Application of Electrodeionization Process for Bioproduct Recovery and CO₂ Capture and Storage

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ARTICLE HISTORY

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DOI: 10.2174/1385272820666160513153 326 **Abstract:** Electrodeionization (EDI) process is a relatively new process applied in environmental engineering, which combines electrodialysis and conventional ion exchange. The EDI process has obvious advantages compared with the traditional deionization equipments based on the chemical regeneration. This review first time evaluated the importance of EDI process for bioproduct recovery (organic acid recovery, bio-methane concentration and bioelectricity production) and CO₂ capture and storage. It highlighted not only the principal mechanisms but also the recent application development. Meanwhile, gaps which limited the large-scale field application of EDI process were also discussed in this paper.



Keywords: Electrodeionization; bioproduct recovery; CO₂ capture and storage; bioenergy.

1. INTRODUCTION

Biomass is an important raw material of highly profitable products including bio-based chemicals, biofuels, bio-monomers, and bio-pharmaceuticals [1]. These reliable and affordable bio-based processes and products have promotive effects on the sustainable development. The Organization for Economic Co-operation and Development (OECD) estimated that the contribution of biotechnology to the gross domestic product (GDP) of OECD could reach 2.7% in 2030 [2]. Adom et al. [3] employed the life cycle assessment technology to evaluate the role of bioproducts on the greenhouse gas emission, and the results showed that the greenhouse gas emission by bioproducts was at least a 39% decrease than that by fossil. Fig. 1 represents the possible lignocellulose utilization route for bio-based chemical production [4]. It should be pointed out that the downstream separation process often contributes most to the total cost [5]. Therefore, a reasonable separation process is necessary.

Carbon dioxide (CO₂) is considered to be the major contributor to the global warming. In order to reduce the possible environmental risk, Intergovernmental panel on climate change (IPCC) has tried to achieve a reduction of CO₂ emission by 50% in 2050 compared with that in 1990 [6, 7]. CO₂ capture and storage (CCS) technology could effectively control the CO₂ emission from the energy industry based on the carbon-intensive operational mode [8].

Many approaches for bioproducts recovery and CCS have been investigated or are being explored (as shown in Fig. 1). Among these technologies, electrodeionization (EDI) technology, also called electrochemical deionization, has attracted considerable attention because of its multi-functions and excellent compatibilities. It not only takes full advantage of the ion exchange and electrodialysis processes, but also effectively avoids the disadvantages of each process [9]. The EDI system has some obvious advantages compared with the traditional deionization equipments with the chemical regeneration: i) The traditional regeneration technology is not required in the EDI system, and the production of related harmful chemicals-acid and caustic can be avoided, improving the workplace health and safety conditions; ii) It generates products with constant quality over time, whereas the quality of products degrades as the resin exhaustion in other regenerable deionization; iii) It reduces the process cost by using a little electricity to take place of labor and reagents [10].

This review evaluated the importance of EDI process for bioproduct recovery and CCS. It highlighted not only the principal mechanisms, but also the recent development in EDI application. Meanwhile, gaps which limited the large-scale field application of EDI process were also discussed in this paper.

2. BACKGROUND OF EDI PROCESS

2.1. EDI Evolution

Table 1 depicts the historical evolution of EDI technology [11]. The EDI process was first time put forward in mid-1950s, and was then employed to produce high purity water in 1971. After that, various novel membranes and other processes such as UV were introduced to EDI system. The first commercial EDI process as thin cell was produced by Minipore in 1986. Nowadays, the development of EDI process is further growing. The major application includes the removal and recovery of heavy metals and organic acids, the production of high pure water, and so on [12-15].

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Fig. (1). Schematic diagram of lignocellulose substrate utilization for bio-based chemical production. Yellow grid within building-block chemicals represents fermentation product, while light-blue grid represents chemical product [4].

