_2PO_4, and NH_4Cl) in the effluent might affect FDCA electronic migration during EDBM operation. Besides, the molecule weight, steric structure, and ionization also influenced the migration of organics in the ED system [28]. The distribution curve of protonation species of FDCA is shown in Fig. S1. When FDCA anions migrated from the salt compartment into the acid compartment. The pH value declined from 6 to <2. This led to different protonic FDCA species co-existing and influenced current efficiency. By reviewing Table 2, it was found that the FDCA recovery efficiency could not be higher than 70 %. Low pH in the acid chamber was expected during EDBM system operation [29]. The pH variations in the acid compartment when various FDCA concentrations were fed as the feedstock are shown in Fig. S2. The literature indicated that FDCA could precipitate under highly acidic conditions (pH < 1) [30]. The pH value in the acid compartment was between 0.59 and 1.85. The pH value was sometimes below 1 and might result in FDCA precipitation. Besides, a little white powder on the AEM surface and at the bottom of the acid compartment was sometimes observed when the EDBM system was disassembled for cleaning (data not shown). To avoid FDCA precipitation in the acid chamber, the EDBM system was replaced by a conventional ED system in the following experiments to recover and concentrate FDCA, hoping to obtain a higher FDCA recovery efficiency.

3.2. FDCA recovery and concentration using the ED system with the artificial bioreactor effluent as feedstock

3.2.1. The effect of feedstock volume

Increasing the feedstock volume of influent means increasing moles of target ions in the solution and the molar flux. However, more time is required for a larger feedstock volume to complete organic acid recovery and concentration [31]. To concentrate FDCA, the effect of feedstock volumes (0.5, 1.0, and 1.5 L) on the FDCA concentration by the ED system was investigated. Fig. 5 illustrates the voltage and FDCA recovery efficiency variations using the ED system as different feedstock volumes as the influent. The curve pattern of each volume group was similar. The initial voltage of each volume group was between 9 and 11 V. As the ED system operated, the anions and cations in the diluted chamber migrated into the concentrated chamber through the anion and cation exchange membranes, respectively. This caused the decline of the diluted chamber conductivity but an increase in the concentrated chamber conductivity. Thus, the voltage gradually rose because of the significant conductive difference between chambers. The larger feedstock volume had higher ionic numbers, which extended the rising voltage time. In the artificial effluent, anions Cl^- HPO_4^{2-}. $H_2PO_4^-$, and SO_4^{2-} coexisted with FDCA anions. These inorganic anions had smaller molecule weights and might migrated faster than FDCA anions. As for the ability of these inorganics to dehydrate FDCA to decrease FDCA radii, further investigation is needed. The FDCA recovery efficiency attended 53.7 % within 15 min as the influent volume of 0.5 L. FDCA recovery efficiencies were 66.3 % and 75.5 % within 30 and 40 min, when feedstock volumes were 1.0 and 1.5 L, respectively. Table 3 shows the initial and final FDCA concentrations in the dilute and concentrated chambers and concentrated fold when the influent feedstock with different volumes. It could be observed that

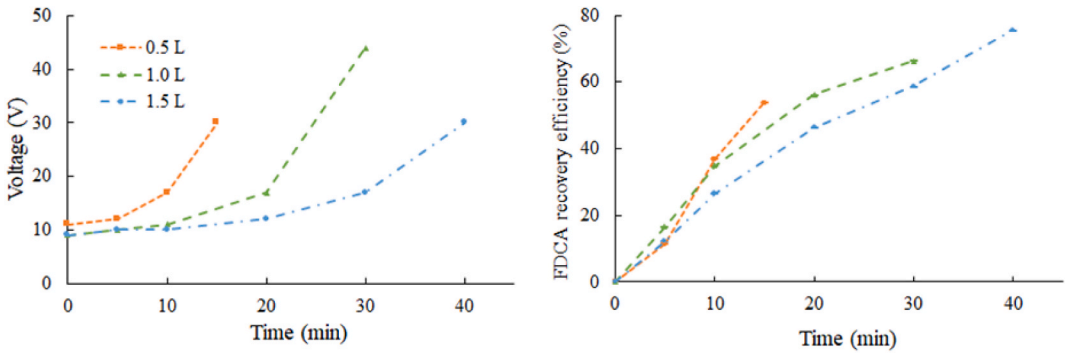

Fig. 5. The voltage and FDCA recovery efficiency variations, using the ED system as different feedstock volumes as the influent.

Table 3
The initial and final FDCA concentrations in the dilute and concentrated chambers and concentrated fold when feeding different feedstock volumes.

Feedstock volume (L)	Initial FDCA conc. in the dilute chamber (mg/L)	Initial FDCA conc. in the concentrate chamber (mg/L)	Final FDCA conc. in the concentrate chamber (mg/L)	Concentrated fold
0.5	1978.9	1023.2	2085.4	2.0 ×
1.0	2048.4	1096.5	3812.7	3.5 ×
1.5	2070.4	1075.9	5767.4	5.4 ×

increasing the feedstock volume could achieve FDCA concentration. The concentrated folds were 2.04, 3.48, and 5.36 in the order of low to high feedstock volumes.

