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Preparation of succinic acid using bipolar membrane electrodialysis

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ABSTRACT

Succinic acid is a key compound in the food, chemicals and pharmaceuticals, with increasing demand in industry. To produce succinic acid in an environmental friendly manner, bipolar membrane electrodialysis (BMED) was used to convert sodium succinate into succinic acid with 0.25 mol/L sodium sulfate as electrode supporting solution. Three stack configurations, such as BP-A-C-BP, BP-C-BP and BP-A-BP, were considered to find the most cost-effective configuration. These results suggested that succinic acid could be purified in a cost-effective manner by using BMED. The voltage drop, concentration, current efficiency, and energy consumption orders of three cell configurations for succinic acid production were $U_{BP\text{-}A\text{-}C\text{-}BP} > U_{BP\text{-}A\text{-}BP} > U_{BP\text{-}C\text{-}BP}$, $C_{BP\text{-}C\text{-}BP} > C_{BP\text{-}A\text{-}C\text{-}BP} > C_{BP\text{-}A\text{-}BP}$, $\eta_{BP\text{-}A\text{-}C\text{-}BP} > \eta_{BP\text{-}C\text{-}BP} > \eta_{BP\text{-}A\text{-}BP}$ and $E_{BP\text{-}A\text{-}BP} > E_{BP\text{-}A\text{-}C\text{-}BP} > E_{BP\text{-}C\text{-}BP}$, respectively. The BP-A-C-BP configuration was found to have the high current efficiency (90%) and the low energy consumption (2.3 kW h/kg).

1. Introduction

Succinic acid ($C_4H_6O_4$) can be used in many areas such as food, chemical and pharmaceutical industries [1]. It is often called a "green" platform chemical because it represents a potential building block, which can be transformed into several commodity or specialty chemicals [2,3]. As one of the most promising commodities in the chemical industry, it currently has a market of over $0.4 billion per year and the potential to increase up to $1.3 billion per year. Currently, it is, by and large, being manufactured by catalytic hydrogenation of petrochemically derived maleic acid or maleic anhydride from n-butane [4]. Because of the scarcity of fossil resources, high crude oil prices, and the environmental benefit of using renewable feedstock, the fermentation technologies applied to the production of succinic acid have arisen as a feasible alternative to chemical processes [5,6]. However, petrochemical derived succinic acid has a lower production price ($1.05 to 1.29 US/Kg) compared with that derived by fermentation ($1.66 to 2.2 US/Kg, sold at $5–9 US/Kg). Therefore, efforts must be done to decrease processing costs in bio-based succinic acid production.

Owing to the majority of processing costs in fermentation are accounted for the separation and purification of the desired product (50–80% of the final cost), many attempts to improve and reduce separation steps have been made, but to date there is not a successful technology scaled up to industrial production. Despite the fact that several separation processes have been proposed to overcome this problem (precipitation [7], reactive extraction [8–10], ion exchange resins [11], crystallization), none addresses the main limiting barriers of large energy and material requirements as well as waste generation.

To achieve this goal, we considered bipolar membrane electrodialysis which is a novel electro-membrane process that follows the heart and soul of industrial ecology. Several studies have indicated that BMED is an economically applicable process for producing organic acids, such as citric acid [12,13], lactic acid [14–16], acetic acid [17], gluconic acid [18,19], tartaric acid [20,21], and salicylic acid [22]. Compared with conventional method-precipitation, BMED has some inherent advantages in the production of the organic acid such as high recovery, simple operation, without notorious solid pollution, environment protecting and so on. Currently, only a limited number of studies [23] on lab scale focused on the feasibility of using BMED for the preparation of succinic acid from sodium succinate. However, the production of organic acids by using BMED is not only related to operation parameters such as current density, electrolyte concentration, but, to a large extent, related to the cell configuration [24,25]. Therefore, it is necessary to find out which configuration has a better performance.

In this study experiments have been performed with the three bipolar electrodialysis configurations. Key variables such as voltage drop, succinic acid concentration and current efficiency and energy consumption have been determined.

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2. Experimental

2.1. Reagents and membranes

In this work, the homogeneous anion exchange membrane (AEM), cation exchange membrane (CEM) and the BPM-I bipolar membrane were purchased from Beijing Tingrun Membrane technology Co., Ltd., China. Their main characteristics were listed in Table 1. All the chemicals including succinic acid, sodium succinate, sodium hydroxide and sodium sulfate, which were purchased from the Chinese medicine group chemical reagent Co., Ltd., were of analytical grade and used without further purification. Distilled water was used throughout.

2.2. The experimental set-up

In this paper, laboratory-scale experimental setup was demonstrated in Fig. 1 in detail. It consisted of an acid, a salt, a base and an electrolyte reservoirs containing 1 L solution each, allowing for continuous circulation by a centrifugal pumps and flow-meters. Centrifugal pumps were used to carry the solutions from the reservoir through the electrodialysis stack. The temperature in all reservoirs was maintained at 20 °C with a recirculating cooling water system.