Table 1.	Electrodeionization	evolution	[11].
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Year	Researcher	Development	Company
1953	Kollsman	CEDI: device for acetone purification	Dutch company
1955	Walters et al.	Electrodeionization: concentration of radioactive waste	Argonne National Lab.
1960	Sammons and Watts	EDI for sodium salts deionization	Harwell Atomic Energy Authority
1971	Matejka	EDI for continuous high-purity water production	NA
1986	Giuffrida, Jha, Ganzi.	Electrodeionization apparatus	Millipore Co.
1989	Parsi	EDI with bipolar membranes for dissolved solids removal	Ionics Inc.
1991	Katz, Elyanow, Sims	Electrodeionization polarity reversal apparatus and process	Ionics Inc.
1992	White	EDI and ultraviolet light for water purification	Millipore Co.
1999	DiMascio, Gary, Ganzy	Electrodeionization apparatus and method	United States Filter Co.
2005	Avijit, Gareth	Continuous EDI	Chemitreat Pte. Ltd.
2008	Barber	Introduction of shifting current	General Electric Company
2012	Riviello	Simultaneous anion and cation suppression in a continuous EDI	Trovion Singapore Pte. Ltd., Co.

2.2. Construction of EDI Model

As shown in Figure 2, the key components of EDI process are ion exchange membranes, which create different compartments for liquid flow with inlets and outlets (dilute compartment, concentrate compartment and/or electrode compartment) [16]. Meanwhile, the different kinds of electrically active media, including ion exchange resin, ion exchange textile and so on, are filled in the dilute compartment to enhance mass transfer and reduce the electrical resistivity. Furthermore, the external source was chosen to apply a transverse DC electrical field. The typical electrically active media, membranes, and electrodes used in EDI systems were systematically summarized by Alvarado *et al.* [11].

It is reported that high selectivity of the electrically active media is not required, because contaminants may not migrate through the dilute compartments easily if the contaminants is held too tightly by the electrically active media. Meanwhile, high adsorption



Fig. (2). Basic electrodeionization cell configuration [16]. (CM: cation exchange membrane; AM: anion exchange membrane).

capacity is also not required, since the contaminants don't need to remain fixed on the electrically active media [17]. The most important feature of electrically active media is the ionic transport properties, which determine the EDI performances [18]. Ion exchange resin is more widely used as electrically active media in the EDI process nowadays.

Several researches added the ion exchange resins in concentrate and/or electrode compartments [18-20]. Some results showed that utilizing ion exchange resins to the concentrate and/or electrode compartment could reduce the overall electrical resistance, which helped to avoid the need for salt injection or recirculation pumps, and then reduced system complexity and operating cost [21]. However, Semmens *et al.* [22] showed that the use of ion exchange resins in the concentrate compartment was not beneficial when operating on high conductivity feed solution.

The novel EDI processes with various configurations have been developed in last few years. Besides the most common EDI process with unipolar membranes (as shown in Fig. **3a**), Grabowski *et al.* [19, 20] improved the EDI process with bipolar membranes (Fig. **3b**), and Dermentzis *et al.* [23-25] constructed the electrostatic

shielding zones, which was made of electronically and ionically conducting media such as electrode graphite powder (Fig. **3c**). These proposed processes showed predominant advantages compared with the classical EDI process, reducing the risk of concentration polarization and membrane fouling.

The most widely used EDI process arranges the ion exchange resins either as a mixed-bed, as a clustered bed, as a layered bed, or as a separated bed [20, 26]. Meanwhile, there are two major module configurations of EDI device: plate-and-frame type and spiralwound type. Currently, more than 90% of the EDI processes are arranged with the plate-and-frame mode, which guarantees the equal distribution of water flow and current in the system [10, 27].

According to the space between the ion exchange membranes, the cells generally are divided into two categories, thin cell (1.5-3.5 mm) and thick cell (8-10 mm). Filling the separated regions with the resins of same polarity will favor the performances of thick cells, while the performances of thin cells are better when anion and cation exchange resins are mixed intimately in the dilute compartments.

2.3. Operating Principle of EDI Model

Two distinct operational regimes, enhanced transfer and electro-regeneration, are considered as the operating principle of EDI process [28]. The conductivity of ion exchange resins is two to three orders of magnitude higher than that of water. Therefore, ion exchange resins in the salt forms are the top priority as the medium when ions transfer from the dilute compartment to the concentration compartment in the enhanced transfer regime [29]. In the electroregeneration regime, the voltage gradient increases with the regional ionic depletion. When the voltage gradient is higher than 0.83 V, water is dissociated to hydrogen and hydroxide ions, and then the ion exchange resins are continuously regenerated [26]. The regenerated resins in the EDI process can be continuously remove the weakly ionized compounds [10]. The extent of water dissociation was a function not only of the water purity, but also of the applied voltage and flow velocity [30].