3.2.2. *The effect of FDCA concentration in the concentrate chamber*

The organic acid concentration in the concentrated chamber affected the ED system performance. The low organic acid concentration in the concentrated chamber needed more energy to overcome the high resistance of the ED system. On the contrary, high organic acid concentration in the concentrated chamber caused reverse diffusion of organic acid and reduced organic acid recovery efficiency [16]. Thus, different FDCA concentrations (500, 1000, 1500, and 2100 mg/L) in the concentrated chamber were studied. The feedstock volume of the diluted chamber was 1.5 L, and the FDCA concentration was 2100 mg/L. According to the previous results, the feedstock pH and current density were 5 and 8.93 mA/cm^2, respectively. The variations of the ED voltage and FDCA recovery efficiency of the ED system with the different initial FDCA concentrations in the concentrated chamber are shown in Fig. 6. The higher the FDCA concentration in the concentrated chamber, the longer the operating time of the ED system. The initial voltage of each concentration group was similar, and between 6 and 9 V. After operation, the final voltages were 13, 28, 28, and 29 V in the order of low to high FDCA concentration in the concentrated chamber. As for FDCA recovery efficiency, the results could be divided into low concentrations (500 and 1000 mg/L) and high concentrations (1500 and 2100 mg/L) according to the curve trends. The FDCA recovery efficiency of the low FDCA concentrations had a similar trend. At the end of the experiment, the FDCA recovery efficiencies of 500 mg/L and 1000 mg/L FDCA groups were 78.3 % and 81.5 %, respectively. The FDCA recovery efficiencies of 1500 mg/L and 2100 mg/L FDCA group were 72.6 % and 75.9 %, respectively, at the end of the experiment. The previous research indicated that the high acid concentration in the concentrated chamber might cause back diffusion of the acid ions to reduce the efficiency of the ED system [20], which is consistent with our results.

Combining the results of the EDBM and ED systems, the optimal condition for FDCA recovery and concentration was summarized as the FDCA inflow concentration of 2100 mg/L, current density of 8.93 mA/cm^2, feedstock pH of 5, feedstock volume of 1.5 L and the initial FDCA concentration in the concentrated chamber of 1000 mg/L. These parameters were used for FDCA recovery and concentration from the real 5-HMF biotransforming bioreactor effluent.

3.3. *FDCA recovery using the ED system with the real bioreactor effluent as feedstock*

To realize that the parameters established above could be used for FDCA recovery from the real 5-HMF biotransforming bioreactor effluent, three batch effluents were used as the feedstock of the ED system. Fig. 7 shows the variations in voltage and FDCA recovery efficiency of the ED system using three real bioreactor effluents as the feedstock. The voltage of each effluent gradually increased. Owing to the low FDCA concentration, the average voltage of the effluent I was lower than those of the other effluents, and the final voltage was 25 V. The initial FDCA concentrations of effluent II and III were similar, but the average voltage of effluent III was lower than that of effluent II. It was speculated that more impurities of the effluent III were removed after pre-filtrating using a 0.22 μm pore size filter. The effluent II and III voltage at the end of the experiment were 36 and 34 V, respectively. The voltage of the actual effluent was much higher than that of the artificial bioreactor effluent. The reason might be the complex composition of the actual effluent. Szczygiełda & Prochaska applied centrifugation, ultrafiltration, and nanofiltration to pre-treat the fermentation broth. Although most

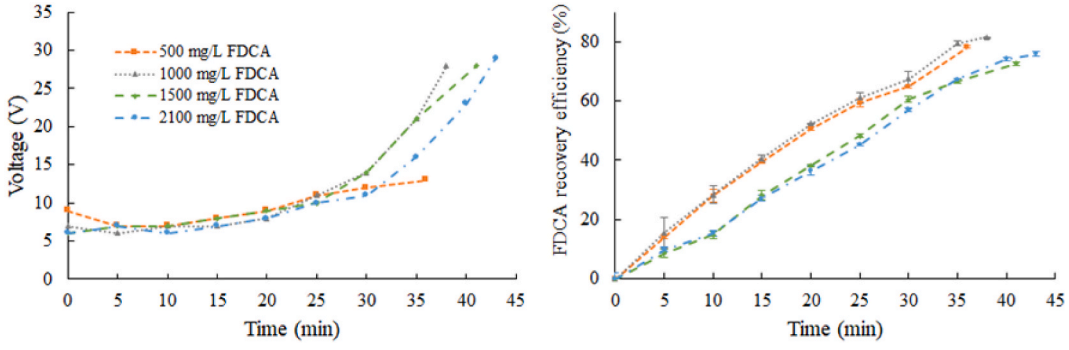

Fig. 6. The variations of the ED voltage and FDCA recovery efficiency of the ED system with the different initial FDCA concentrations in the concentrated chamber.

