



## Transport properties of amino acid ions at isoelectric point in electrodialysis



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### ABSTRACT

The protonation and deprotonation reactions in systems containing amino acids play an important role and lead to some principal peculiarities of their transport in solution and membrane. A better understanding of the behavior of bipolar amino acid ions in electrodialysis would help a lot to facilitate or restrain the transfer of amino acids depending on the applications. In this work, the dissociation and migration of glutamic acid (acidic) and lysine (basic) in electrodialysis was ascertained using the special seven-compartment assemblies. It showed that acidic glutamic acid was inclined to be negatively charged and migrate towards the anode, while basic lysine preferred to be positively charged and migrate towards the cathode. Besides, the process performances of the same amino acids were also investigated during the conventional electrodialysis with the five-compartment assemblies. The pH and adsorption characteristics of membranes indicated that the above conclusion was valid for the conventional electrodialysis as well. Moreover, process parameters, i.e. overall current efficiency and energy consumption, were also assessed and compared for the electrodialysis of glutamic acid and lysine solutions.

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### 1. Introduction

Amino acids (AA) are biologically important organic compounds composed of amine ( $-\text{NH}_2$ ) and carboxyl ( $-\text{COOH}$ ) functional groups, along with a side-chain specific to each amino acid. They have been widely used in foods, beverages, pharmaceuticals, cosmetics, biodegradable plastics, chiral catalysts, and other biochemical or chemical products, and thus have a close relationship with human's daily life. Nowadays, most amino acids can be produced by fermentation at industrial scale, while the separation, concentration, and purification of amino acids is still challenging [1].

The electrodialysis technologies (ED), such as conventional electrodialysis (CED) [2,3], electrometathesis (EMT) [4], electro-ion substitution (EIS) [5], electrodialysis with bipolar membranes (BMED) [6] and electrodeionization (EDI) [4,7], have been proved their advance in the fields of amino acid production. According to their application, the two most important functions of ion-exchange membrane separation processes are demineralization and concentration of amino acids [8].

In the case of demineralization, it is better to keep the feed pH at the isoelectric point of amino acids so that most of amino acids

exist in the form of bipolar ions and stay in the feed, and more inorganic salts but less amino acids can migrate into the adjacent compartment. Shen et al. studied the desalination of glutamine fermentation broth [9]. The main ingredients of the fermentation broth were 40.0 g/L glutamine, 5.0 g/L glutamic acid, 1.8 g/L glucose, 0.42 mol/L sulfate and 0.05 mol/L of phosphates, pH 6.0. It showed that a salt removal of 95% could be achieved easily. Meanwhile, the loss of glutamine could be reduced effectively when the current density was kept at 204 A/m<sup>2</sup> and pH was controlled near the isoelectric point of glutamine (pI: 5.65) during the process. However, the minimum loss of glutamine was still as much as 20%.

In the case of concentration of amino acids, methods have to be taken to facilitate the transport of amino acids. However, the migration of amino acid ions across ion exchange membranes in the ED process is usually unsatisfactory due to their weak dissociation and complex chemical structure [10–13]. Zhang et al. [6] compared two electrodialysis processes, two-compartment BMED and modified CED, for the recovery of glutamic acid from isoelectric supernatant in monosodium glutamate production. The results indicated that the modified CED was more efficient method than two-compartment BMED. The former method gave the best performance when the operation voltage was kept at 20 V, and the initial concentrations of sulfate radical and glutamic acid were 3.5 g/L and 20 g/L, respectively; that is, the transfer rate of glutamic acid was 88.3%, the average current efficiency was 24.9%, and the

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energy consumption was 3.67 kWh/kg, which definitely need improvement.

Therefore, how to facilitate or restrain the transfer of amino acid ions in electro dialysis are of considerable importance. Although transport of amino acid ions has already been studied, the behavior of bipolar amino acid ions in electro dialysis is not yet understood in detail. In this study, the dissociation and migration direction of amino acids at their isoelectric point was ascertained using the special seven-compartment assemblies. Besides, the specific features of the amino acids recovery process by conventional electro dialysis with the five-compartment assemblies were also investigated. The objective of this study is to reveal the transport regularities of amino acids in electro dialysis, and provide the theory basis for the regulation of the transfer of amino acid ions according to the application of ion-exchange membrane separation processes.

## 2. Experimental

### 2.1. Reagent and membranes

Glutamic acid (147.13 g/mol (molecular weight), 3.22 (isoelectric point)), lysine (146.19 g/mol, 9.74), sodium sulfate and sodium chloride were of analytical grade and used without further purification. In this experiment, the anion-exchange membranes (AM) and bipolar membranes (BM) were supplied by Guangya Co. Ltd. (Hebei, China), and the cation exchange membranes (CM) were obtained from Xiangfeng Co. Ltd. (Shanghai, China). The cation- and anion-exchange membranes were converted into Na-type and Cl-type respectively by immersing the membranes into 1 mol/L NaCl solution and washed by deionized water before electro dialysis.