The water dissociation preferentially occurs at the bipolar interfaces of resin/resin and resin/membrane in ion-depleting compartment. The position of water dissociation can be simplified into four



Fig. (3). Schematic diagram of EDI process with unipolar membranes (a), with bipolar membranes (b), and with electrostatic shielding zones (c). (CM: cation exchange membrane; AM: anion exchange membrane; BM: bipolar exchange membrane; IER: ion exchange resin; ICS: ion conducting separator; EICM: electronically and ionically conducting media).



Fig. (4). Schematic representation of the contact forms of ion exchange resins and ion exchange membranes in the dilute compartment [31].

Table 2. A	list of	using	EDI	process f	or the	removal	and	recovery o	f organic ac	cids.

Technology	Process	Inhibitory Compounds	Results	References
Sequential polyelec- trolyte adsorption+ RW-EDI	Removal of enzymatic and fermenta- tion inhibitory	Acetic acid; 5-hydroxymethyl furfural (HMF); Furfural; Sulfu- ric acid	The removal efficiency of acetic acid, 5-hydroxymethyl furfural, furfural, and sulfuric acid was 77 %, 60 %, 74 %, and more than 97 %, respectively.	[15]
EDI	Production of lactic acid by <i>Pedio</i> - coccus pentosaceus	-	The lactate concentration in concentrate compartment was up to 185 g/L.	[32]
EDI (Bipolar mem- brane)	Production of tartaric acid	-	The process could produce a relative high purity of tartaric acid.	[33, 34]
RW-EDI	Removal of acidic impurities from corn stover hydrolysate liquor	Sulfuric acid; Acetic acid; Furfural; 5-hydroxymethyl furfural (HMF)	The removal efficiencies of sulfuric and acetic acids were higher than 99 % and 95 %, respectively.	[41]
EDI	Recovery of citric acid from fermen- tation broth	-	The overall current efficiency was in the range of 40-96 %.	[42]
RW-EDI	Production of butyric acid with <i>Clos-</i> tridium tyrobutyricum	Butyric acid; Acetic acid; Lactic acid	Higher concentration of acetate in the reactor was reme- tabolized to butyrate, allowing >91 % selectivity of butyric acid.	[43]
RW-EDI	Enzymatic or fermentative reaction of glucose	Gluconic acid; Lactic acid	The separation efficiency of the pure gluconic acid or pure lactic acid was ≥99 %.	[44]

types according to the contact forms of ion exchange resins and ion exchange membranes in dilute compartment. As shown in Figure 4, Type a shows that the anion exchange resin (AER) contacts with AM and cation exchange resin (CER), and CER contacts with AER and CM, respectively. On this occasion, the ions transfer fast between the AER and CER, thus leading to rapid decrease of ion concentration between the resins and subsequent water splitting. As shown in Type b, the CER contacts with AM and AER, whereas the AER comes into contact with CM and CER. Due to this kind of connection, the rapid decrease of ion concentration results in the water splitting at the AM-CER contact surface and AER-CM contact surface. Two AERs contact each other are represented in Type c, following the contact with AM and CM, respectively. Under the action of AER resins, anions transfer rapidly and water was split at the AER-CM contact surface. Conversely, when two CERs contact each other and with AEM and CEM respectively (Type d), the water will be dissociated at the AM-CER contact surface. Under this condition, cations can transfer rapidly by CERs from the dilute compartment to the concentrate compartment [31].

