



## Enhanced capacitive deionization performance with carbon electrodes prepared with a modified evaporation casting method



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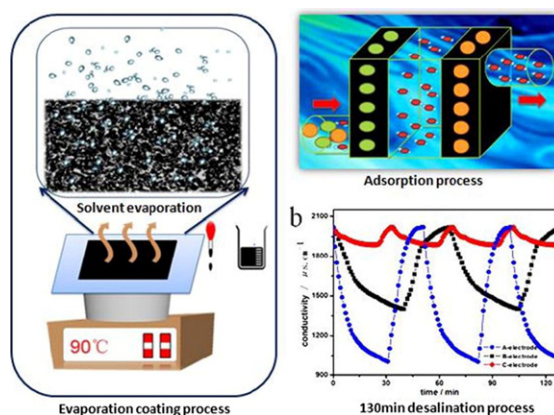
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### HIGHLIGHTS

- Electrode is prepared by evaporating the precursor mixture on a conductive substrate.
- Promising mechanical stability, porosity and hydrophilic property.
- A specific capacitance of 14.5 F/g and a desalination efficiency of 50.4%.
- CDI unit has a regeneration time of 19 min and an electrosorptive capacity of 14.4 mg/g.

### GRAPHICAL ABSTRACT



### ARTICLE INFO

#### Article history:

Received 6 November 2015

Received in revised form 29 January 2016

Accepted 4 February 2016

Available online 2 March 2016

#### Keywords:

Capacitive deionization  
Carbon electrode  
Evaporation casting  
Spray coating  
Roller coating

### ABSTRACT

A carbon electrode was fabricated with a newly developed evaporation casting method to enhance the desalination performance of a capacitive deionization (CDI) system. The surface and structural properties, as well as the electrochemical behaviors of the prepared electrode were systematically characterized by digital photo images, contact angle measurement, scanning electron microscopy (SEM) and cyclic voltammetry (CV). Capacitive deionization experiments were carried out at various operating conditions using a CDI cell with the as-prepared carbon electrodes. The specific capacitance of the carbon electrode and the desalination efficiency of the CDI cell were 14.5 F/g and 50.4% respectively, which are promisingly higher than that obtained with other carbon electrodes prepared with traditional spray coating and roller coating methods. Moreover, a regeneration time of 19 min and an electrosorption capacity of 14.4 mg/g were achieved for capacitive deionization in 25 mL NaCl aqueous solution with an initial conductivity of 2020  $\mu\text{S}/\text{cm}$  by using the new carbon electrodes. The enhanced CDI performance was attributed to the loose structure and favorable channels of the carbon electrode, which were formed in a controllable fabrication manner with the modified evaporation casting method. Therefore, the prepared electrodes by using this novel method show promising stability, high porosity, good pore distribution, and favorable hydrophilic property.

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### 1. Introduction

Capacitive deionization (CDI) has received considerable attentions since 1960s as a water treatment technology due to its simple principle and low cost [1,3,5,13,32]. Generally, when an external voltage is

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applied to a pair of highly porous electrodes, the charged ions are electrostatic adsorbed and stored on the surface of porous electrodes that consists of electrical double layer [6,38,40,44]. When the potential is removed, the adsorbed ions are released to the bulk solution. Repeating these processes can essentially purify salty water in an energy efficient and environmentally friendly way [7,8,28,42].

To assemble a practical CDI unit, Caudle et al. firstly used porous carbon electrodes made of activated carbon powder (ACP) in a flow-through mode [14]. This work was later followed by Johnson et al. that various electrode materials were thoroughly investigated [26]. However, these studies were eventually discontinued due to the instability of the fabricated electrodes. After that, tremendous efforts have been triggered by the pioneering works of Caudle and Johnson, and research has been mainly focused on fabrication of electrodes with novel materials to improve CDI performance [9,18,21–23,29]. Generally, porous electrodes with high pore volume, that enable the access of salty solution and promote the formation of electrical double layer, are substantially preferred for effective electro-adsorption of salt ions [10,12,33]. Importantly, high stability of the electrodes is a prerequisite for practical operation of a CDI cell [11,25,43]. It means that the electrode materials, such as active carbon powders, should strongly adhere to the substrate without fall off during the CDI operation. On the other hand, binder is generally used for preparation of CDI electrodes [17] while the amount of binder plays crucial role in determining the performance of the resulting electrodes. Although, the binder-free composite electrode is a promising candidate for high performing supercapacitors and capacitive deionization, the binder-free composite electrode requires complicated equipment, and is difficultly operated. Unfortunately, how to adhere the electrode active materials to a substrate in a viable manner is still a technical challenge. As known, using too much amount of binder can definitely lower the electrode conductivity due to the insulation of binder, while less content of binder can correspondingly lead to instability of the electrode [19,41]. Therefore, it is desirable to control accurately the amount of binder during the preparation process in order to balance the conductivity and stability of the resulting electrodes.

