

Supporting information for

# Retrieval of CO<sub>2</sub> from carbonate solution for its sequestration by a novel electrochemical decarbonizing and ingathering method

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### **The calculation of the EDI scaling up:**

It is highly interesting for us whether the electrochemical decarbonizing and ingathering method can be potentially put forward its application. Suggested by reviewers of the manuscript, we have made a calculation of the EDI system according experimental data in order to demonstrate its feasibility to scale up and presented it as the supporting information.

For a 100MW coal-fired power plant as example, the emission amount of CO<sub>2</sub> to be treated is about 75 metric tons per hour. If the concentration of CO<sub>2</sub> in the flue gas is at 10% and 90% of the CO<sub>2</sub> in the flue gas is captured, designed equipment should have a capacity of about 3600m<sup>3</sup>/h flow volume of the EDI process to handle. This EDI device / equipment in size is comparable to amine absorption <sup>1</sup> and far less than the engineering-scale EDI's devices because of the engineering application of the electro-deionization process (110000m<sup>3</sup>/day capacity) <sup>2</sup> already existed.

In the experiment, we addressed to treat lower concentration (around 10 mmol/L) of the absorbent. Generally, the treatment of a low concentration is supposed to be much difficult and more energy input is needed for absorption / thermal desorption

process. While in the EDI process, carbonates (dissolution of  $\text{CO}_2$ ) in lower concentration solution would be easily transmitted through a membrane and concentrated under the electric field, and thus less energy is required. Moreover, the alkaline solution was regenerated for its recycling after the  $\text{CO}_2$  gases separated. Therefore, the EDI technology is easier to be scaled-up with a high efficiency, so that EDI is not only applicable but also alternative to other technologies such as amine absorption/thermal desorption.

### **The comparison between amine absorption and EDI**

It is well-known that the thermal treatment like amine absorption requires a large amount of thermal energy, in the range of 3-4 MJ/kg- $\text{CO}_2$ , of which 28% to 42% represents the thermal desorption energy<sup>1</sup>. In EDI process, energy consumption is only 1.41 MJ/kg- $\text{CO}_2$  with comparable recovery efficiency (90%). Similar to amine absorption, alkaline solution (NaOH) in the EDI method will be electrochemically regenerated and recycled. The capacity of treatment will be depended on the absorbents' flow volume instead of the device itself. Therefore, the equipment size required can be comparable. Once more, it is meaningful to make the comparison within "desorption stage" between this technology and amine absorption because the capture stage will be the same if processing  $\text{CO}_2$  emissions from a power plant.

### **The applied voltage and its measurement:**

In this experiment, a broad voltage range is tested and about 10V voltage is used in our following sections. This applied voltage is measured across the whole EDI device and it is conceptually different from the theoretical voltage for splitting water (1.23V). This is due to ion migration and water dissociation is both effected by voltage. It is generally believed that the higher voltage accompanied by severe hydrolysis degree. At that condition, the regeneration of resin is fully regenerated and also ion removal efficiency is improved. Our selection on working voltage is on the premise that water dissociation mechanism didn't exceed ion removal mechanism in order to increase current efficiency and reduce energy consumption. 1.23V condition

is enough for splitting water but not promotes ion immigration. In order to clearly discuss the phenomena, we measured the conductivity of outlet solution at different voltage as shown in Figure S1.

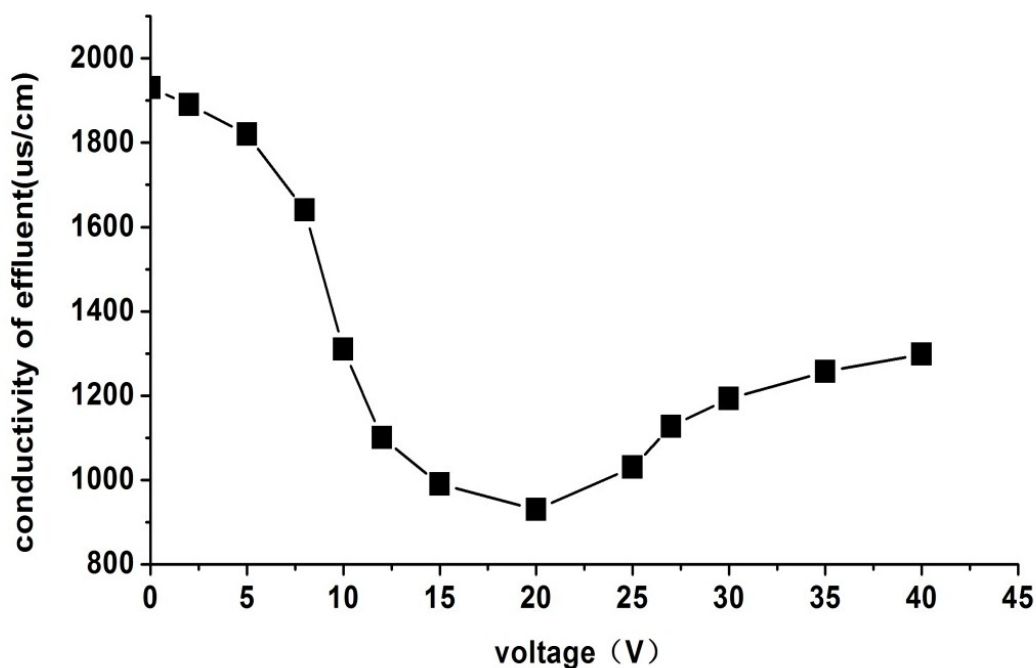


Figure S1. The conductivity of outlet solution at different voltage

From Figure S1 it clearly showed that there is an optimal condition from the conductivity of outlet solution against different voltage and implicated that at lower voltage supplies the carbonate ions can not completely migrate through the cell and higher voltage there are competitive ions due to the dissociation of water. That is, it decreased slowly in the first region, stagnated at region 3 and the eliminated flux is high in region 2. Thus we concluded that the high voltage is necessary though unavoidably water dissociation phenomenon happens.

It is deserved to be mentioned that the cell distance is 7.0 mm in the experiment which was designed as same configuration of industrial equipment and large distance may increase the voltage. As shown in Table S1, the applied voltage of each cell increase when the distance increase. The addition of resin in the cell will significantly reduce the resistance thus leading to lower voltage in comparison of an electro dialysis cell in the same distance.

In our EDI device, there are four compartments and every compartment needs about 2.5-5.0 V, and thus it is reasonable to supply around 10-20 applied voltage across the whole EDI cell.

Table S1. The effect of applied voltage in EDI system

System	Cell distance(mm)	Applied voltage of each cell (V)
ED	1.0	1-2
EDI	1.0	0.5-1.0
EDI	5.0	>2.5
EDI	7.0	2.5-5.0

#### **Limiting current:**

No current is found here as CV technique. About current limit: in an EDI device, I-V curves exhibits the full cell properties (including ions transportation across membranes and resins) so that the limiting current is different from the current limit in CV technique. Also, the I-V relations measured are attributed by the effects of ionic transport and water dissociation phenomena which are generally accepted in bipolar membrane systems<sup>3</sup>.

## **References:**

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