### 2.2. Electro dialysis equipment and membrane configurations used

Electro dialyzer supplied by Sanyuan Bada Co. (Beijing, China) was used in this study. Fig. 1 indicated two types of membrane configurations: the seven-compartment assemblies used to investigate the migration direction of amino acids, and the five-compartment assemblies for the recovery of amino acids.

The seven-compartment assemblies involved five independent streams: D4, C3, C5, D2-6 and C1-7; i.e. the flowing streams circulating in diluted compartment 4 (D4), concentrated compartment 3 (C3), concentrated compartment 5 (C5), diluted compartments 2 and 6 (D (2,6)), as well as concentrated compartments 1 and 7 (C (1,7)), as shown in Fig. 1a. Compartment 4 was fed with amino acid solution, while compartments 2 and 6 were fed with inorganic salt

solution. Thus, the migration direction of amino acids in the D4 stream can be defined by monitoring the variations of amino acid concentration in the C3 and C5 stream. Moreover, two pieces of bipolar membranes were placed on both sides of the membrane stack, which effectively prevented ions from transferring between the electrode compartments and the adjacent compartments [6]. Herein, the water dissociation induced by the bipolar membranes could make no significant effect on the pH of the concentrate, since the  $H^+$  and  $OH^-$  ions generated by the bipolar membranes in compartments 1 and 7 can be neutralized by each other in the reservoir tank (Fig. 1a). So, in total three pieces of anion-exchange membrane, three pieces of cation-exchange membrane and two pieces of bipolar membrane were used. For each membrane, the active surface area was  $80\text{ mm} \times 120\text{ mm}$ , and the spacer thickness between two membranes was 1.2 mm.

The five-compartment assemblies is based on conventional electro dialysis by adding two pieces of bipolar membranes on both sides of the membrane stack as depicted in the seven-compartment assemblies. The amino acid solution was pumped into compartments 2 and 4 as diluted stream, and distilled water pumped into compartments 1, 3 and 5 as concentrated stream; the overall result is an enrichment of ions in the concentrated stream and a depletion of ions in the diluted stream. For the five-compartment assemblies, there are two cell pairs in the stack, each containing a diluate and a concentrate compartment (Fig. 1b). So, in total two pieces of anion-exchange membrane, two pieces of cation-exchange membrane and two pieces of bipolar membrane were used. For each membrane, the active surface area was  $50\text{ mm} \times 100\text{ mm}$ , and the spacer thickness between two membranes was 1.2 mm.

### 2.3. Experimental procedure

Primarily, the migration direction of amino acids was studied. For the seven-compartment assemblies (Fig. 1a), the setup consisted of six vessels for the D4, C3, C5, D (2,6), C (1,7) and electrode rinsing solution (ERS). The initial composition of the various streams is given in Table 1a.

Second, the performance of the recovery process of amino acids in electro dialysis was examined. For the five-compartment assemblies (Fig. 1b), apart from ERS, the stack configuration was composed of two separate loops: D (2,4) and C (1,3,5); namely the flowing streams circulating in diluted compartments 2 and 4 (D (2,4)) and concentrated compartments 1, 3 and 5 (C (1,3,5)). The initial composition of the streams is given in Table 1b.

Both of the ED process with seven-compartment assemblies and that with five-compartment assemblies were carried out in

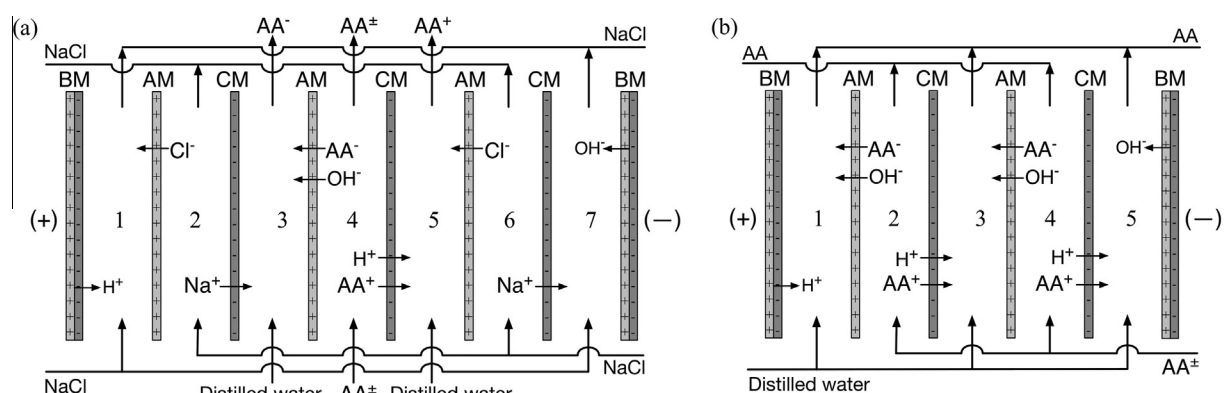


Fig. 1. The membrane configuration and principle of seven-compartment assemblies (a) and five-compartment assemblies (b) (AM, anion-exchange membrane; CM, cation-exchange membrane; BM, bipolar membrane; AA, amino acid).

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