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Separation of methionine from the mixture with sodium carbonate using bipolar membrane electro dialysis

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ABSTRACT

Methionine is an important amino acid can be only obtained through ingestion of particular foods. The so called hydration method is the common route for methionine preparation, which often involves many disadvantages such as environmental pollution and low methionine recovery. To avoid these problems, bipolar membrane electro dialysis (BMED) was introduced as alternative. The feasibility of BMED for methionine separation from its mixture with sodium carbonate was investigated using a series of mono-polar membranes and bipolar membranes (BM) with different cell-configurations. Membrane fouling was investigated using SEM and ATR-FTIR methods. The final results indicate that ChemJOY membranes are suitable for methionine separation with excellent anti-fouling characters. The BM-cation exchange membrane (C) is the most suitable cell-configuration. Following the success of lab-scale experiments, a pilot-scale setup was built and the results indicated that BMED could produce methionine efficiently at recovery ratio of ca. 83.23% and desalination ratio of ca. 95.18%.

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1. Introduction

Methionine is a very important amino acid for the metabolism of mammal which cannot be synthesized in vivo, and could only get in vitro to meet the normal metabolism [1]. This suggests that a sufficient supply of methionine in the diet or as a dietary supplement is of particular importance. Recently, a synthetic route that called Hydration method (a ring-closure reaction and ring-opening reaction) was applied to produce methionine from fine chemicals since the production capacity by this chemical synthesis is much larger than conventional fermentation method (see Scheme 1). However, a large amount of sodium hydroxide was consumed to promote ring-opening reaction for producing intermediate sodium methionine. Furthermore, large quantities of sulfuric acid were needed to acidify the sodium methionine into methionine at isoelectric point. These will generate enormous amount of sodium sulfate waste and acidified wastewater, and hence give rise to serious environmental problems.

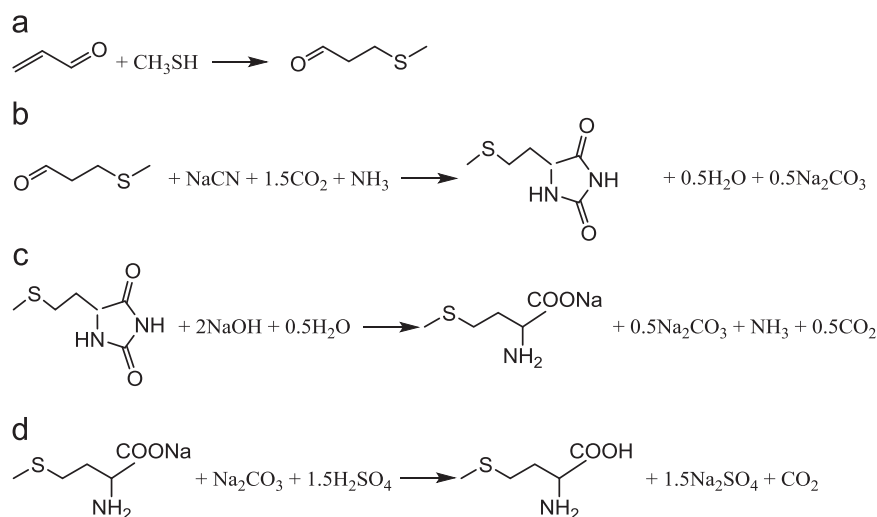
From viewpoint of environment protection and process cost, a new process of bipolar membrane electro dialysis (BMED) was suggested as an alternative to produce methionine. Since bipolar-membrane can dissociate water into protons and hydroxyl ions at the interface layer, under the driven force of potential difference.

Acid and base will be produced at corresponding compartment of BMED stack. For this case, BMED has been widely used for organic and inorganic acid/base production [2–5], environmental protection [6–9] and resources isolation [10–13], especially for the amino acid production and separation from fermenters [14–17]. However, BMED has never been used for the reclamation of methionine from its carbonate mixtures. Methionine and sodium hydroxide will be produced at acid compartment and base compartment correspondingly, when the mixture of sodium methionine and sodium carbonate solution was introduced into BMED stack. There is no need to introduce large amounts of sulfuric acid for the original synthesis route anymore. Moreover, sodium hydroxide produced during BMED process can be recycled to neutralization route and reduce the production cost and thus extra-addition of sodium hydroxide is unnecessary anymore.