For the experiment, the electrodialysis stack was supplied by the Shandong Tianwei Membrane technology Co., Ltd., China. Illustrated in Fig. 2, there were three conventional bipolar membrane configurations (BP-A-C-BP, BP-A-BP and BP-C-BP configuration), which could be used to convert the sodium succinate into succinic acid. In the BP-C-BP configuration, when a direct current passed through the stack, sodium ions in the feed transported into the base compartment across the cation exchange membrane and reacted with OH^- generated by water-splitting in the intermediate layer of bipolar membrane. Simultaneously, the remained succinate ions in the feed compartment reacted with hydrogen ions to form succinic acid. In the BP-A-BP configuration, succinate ions transported through the anion-exchange membrane and combined with hydrogen ions in acid compartment. As for BP-A-C-BP configuration which contained three compartments, both succinate ions and sodium ions migrated across the anion exchange membrane and cation exchange membrane, and combined with hydrogen ions and hydroxide ions produced by bipolar membrane intermediate layer, to form the succinic acid and sodium hydroxide in acid compartment and base compartment, respectively. The sodium succinate solution with onset concentration of 0.5 M was fed to salt compartment. The effective area of the membranes was 80 cm^2. Furthermore, succinic acid solution (0.05 M) and sodium hydroxide (0.05 M) were added into the acid compartment and the base compartment to decrease the cell voltage at the beginning of the experiment, respectively. Two surface platinized electrodes made of titanium coated with ruthenium terminated the setup with sodium sulfate (0.25 M) as a rinsing solution. The feed solutions were circulated through these compartments at a flow rate of 15 L/h.

All experiments were conducted in a batch working in the constant current mode. The direct current was supplied by a CV/CC regulated power supply (WYL1702, Hangzhou Siling Electrical Instrument Ltd.). The value of current density (12.5 mA/cm^2, 25 mA/cm^2 and 37.5 mA/cm^2) used in the stack was used as the design variable during the experiments in order to quantify its impact on the preparation of succinic acid. The electrical voltage across the stack could be read from the indicators of the power supply every 10 min.

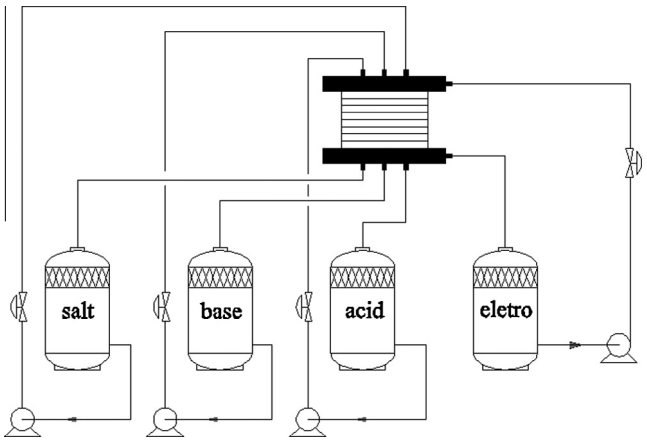

Fig. 1. Schematic diagram of the experimental set-up and flow.

2.3. Analysis methods and calculations

2.3.1. Succinic acid concentration, pH and conductivity

The concentration of succinic acid was determined by titration with a standard sodium hydroxide solution using phenolphthalein as an indicator. The pH of solutions in each compartment was continuously measured using a pH-meter (DELTA 320, Mettle–Toledo international incorporated company). And the conductivity of solutions in each compartment was continuously measured with an YSI conductivity meter (DDS-307A, Shanghai instrumental analysis scientific instrument Co., Ltd.) at the same time. Samples were taken every 0.5 hour from each reservoir. It is worth mentioning that three replicates of each experiment were performed and then their average value was reported in order to minimize the experimental errors. The circulation of solutions in the cell was started 5–10 min before the electric field was exerted.

2.3.2. Current efficiency and energy consumption

The performance of an electro-membrane process was considered in terms of current efficiency, which represents the ratio of the number of moles produced or transferred and the number of charge units passed in the system. The current efficiency ŋ was calculated as Eq. (1),

$$\eta = \frac{(C_t - C_0)ZV_tF}{Nit} \times 100\% \quad (1)$$

where C_0 and C_t are the acid concentration in acid compartment (mol/dm^3) at time 0 and t (s), respectively; V_t is the circulated volume of solution in acid compartment at time t (dm^3); Z is the ion's absolute valence ($Z = 2$); i is the current (A); F is the Faraday constant (96,485 C/mol), and N is the number of repeating units ($N = 4$).

Table 1
Main characteristics of membranes used in the experiments.

Membrane	Area resistance (Ω cm^2)	Thickness (mm)	Exchange capacity (mequiv/g)	Selectivity (%)	Heat stability (⩽°C)
AEM	5–9	0.16–0.23	1.8–2.0	90–95	40
CEM	2–5	0.16–0.23	1.8–2.2	95–99	40
BPM-I		0.16–0.23			45