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The cleaner production of monosodium L-glutamate by resin-filled electro-membrane reactor

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ABSTRACT

In this study, electro-membrane reactors were developed for the cleaner production and *in situ* separation of monosodium L-glutamate (NaGA) from ammonium glutamate (NH_4GA) solution. The process performances were compared both before and after the addition of resins to the compartments of the reactors. The recovery rate of the resin-filled electro-membrane reactor in which all of the compartments were filled with resins was higher than that of the other reactors, while the reaction time and energy consumption were relatively low. Additionally, the effects of the current density, the initial concentration and the volumetric ratio of product solution and feed solution on the performance of the resin-filled reactor were investigated. Under the optimized conditions, 94.15% of the glutamate was successfully recovered, and the glutamate migration rate and energy consumption were 3.132 mol/(m^2 h) and 0.920 kWh/kg NaGA, respectively. The final concentration of NaGA reached 0.603 mol/L. The resin-filled electro-membrane reactor provides a novel, environmentally friendly and energy-efficient method for the production of NaGA.

1. Introduction

Monosodium L-glutamate (NaGA) is widely used as a flavor enhancer, particularly in China. The annual production of NaGA is estimated to be 2 million tons, which is nearly half of the total amino acid production [1–3]. A microbial fermentation method is typically used for its industrial production. However, to overcome the product-inhibition effect during the fermentation process, ammonia water is continuously added to the fermentation broth to maintain a constant pH. Therefore, the fermentation product is in the form of ammonium glutamate (NH_4GA) and the transformation of NH_4GA in the fermentation broth to NaGA is the critical process during the process of NaGA production [4,5]. Traditionally, this process was consisted of isoelectric point crystallization and neutralization. These additional treatments are complex and consume large amounts of acids and bases and generate large amounts of wastewater simultaneously. Specifically, the production of 1 t of NaGA consumes 0.8 t of concentrated sulfuric acid and discharges 10–15 t of highly concentrated organic wastewater [6]. In addition, a large proportion of the production cost of NaGA is associated with these recovery and purification steps. Therefore, the development of a convenient and economical approach for the production and separation of NaGA from fermentation broth is necessary.

The electro-membrane reactor is a typical electrodialysis (ED) technique that is used to produce new substances by ion recombination. Depending on the type of ion exchange membrane used, the electro-membrane reactor can be categorized as either a bipolar membrane electrodialysis (BMED) reactor or a monopolar membrane electrodialysis reactor. The former takes advantage of the H^+ and OH^- produced by water splitting in the bipolar membranes and has been used for the production of organic acids or bases from its salts [7,8]. In the monopolar membrane electrodialysis reactor, repeating units are located between two electrodes, and each repeating unit consists of four alternately arranged monopolar membranes (two cation-selective membranes and two anion-selective membranes). Two different feed solutions are introduced into the reactor, and two outputs can be recovered in the adjacent compartments under the action of a direct current (DC) electric field. The reactor can be used to perform metathesis reactions, which can be described as $AX+BY \rightarrow BX+AY$ [9]. There are two obvious advantages of the monopolar membrane electrodialysis reactor: (1) a minimum economic cost without the use of a bipolar membrane and (2) the fact that it can be used for the production of both acids/bases and salts. It has been previously demonstrated to enable the recovery of inorganic salts [10]. However, few reports can be found in the literature regarding the production of organic salts. In the present work, the electro-membrane reactor (monopolar membrane) was investigated for

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the transformation of NH_4GA in the fermentation broth to NaGA. Na_2SO_4 was introduced into the reactor as another feed solution providing Na^+. Furthermore, the ion transfer rate and recovery rate of the electro-membrane reactor during the process of organic acid production were relatively low while the energy consumption was high [11,12]. Therefore, before application, the reactor requires further optimization to achieve more efficient production.

Electrodeionization (EDI) technology is a hybrid separation process that promotes the elimination of ionized species from liquids via the use of electrically active media in the ED compartments and is mainly used to produce pure-water [13,14]. The electrically active media, such as ion exchange resins, function by alternately collecting and discharging the ionized species or by facilitating the transport of ions continuously through ionic substitution mechanisms [15,16]. According to previous reports, EDI has been used to extract lactic acid [17,18] and citric acid [19] directly from fermentation broths to avoid product-inhibition effects. Enlightened by EDI technology, the electro-membrane reactor was improved by filling resins into each compartment.

In the present work, a resin-filled electro-membrane reactor was investigated for the production of high-purity NaGA solutions, and the performances of different reactors with/without resins were compared. The influences of the current density, the initial concentration of NaGA and the volumetric ratio of feed and product solutions on the performance of the resin-filled reactor were also investigated in detail.