3. APPLICATION OF EDI PROCESS

3.1. Organic Acid Recovery

Organic acids have been extensively applied in different fields, including food, pharmaceutical, and chemical industries [32]. Some

researchers have demonstrated that the EDI process is useful for the removal and recovery of organic acids [33, 34]. Recently, research has concentrated on the EDI application in microbial production of organic acids, which not only reduces the petroleum use and greenhouse gas emission, but also increases the market for organic acids because of the economical production process [35]. However, during the microbial production process of organic acids, such as enzymatic hydrolysis and fermentation, the removal of enzymatic and fermentation inhibitory compounds is very important, because these inhibitory compounds can inhibit the microorganism growth and reduce the product yield of organic acids [15]. Several processes can be used to remove inhibitory compounds from solution such as alkali treatment [36], sulfite addition [37], activated carbon treatment [38], bioabatement by microorganisms [39], multi-component electrodialysis [40], and so on. Among them, EDI process was more excellent, which could decrease the production cost and provide an efficient pathway to scale up at a commercial scale [41]. A list of using EDI process for organic acid recovery was summarized in Table 2.

Fig. **5** shows a schematic of organic acid production, which includes three EDI routes. Firstly, due to the recalcitrant nature of some raw materials, such as lignocelluloses, a pretreatment step with dilute acid is required for the disruption of lignocellulose structure and release of polymeric sugars from the internal structure before the enzymatic hydrolysis [45]. In this step, the EDI process



Fig. (5). Schematic of production of organic acids, EDI route for pretreatment process (a), for ex situ production of organic acids (b), and for in situ production of organic acids (c).



Fig. (6). One embodiment of RW-EDI apparatus, in which the pluralities of wafers were arranged in pairs (a), and one embodiment of RW-EDI apparatus, in which each wafer included a basic portion and an acidic portion (b).

can be used as an effective method for the recovery of pretreated dilute acid. As shown in Fig. **5a**, the inhibitory compounds can be removed by the EDI process (ex situ production of organic acids), which can facilitate the subsequent cyclic process, as shown in Fig. **5b**. Meanwhile, Arora *et al.* [44] demonstrated that one-step separative bioreactor (in situ production of organic acids) provided a good platform for a continuous process with simultaneous product formation and recovery in the same localized area, where efficient product capture, separation and recovery proceeded. It not only avoided the product accumulation and inhibition, but also enhanced the transport rates without pH control (Fig. **5c**). Datta *et al.* [41] provided a promising way for sequential recovery of pretreated dilute acid (sulfuric acid) and targeted organic acid (acetic acid) from the hydrolysate liquor by adjusting the operating conditions.

3.2. Bio-methane Concentration

Natural gas (methane) has developed into a fungible form of clean energy, and its market demand is increasing [46]. In order to create the desired high methane concentration in fungible natural gas, carbon dioxide (CO₂) must be separated from the biogas produced in anaerobic digestion [47]. Current methods to separate CO_2 include the utilization of a vacuum to pull the CO₂ across a membrane and elevating the temperature of a solvent to drive the CO₂. However, these methods lead to an increase in energy cost.

Seth W. Snyder [48] developed a resin-wafer deionization (RW-EDI) process to effectively separate CO_2 and CH_4 from the biogas produced by the anaerobic digestion of biomass. Fig. **6a** represented one embodiment of RW-EDI apparatus, in which the pluralities of wafers were arranged in pairs. Each basic wafer contained a porous basic ion exchange media and a biogas inlet. The biogas inlet of RW-EDI apparatus connected with the biogas outlet of anaerobic digestion system. After the biogas passed through the basic wafer, the CO_2 in biogas was converted into bicarbonate ion and the concentrated natural gas was obtained. Each acidic wafer contained a porous acid ion exchange media and a bicarbonate inlet. They were configured to receive and convert the bicarbonate ions to CO_2 gas. Fig. **6b** represented one embodiment of RW-EDI apparatus. A vented juncture was inserted between acidic and basic por-



Fig. (7). Microbial electrodeionization cell configuration [50].

tions of the wafer, which can comprise of hollow tubes. The hollow tubes of vented juncture collected the CO₂-depleted, methaneenriched biogas, while the bicarbonate outlet passed through the spaces between the hollow tubes and entered the acidic portion of the wafer.