Developing rational technology of preparing electrode is substantially crucial to resolve the abovementioned problems, as well as to achieve ideal CDI performance. Indeed, several traditional methods, such as roller coating method and spray coating method, have been used to fabricate CDI electrodes, which are composed of electrode materials coated on conductive substrates, for example, carbon clothing and carbon paper. Typically, roller coating method [16,28,30] refers to pressing electrode materials onto substrate with extra pressure. This method doesn't require complicated equipment, and is easily operated. However, the prepared electrodes are prone to tightly compacted and resulting poor electrode structure since an extra pressure was valid applied during the preparation process. Spray coating method [34] refers to spraying active materials onto the substrate with the assistance of a spray gun. Although these methods have been well adopted, the CDI requests to explore a simple and low cost way with high performance application to prepare CDI electrodes. Keeping these in mind, we are here devoted to developing a novel technology of preparing CDI electrodes in a controllable and easily manipulated way. In this work, we report a modified evaporation casting method, which is simply relied on step-by evaporation the slurry of the precursor mixture on a heated conductive substrate. As a result, the prepared electrodes by using this novel method show promising stability, high porosity, good pore size distribution, and favorable hydrophilic property. Significantly, the electrodes exhibit a specific capacitance of 14.5 F/g in 0.5 M NaCl solution, which is apparently higher than that of other two electrodes obtained with traditional roller coating and spray coating methods. Furthermore, when using for CDI application, the newly developed electrodes afford an electrosorption capacity of 14.4 mg/g and fast regeneration. Thereby, this work provides an effective technology of

preparing electrodes in simple and low cost way with high CDI performance application that is possibly scale-up for water purifying.

## 2. Materials and methods

### 2.1. Materials

Activated carbon (AC, TF-B818) was purchased from Shanghai Carbon Materials Co., Ltd. Carbon fiber paper (CFP) was obtained from Beijing Ji-Xingsheng Industry and Trade Co., Ltd. Conductive black carbon was obtained from Tianjin Yiborui Chemical Co., Ltd. Ethanol (99.7 wt.%) was bought from Beijing Reagent Company. Polytetrafluoroethylene (PTFE) suspension solution (16 wt.%) was home-prepared.

### 2.2. Preparation of electrodes

#### 2.2.1. Evaporation casting of AC on CFP

First, 85 wt.% activated carbon and 10 wt.% conductive carbon black were mixed, and then the mixed carbon materials were uniformly dispersed in 30 mL of ethanol with the assistance of ultrasonication. Subsequently, 5 wt.% PTFE suspensions were added to the solution followed by ultrasonic treatment for 0.5 h. The CFP was placed on a clean horizontal glass plate which was heated by water steam. Subsequently, a certain amount of the carbon mixture solution was dropped onto the steam-heated CFP. After the solvent was fully evaporated, the carbon coated CFP electrode was cut with a size of a 5 cm × 5 cm. The obtained electrode was further dried in a vacuum oven at 60 °C for 48 h to remove all the solvent. The obtained electrode was referred as A-electrode. Fig. 1 shows the detailed steps of the preparation of the carbon coated CFP electrode.

#### 2.2.2. Roller coating method

The carbon mixed solution was prepared with the same procedure as described above. Then, same amount of the dispersion was coated and pressed with a spoon onto the CFP. The prepared electrode was dried in a vacuum oven at 60 °C for 48 h to remove all the solvent. The obtained electrode was referred as B-electrode.

#### 2.2.3. Spray coating method

The obtained carbon mixed dispersion was filled into a spray gun container. Then a certain amount of carbon was sprayed onto the CFP with a certain pressure. After that, the prepared electrode was dried in

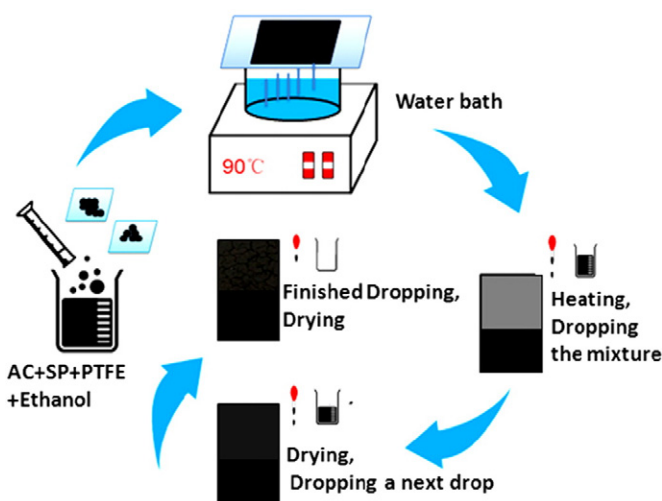


Fig. 1. Illustration of the preparation procedure of the carbon coated CFP electrode with a modified evaporation method.

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