

# An asymmetrical activated carbon electrode configuration for increased pore utilization in a membrane-assisted capacitive deionization system

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**Abstract:** A membrane-assisted capacitive deionization (CDI) system was developed for the purification of water containing sodium chloride using activated carbon fibers (ACFs) as capacitor electrode materials. The ACFs have different degrees of activation with different surface areas and pore size distributions. Their desalination performance for sodium or chloride ions was investigated. Results indicate that the salt removal efficiency and surface area-normalized electrosorption capacity for each ion depend on the surface area, pore depth and the match between the pore sizes of the ACFs and the radius of each hydrated ion. A high surface area and shallow pores favor the salt removal efficiency and a high surface area-normalized electrosorption capacity. The ACF with a median pore size of 0.69 nm performs best for sodium ion removal and those with median pore sizes of 1.09 and 1.52 nm are best for chloride ion removal, which could be ascribed to the fact that the radius of a hydrated sodium ion (0.66 nm) is smaller than that of a hydrated chloride ion (0.72 nm). An asymmetric electrode material configuration is needed to optimize both the anion and cation adsorption in the membrane-assisted CDI system.

**Key Words:** Capacitive deionization; Activated carbon fiber; Pore size; Sodium and chloride ions; Asymmetric electrode

## 1 Introduction

A desalination process that can remove some amount of salt or minerals from water has been drawn an increasing attention due to the global water crisis<sup>[1]</sup>. Most currently used methods such as thermal evaporation, reverse osmosis and electro-dialysis must overcome the high cost of facility and maintenance<sup>[2,3]</sup>. Recently, capacitive deionization (CDI), an electrochemical water purification method by reversible adsorption and desorption of ions onto the surface of electrical charged electrode under an external power source, has attracted a great deal of attention because it is an alternative low-energy-consumption and eco-friendly technology<sup>[4-8]</sup>.

In recent years, the studies have been focused on the electrode active material to improve desalination performance. Carbons with a variety of forms and porosity have been chosen as electrode active materials owing to their excellent characteristics, such as large surface area, good electrical conductivity and chemical stability<sup>[9-11]</sup>. Many studies have been carried out to improve CDI performance using various forms of carbon materials including carbon aerogels<sup>[12-14]</sup>, activated carbon fibers<sup>[15,16]</sup>, activated carbon fiber cloth<sup>[17]</sup>, carbon nanofiber networks<sup>[18]</sup>, mesoporous carbons<sup>[19,20]</sup>, graphene<sup>[21-23]</sup>, carbon nanofibers<sup>[24]</sup>, carbon nanotubes<sup>[24]</sup> and carbon-based composites<sup>[25]</sup>. However, basic investigations on

achieving a high ion removal capacity and rapid kinetics of CDI systems with a low cost have not yet been sufficiently conducted. Because ions are held through the electrical double layers on the internal surface of carbon pores, revealing the relationship between the internal structure of carbons and the size of ions is essentially important to achieve a high desalination performance. Nevertheless, a few experiments and theoretical predictions are reported about the relations between pore structure and desalination performance<sup>[26-28]</sup>.

Activated carbon fibers (ACFs), which have a large surface area and good conductivity, have been used in various fields such as catalyst supports<sup>[29]</sup>, energy storage materials and adsorbents<sup>[30]</sup>. There have been numerous efforts to elucidate the pore structure of ACFs to extend their application fields. Among the conventional ACFs, the OG-series pitch-based ACFs from Osaka Gas Co., Ltd., Japan, have high surface areas and tunable pore structures depending on their degree of activation. The structural analysis and practical use of OG-series ACFs as chemical adsorbents have been already demonstrated in many papers<sup>[31-34]</sup>.

The present study investigated the relationship between the pore structure of ACFs and desalination performance for sodium chloride aqueous solution using CDI. A powder activated carbon (Maxsorb-III) with an extremely high surface

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area was used as a counter and reference electrode materials, and four types of OG-series ACFs with predominant open pores were employed as working electrodes to study their pore structure effects on the desalination performance of sodium chloride solutions by CDI.

## 2 Experimental

### 2.1 Materials

Four types of pitch-based ACFs (OG-series: OG7A, OG10A, OG15A and OG20A with an increased the degree of activation) were obtained from Osaka Gas Co., Ltd. (Japan). The extremely high surface area activated carbon for the counter/reference electrode material (Maxsorb-III) was provided by Kansai Coke and Chemicals Co., Ltd. (Japan). Ketjen Black was selected as the conductive additive to reduce electrode resistance. Carbons were sufficiently pulverized by a milling machine (FRITSCH, Germany; with 0.2 mm sieve rings) and ball-milled in ethanol for 24 h before the all experiments. Poly (vinylidene fluoride) (PVdF, Mw ~534,000) and N, N-Dimethylacetamide (DMAc, 99.8%) were purchased from Sigma-Aldrich and sodium chloride (NaCl, 99.5%) from Junsei Chemical Co. (Japan).