3.3. Bio-electricity Production

Microbial electrodeionization cell (MEC) (also called ion exchange resins coupled microbial desalination cells (MDC)) is thought as a new process for bioenergy production, wastewater treatment and production of valuable chemical substances [49]. The basic configuration of MEC is shown in Figure 7 [50]. The ion exchange resins were added in desalination chamber, and the ions transfer to two neighboring compartments. In the anode chamber, the bio-convertible substrates were oxidized to form biofilm by exoelectrogenic bacteria, with the electrons released to the circuit and protons into solution. Electrons from the anode flow to the cathode, where they combine with protons and oxygen to form water.

As shown in Figure $\mathbf{8}$, the forming process of a biofilm is similar with the growth of multicellular organism, during which the

intercellular signals adjust the growth and differentiation. The biofilm formation proceeds through the adsorption of freeswimming planktonic bacteria to a biotic or abiotic surface, and this process is reversible at first, and then irreversible. Adhesion is an important trigger for the initially physiological changes of the biofilm. With the growth and division of bacteria, molecular signals pass between the cells and provide information on cell density. This phenomenon is called quorum sensing. The extracellular polymeric substances are produced by microorganisms in a mature community to encase the microcolony. However, the planktonic cells may leave the formed biofilm to establish new biofilm. There are also signals from the collectivity, attracting new microbial species to join the consortium [51].

Shehab *et al.* [52, 53] developed an air-cathode stacked microbial electrodeionization cell (SMEDIC), made of a stack of membranes. In order to improve the performance by lessening the ohmic resistances, the spacers between membranes were packed with ion exchange resins (as shown in Fig. 9). The results demonstrated that the addition of ion exchange resins and elimination of spacer between ion exchange membranes were beneficial to enhance the desalination performance of SMEDIC. When the salt concentration was 35 g L⁻¹, the desalination efficiency was 61%-72%.

In order to improve water desalination rate, Morel et al. [54] proposed a new process, namely ion exchange resins coupled with MDC. When the NaCl concentration of influents was 2-10 g L^{-1} , the desalination rate by MDC coupled with ion exchange resins was 1.5-8.0 times of that by MDC without ion exchange resins. And the charger efficiency in the MDC with ion exchange resins has also been improved. Zuo et al. [55] studied the influence of multiple ions migration on the desalination performance of MDC packed with ion exchange resins. The results showed that the mixed anion exchange resin-packed microbial desalination cell (AR -MDC) had better performance than the mixed cation exchange resin-packed microbial desalination cell (CR -MDC) at different flow rates (as shown in Fig. 10). Competitive migration order of anions was $SO_4^{2^2}$ $> NO_3^- > Cl^-$ and that of cations was $Ca^{2+} \approx Mg^{2+} > NH_4^+ > Na^+$, which were jointly affected by molar conductivity and exchange selectivity of resins. Furthermore, higher concentration of Ca²⁺ and



Fig. (8). Formation of a biofilm [51].



Fig. (9). Schematic of bench scale SMEDIC with parallel continuously recycled flow through 5 cell pair ED stack, anion exchange membrane (AEM), and cation exchange membrane (CEM) (a), Photograph of the reactor in operation (b) [52].



Fig. (10). Cell voltage and effluent conductivity changes of mixed anion exchange resin-packed microbial desalination cell (AR-MDC) and mixed cation exchange resin-packed microbial desalination cell (CR-MDC) at different flow rates. (Red arrows represent substrate replacement. A1-effluent conductivity of AR-MDC, A2-cell voltage in AR-MDC; B1-effluent conductivity of CR-MDC, B2-cell voltage in CR-MDC).

 Mg^{2+} resulted in the decrease of electric conductivity of mixed resins and scaling on the surface of cation exchange membrane adjoined with cathode chamber. The results suggested that mixed ion exchange resin-packed MDC was more appropriate for water desalination, when the water hardness was low. Similarly, in order to improve water desalination and electricity production of a MDC at a NaCl concentration lower than 0.7 g L⁻¹, ion exchange resin was also filled in the desalination compartment of MDC by Zhang *et al.* [50]. The desalination process followed pseudo-first-order kinetics. When the initial NaCl concentrations were 700 and 100 mg L⁻¹, the kinetic constant of MDC with resins increased by 2.5 times and 3.9 times than that without resins, while the infinite concentration decreased by 6.6 times and 2.6 times, respectively. Meanwhile, the current and power densities were also improved when the MDC was packed with the ion exchange resins.