Ion exchange membranes fouling is a general phenomenon during electro dialysis (ED), which is often caused by the low soluble inorganic compounds such as Ca^{2+} , Mg^{2+} and Cr^{3+} salts, organic compounds such as amino acid, macromolecular organic compounds and polymers, and even suspended solids and colloids such as poly-electrolytes, organic anions. Inorganic salts would generally form surface crystallization on membranes surface [18]. These elements mentioned above could cause severe problems to ion exchange membranes due to the precipitation at the membrane surface or partial penetration into the membranes [19]. This precipitation would results in the increment of membrane area resistance, deactivate the membrane functional group, and thus

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Scheme 1. Chemical synthesis procedure for methionine.

lead to the physical/chemical damages of the membranes. In particular, methionine anions can precipitate on the membrane surface or be absorbed into membrane matrix due to their low mobility inside membrane phase. Generally, anion exchange membranes are more sensitive to anion organics and more susceptible to be fouled during ED [20–22]. In this case, the membrane fouling was also investigated by observing morphological changes between pristine and treated membranes using ATR-FTIR and SEM.

2. Experiment

2.1. Materials

The feed mixture containing sodium methionine and sodium carbonate intermediate was kindly supplied by a related company in China. The mixture was pretreated with active carbon powder (AR, total surface area of 500–1000 m² g⁻¹, purchased from Sinopharm Chemical Reagent Co., Ltd.) at 60 °C last for 6 h. The anion exchange membranes used for BMED were JAM-II-05 (Beijing Tingrun Membrane Technology Development Co., Ltd., China), Neosepta AMX (Tokuyama Co., Japan), LabA (Hefei ChemJOY Polymer Materials Co., Ltd., China). The cation exchange membranes used were JCM-II-05 (Beijing Tingrun Membrane Technology Development Co., Ltd., China), Neosepta CMX (Tokuyama Co., Japan), LabC (Hefei ChemJOY Polymer Materials Co., Ltd., China). The bipolar membranes used were Neosepta BP-1 (Tokuyama Co., Japan) and TR BPM-I (Beijing Tingrun Membrane

Technology Development Co., Ltd., China). The main properties of the membranes were illustrated in Table 1. The used reagents including Na₂SO₄, HCl and NaOH, are AR, and purchased from Sinopharm Chemical reagent Co., Ltd., China.

2.2. Electrodialysis devices

A scheme diagram of the experiment setup is illustrated in Fig. 1. The setup is a lab-scale BMED system designed and installed in our lab. The effective area of membrane was 7.07 cm². Two repeat units were installed inside the BMED stack. Teflon plate with width of 10 mm was installed between adjacent membranes. Anode and cathode were made of titanium coated with ruthenium. The pilot-scale setup (No. CJ-BMED-1) is also designed and installed in our lab with effective area of 120 cm² (8*15) and three repeat units. Different from lab-scale setup, pilot-scale one is configured with four BMs (BPM-I) and three LabCs (BM-C configuration). Both cathodes were made of titanium coated with ruthenium. The setup consists of several separate circuits with 1 L vessel for the acid, base, feed and electrode solution. Dilute sodium hydroxide (0.1 M) was used as initial base solution and supplied to base compartment, dilute hydrochloric acid (0.1 M) was used as initial acid solution and supplied to acid compartment, sodium methionine and sodium carbonate mixture was used as feed solution and supplied to feed compartment. Sodium sulfate (3%) was used as electrolyte and supplied to electrode compartment. For lab-scale setup, each solution was circulated at flow velocity of 200 ml min⁻¹, using peristaltic pump (BT600L,

Table 1
Types of the used membrane and their properties.

	Membrane type	Thickness (μm)	IEC ^a (meq g ⁻¹)	Area resistance (Ω cm ²)	Water uptake ^b (%)	Transport number (%)
Tingrun, China, http://www.tingrun.com/	JAM-II-05	160–230	1.8–2.2	4–8 ^c	24–30	90–95
	JCM-II-05	160–230	2.0–2.9	1–3 ^c	35–43	95–99
	TR, BPM-I	160–230	–	–	35–40	–
Neosepta, Japan, http://www.astom-corp.jp/	AMX	134	1.25	2.35 ^d	16	91
	CMX	164	1.62	2.91 ^d	18	98
	BP-1	200–300	–	–	23	> 98
ChemJOY, China, http://www.cj-membrane.com/	LabA	140–160	1.5	2.5–3.5	20–30	92
	LabC	140–160	1.2	1.5–3.0	20–30	86

The data were collected from the product brochure provided by manufacturers and available in the website.

^a Anion-exchange membrane and Cation-exchange membrane IEC were determined in meq g⁻¹ in Cl⁻ and Na⁺ form respectively.

^b Water uptake/swelling degree were determined by equation of gH₂O/gdry.

^c Anion-exchange membrane and Cation-exchange membrane area resistance was measured as Cl⁻ and Na⁺ form in 0.1 M NaCl @T=25 °C, respectively.

^d Anion-exchange membrane and Cation-exchange membrane area resistance was measured as Cl⁻ and Na⁺ form in 0.5 M NaCl @T=25 °C respectively.

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