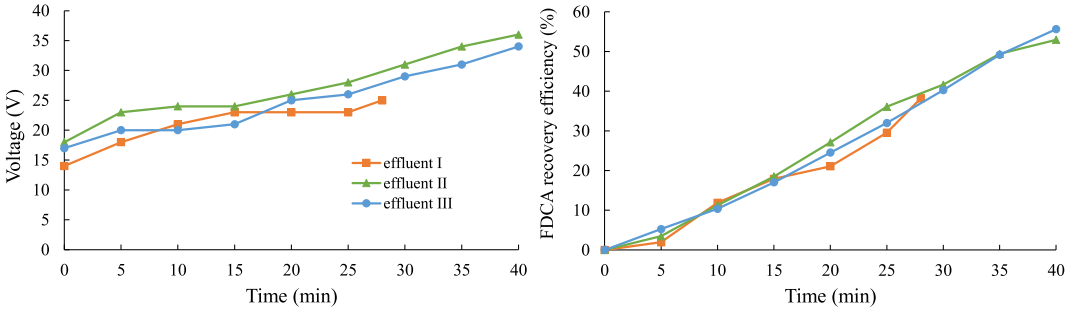

Fig. 7. The voltage and FDCA recovery efficiency variations of the ED system using three real bioreactor effluents as the feedstock.

particles were removed, some inorganic ions were left in the broth, hindering electrodialysis performance [22]. The pretreatment procedure used in this experiment was much more straightforward than in the literature. Thus, more impurities remained to increase the voltage of the ED system. In addition, the higher the viscosity of the liquid, the lower the migration rate of charged ions or colloids, resulting in a decrease in membrane filtration rate [32]. The real effluent used in the experiment had a certain viscosity, leading to an increase in resistance and voltage. The FDCA recovery efficiency of each effluent gradually increased during the whole experimental period. The FDCA recovery efficiencies in order from effluent I to III were 38.2 %, 53.0 %, and 55.6 %, respectively, which was significantly lower than the FDCA recovery efficiency of the artificial bioreactor effluent. The reason was speculated that the products produced by microorganisms were in the real effluent, which influenced the migration and recovery of FDCA. Comparing the FDCA recovery efficiencies of effluent II and III, the FDCA recovery efficiency of effluent III was slightly higher than that of effluent II by 2.6 %. This was mainly due to the different pore sizes in the pre-treatment filtration procedures.

Fig. 8 shows the FDCA flux change of effluent III and the artificial bioreactor effluent. The FDCA flux of the real effluent was lower than that of the artificial bioreactor effluent during the whole experimental period. The average FDCA flux of the actual effluent III was about 57.5 % lower than that of the artificial bioreactor effluent. Szczygiełda et al. used an EDBM system to recover succinic acid from the synthesized and real fermentation broth. When using real fermentation broth, the acid ion flux was reduced by 35 % compared with the synthesized fermentation broth. Due to the complex composition of the actual fermentation broth, the deposition of inorganic compounds and other biological components in the fermentation broth accumulated on the ion exchange membrane, resulting in a significant flux decline [21].

4. Conclusions

The optimal conditions for the EDBM system to recover FDCA were to adjust the pH of the artificial feedstock containing 2100 mg/L FDCA to 5 and operate at the current density of 8.93 mA/cm^2. The FDCA recovery efficiency was 67.0 %. The optimal conditions for the ED system to further concentrate and recover FDCA were feeding the artificial feedstock volume of 1.5 L and preparing 1000 mg/L FDCA in the concentration chamber. The highest FDCA recovery efficiency was 81.5 %, and the FDCA concentrated fold was 5.4×. The highest FDCA recovery efficiency, 55.6 %, was obtained when pre-filtrating the real effluent with a 0.22 μm pore size filter (effluent III), showing that it was feasible to concentrate and recover FDCA in the effluent of the 5-HMF biotransformation bioreactor using ED technology.

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Data availability

Data will be made available on request.

CRediT authorship contribution statement

Chih-Ming Liang: Writing – original draft. **Chun-Chin Wang:** Writing – review & editing. **Yi-Ting Hung:** Visualization, Investigation. **Hao-Wei Cheng:** Visualization. **Chu-Fang Yang:** Project administration, Funding acquisition.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Chu-Fang Yang reports financial support was provided by National Science and Technology Council (former Ministry of Science and Technology), Taiwan. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

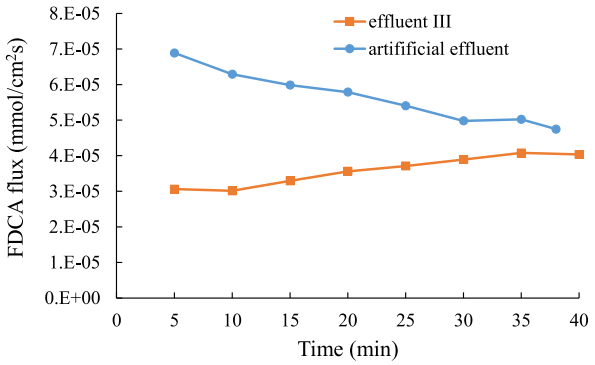

Fig. 8. The FDCA flux change of effluent III and the artificial bioreactor effluent.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2024.e34706.

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