### 2.2 Structural and electrochemical characteristics of ACFs

The porous structure of pulverized ACFs was evaluated by nitrogen adsorption at 77K using a BELSORP-mini adsorption apparatus. The specific surface areas were calculated by the BET (Braunauer-Emmett-Teller) equation, and their pore size distributions were derived from adsorption isotherms using an NLDFT method.<sup>35</sup> Electrochemical capacitances of ACFs were investigated by cyclic voltammetry using a conventional three-electrode electrochemical cell configuration in a 0.5 M NaCl aqueous solution. A Pt wire and a silver/silver-chloride electrode (SSCE) were used as the counter and reference electrodes, respectively. The dispersed solution of activated carbons and binder substances was dropped onto the glassy carbon as a working electrode, after which the electrode was immersed in

electrolyte and outgassed under vacuum for 10 min. Cyclic voltammogram for the assessment of carbon capacitance was performed with a potential sweep from -0.4 to 0.6 V at a scan rate from 5 to 100 mVs<sup>-1</sup> in 5 cycles. The average capacitance of samples was calculated from the 5<sup>th</sup> sweep from -0.2 to 0.4 V. The capacitances were also evaluated by the same procedure in an electrolyte (0.1 M HClO<sub>4</sub>) and with a sweep range from -0.2 to 0.8 V.

### 2.3 Capacitive deionization testing

An activated carbon paste was prepared by a mixture of 88 wt% of activated carbon, 5 wt% of Ketjen Black as conductive filler and 7 wt% of PVDF as a binder. The carbon paste was coated on the graphite foil as a current collector by a bar-coating machine with a bar height of 400 μm. The carbon electrode was dried at 80 °C in a vacuum oven for 2 h and cut into an effective area of 10 × 10 cm<sup>2</sup>. As shown in Fig. 1, the CDI unit-cells for the ion adsorption experiments were fabricated with a pair of carbon electrodes with a nylon spacer to enable streaming of the salty solution. Within the carbon electrode and spacer, ion-selective membranes (NEOSEPTA AMX/CMX membranes from ASTOM Corporation, Japan) were used to prevent adsorption on the opposite site during the desorption period to enhance the desalination performance.<sup>36</sup> An OG-series ACF electrode with a cation-permeable membrane was coupled with a negative electronic source to evaluate the adsorption performance of sodium ion in relation to the pore structure of ACFs. The counter electrode was made of an extremely high surface area activated carbon, Maxsorb-III. The opposite arrangement was used to evaluate the adsorption performance of chloride ion. The CDI experiments were conducted in a continuous flow mode. The salty water was fed from a reservoir to the CDI cell. The conductivity meter was placed at the outlet of the cell to measure the concentration of the effluent solution, purified or concentrated water. The effect of salt concentration and feeding rate of salty water on the adsorption performance was also tested. For the salt concentration tests, the 250 or 1000 ppm NaCl aqueous solution was chosen with a flow rate of

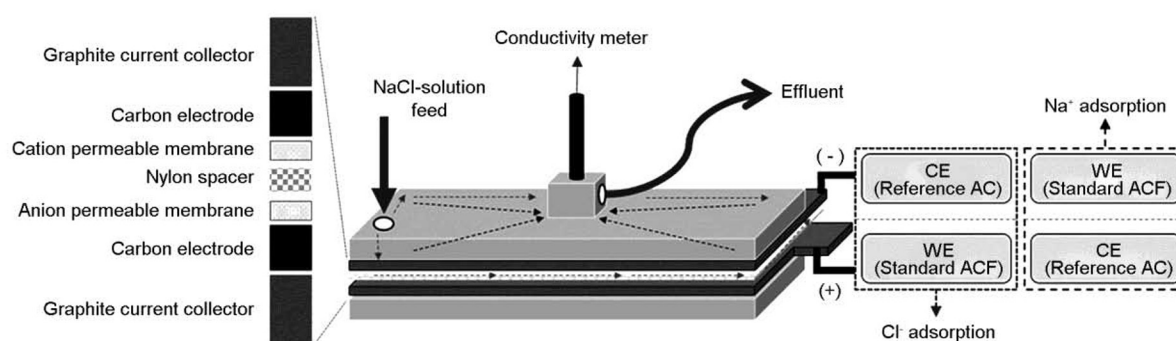


Fig. 1 Schematic of the capacitive deionization experiment.

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