3.4. CO₂ Capture and Storage

In the process of CO_2 capture and storage (CCS), the CO_2 is isolated from industrial and energy related sources, compressed,

transported and stored underground or utilized in other processes, such as enhanced oil recovery after compression [56]. In a weight per volume basis the total carbon dioxide concentration in seawater is about 140 times as high as that in air [57]. So if cost-effective methods are successfully developed to extract CO_2 and H_2 from seawater, the products could be used in marine engineering processes, including conversion it to value added hydrocarbons through catalytic polymerization, and so on [58-61]. Among these technologies, the novel electrochemical acidification technology utilizes continuous EDI principles to recover CO_2 from seawater [62].

Fig. 11 shows the configuration of a three-compartment electrochemical acidification cell [57]. Cation-permeable membrane was used to create different compartments in the system. The migration of ions (Na⁺ and H⁺) and the separation of anions were realized by the channels on the surface of acid-functional membranes. The reactions within the electrochemical acidification cell are as Eqs (1-5):

$$HCO_{3}^{-} + H^{+} \leftrightarrow H_{2}CO_{3} \leftrightarrow H_{2}O + CO_{2}(g) \uparrow$$
(1)

Anode:
$$2H_2O \rightarrow 4H^+ + O_2 + 4e^-$$
 (2)



Fig. (11). Electrochemical acidification cell configuration [62].

Dilute: 4[NaCl] _{Seawater} + 4H⁺ \rightarrow 4Na⁺ + 4[HCl] _{Acidic Seawater} (3)

Cathode:
$$4H_2O + 4Na^+ + 4e^- \rightarrow 4NaOH + 2H_2$$
 (4)

 $\begin{array}{l} \text{Overall: } 6\text{H}_2\text{O} + 4[\text{NaCl}] \rightarrow 4\text{NaOH} + 4[\text{HCl}]_{\text{Dilute}} + 2[\text{H}_2]_{\text{Cathode}} + \\ [\text{O}_2]_{\text{anode}} \end{array} \tag{5}$

Willauer et al. [58] utilized the electrochemical acidification cell as an effective alternative to extract CO₂ from synthetic seawater. When the pH value of seawater was lower than 5.0, the degassing efficiency almost reached 100%. Meanwhile, Willauer et al. [62-64] chose natural seawater as raw material for the simultaneous recovery of CO₂ and production of H₂. The results demonstrated that about 30% of the total CO2 was collected compared with that from synthetic seawater under the similar pH, which might contribute to the different $CO_3^{2^2}/HCO_3^{-1}$ concentration ratio in two kinds of solution. Besides, the RW-EDI cell was employed by Datta et al. [65] for CO₂ capture from flue gas. The results demonstrated that the CO₂ concentration in the recovery stream was higher than 98%, which could be released at atmospheric pressure without other supplementary measures. Moreover, the effect of enzyme/catalyst on the reaction kinetics was also investigated in this paper. It was found that carbonic anhydrase enzyme has promotive effect on the kinetics when the pH was close to neutral, while the enzyme might lose activity because of the instability.

4. PERSPECTIVE AND CHALLENGES

As proven above, the EDI process inherently possesses economic and environmental benefits for bioproduct recovery and CCS. However, there are still some limiting factors and bottlenecks in practical application, and the other novel functions of EDI process need to be explored. For the recovery of organic acids and biomethane, the one-step separative bioreactor will be the research interest in the future, which not only avoids product accumulation and inhibition but also reduces cost. However, how to avoid the blocking of ion exchange membranes and resins by microorganisms and influent solution is a key technique. For CO_2 capture and storage, much more effort should be made to investigate the performance of EDI process when natural seawater was chosen as the influent. Besides, further investigation can be focused on the improvement of ion exchange resins, including producing cheap substitutes of conventional ion exchange resins, reducing possibility of resin fouling, and maintaining long-term stability. More breakthroughs should be motivated to reduce expenses of EDI process through optimizing the design of piping, power supply and control system.

CONFLICT OF INTEREST

The author(s) confirm that this article content has no conflict of interest.

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