2. Materials and methods

2.1 Reagents

Sodium sulfate, ammonium sulfate and monosodium L-glutamate were of analytical grade and used without further purification. The NH_4GA solution was fed into the reactor instead of the real fermentation broth pretreated by filtration. Because NH_4GA is not available commercially, the NH_4GA solution was prepared by adding ammonia to a solution of L-Glutamic acid until the pH reached 6.5.

2.2 Membranes and resins

The homogeneous anion exchange membranes (AEMs) and cation exchange membranes (CEMs) used in all of the experimental setups were purchased from Beijing Tingrun Membrane Technology Co. Ltd., China. The macro-porous resins were purchased from the Chemical Plant of Nankai University, China. The main characteristics of the resins and membranes are listed in Table 1. The type of resins purchased from the manufacturer were the Na^+ and Cl^- forms. The resins were pretreated firstly according to Ref. [20] and then saturated by the corresponding solution before application to avoid the subsequent adsorption of ionized species.

Table 1
Main characteristics of membranes and resins used in the experiments.[a]

(a) Main characteristics of membranes				
Membrane	Area resistance ($\Omega\,cm^2$)	Thickness (mm)	Exchange capacity (mmol/g)	Selectivity (%)
AEM	4–8	0.16–0.23	1.8–2.2	90–95
CEM	1–3	0.16–0.23	2.0–2.9	95–99
(b) Main characteristics of resins				
Resin	Type	Functional group	Exchange capacity (mmol/mL)	Water content (%)
D001	Strong-acid	$-SO_3^-$	≥ 1.4	45–55
D296	Strong-base	$-N^+(CH_3)_3$	≥ 1.1	50–60

[a] The data were collected from the product brochures provided by the manufacturers.

Table 2
Configurations of the different reactors.[a]

Reactor	Feed compartments: Na_2SO_4 and NH_4GA	Product compartments: NAGA and $(NH_4)_2SO_4$
A	Mesh spacer	Mesh spacer
B	Mix-bed resins	Mesh spacer
C	Mesh spacer	Mix-bed resins
D	Mix-bed resins	Mix-bed resins

[a] The electrode compartments of all reactors were not filled with resins.

2.3 Experimental setup

Table 2 shows the stack configurations of various reactors. Among them, reactor A was a traditional electro-membrane reactor in which PP welded mesh spacers with a thickness of 0.9 mm were placed between the ion exchange membranes. In reactors B and C, the feed compartments and product compartments, respectively, were filled with mixed-bed resins, where spacers with a thickness of 5 mm were used for the resin filling. In reactor D, which is also defined as a resin-filled reactor, both the feed and product compartments were filled with the mixed-bed resins. The volumetric ratio of the mixed-bed resins was 6:4 (anions to cations). The effective surface area of each membrane was 158 cm^2. Both the anode and the cathode were composed of ruthenium-coated titanium.

The inner configuration of the resin-filled electro-membrane reactor (reactor D) with two repeating units is shown in Fig. 1. For the feed solutions, a Na_2SO_4 solution and a NH_4GA solution were added to the corresponding compartments. Under the effects of the DC field, the glutamate ions (GA^-) migrated from the NH_4GA compartment to the NaGA compartment through the AEM. Na^+

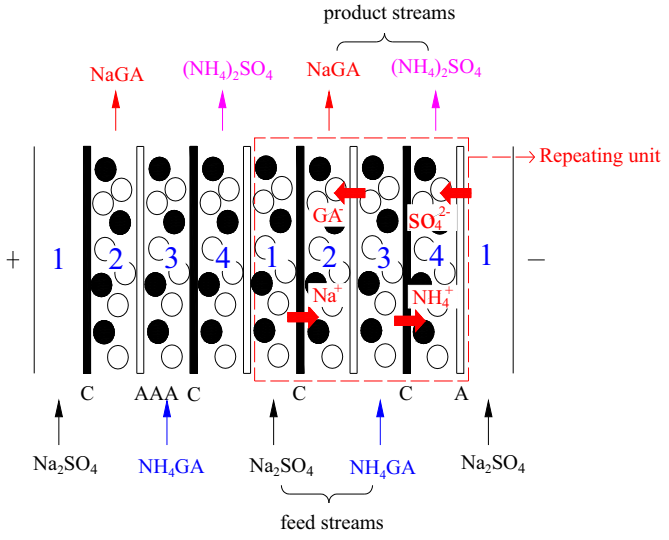

Fig. 1. Schematic representation of the resin-filled electro-membrane reactor (reactor D) for the production of NaGA. A and C represent the anion- and cation-selective membranes, respectively. 1—Na_2SO_4compartment; 2—NaGA compartment; 3—NH_4GA compartment; 4—$(NH_4)_2SO_4$compartment. ○—anion exchange resins; ●—cation exchange resins.