



Hybrid capacitive deionization with Ag coated carbon composite electrode

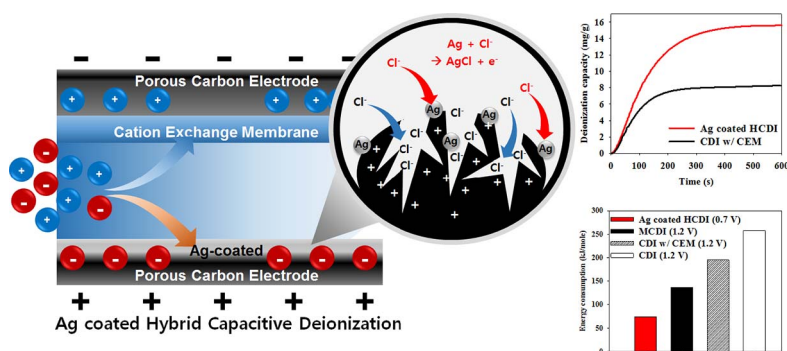
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GRAPHICAL ABSTRACT



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ABSTRACT

Capacitive deionization (CDI) using capacitive electrodes is highlighted as an alternative desalination technology because of its advantages of low cost and high energy-efficiency. However, the deionization capacity of CDI is somewhat limited because its capacity relies on the double layer capacitance of a carbon electrode. Thus, improving the deionization capacity of a CDI system is one of the most urgent issues in CDI technology. Herein, Ag coated carbon composite electrode employed hybrid CDI system (Ag coated HCDI) was investigated to enhance the deionization performances. The Ag coated carbon composite electrode was made by coating a small amount of Ag onto a carbon capacitive electrode, exhibiting the characteristics of a battery and a capacitor together. As major results, the CDI deionization capacity (88% more), rate (39% more), and charge efficiency (76% → 92%) was dramatically enhanced due to the Ag coating. The significant improvement in deionization performance is explained by the enhanced specific capacity combining the capacitance in the carbon electrode with the Ag mediated charge transfer reaction. In addition, the Ag coated HCDI (73.3 kJ mole⁻¹) is superior to membrane assisted CDI (136.7 kJ mole⁻¹) in terms of energy consumption for deionization due to its low voltage feasible operation.

1. Introduction

Capacitive deionization (CDI) is an alternative desalination technology based on electrochemical adsorption/desorption [1,2]. CDI stores ionic species in the electrical double layer (EDL) formed on the

surface of electrodes. CDI has been gaining attention for its high energy-efficiency especially in deionizing low-grade saline water [3,4]. Thus, various applications have been reported such as brackish water desalination [5], distilled water production [6], wastewater treatment [7], and hardness control [8,9].

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However, CDI still has an intrinsic limitation regarding the low capacity of the electrode because traditional CDI electrodes store ions only in the EDL at the surface of the electrodes. There have been several approaches to improve the CDI performance. For example, CDI electrode materials were developed to have a large surface and hierarchically porous structure [10–18]. In addition, novel innovative CDI systems such as membrane assisted CDI (MCDI), inverted-CDI and Flow-CDI have been introduced [7,19–23].

Recently, hybrid capacitive deionization (HCDI) was reported opening new stage of CDI technology [24]. HCDI is an asymmetric system consisting of two different types of electrodes: a capacitive electrode from CDI and a battery electrode from a battery desalination system [25]. In the battery electrode where sodium manganese oxide, manganese dioxide-carbon composite, sodium iron pyrophosphate, silver nanoparticles, and MoS_2 have been employed [24,26–30], the ions are stored through the charge transfer reaction, which contributes to the large amount of electrical capacity. Indeed, HCDI demonstrated superior deionization capacity compared to the conventional CDI [24]. Although not in HCDI, MXene and nickel hexacyanoferrate were used as battery materials for CDI and cation intercalation desalination, respectively [31,32]. However, no researches in HCDI field have been reported using a Ag-carbon composite electrode which has both the characteristics of a battery and a capacitor as one of the electrodes in the system.

Here, new HCDI with an Ag-coated carbon composite electrode (Ag coated HCDI) is proposed where this composite electrode has the characteristics of a battery and a capacitor together (Fig. 1). The Ag coated carbon composite electrode is fabricated by coating Ag onto the capacitive carbon electrode so that it has both characteristics of a battery and a capacitor. In this study, as a battery composite material, Ag was chosen due to its anion capturing ability through the charge transfer reaction ($\text{Ag} + \text{Cl}^- \rightleftharpoons \text{AgCl} + \text{e}^-$) and electrochemical stability of Ag in an aqueous system with a large amount of electrical capacity [33]. Since Ag is an expensive rare metal, small amount of Ag was deposited onto the capacitive carbon electrode with a photo induced reduction [34,35].

2. Materials & methods

2.1. Preparation of the capacitive carbon electrode

The carbon electrode was fabricated as previously described [8,15,24]. Activated carbon (YP50, Kuraray Chemical Co, Japan), carbon black (super P, TIMCAL Graphite & Carbon, Swiss), and polymeric binder (polytetrafluoroethylene, Sigma-Aldrich, USA) were mixed to prepare the electrode. The mixture was blended and roll-pressed to make a sheet-type carbon electrode.

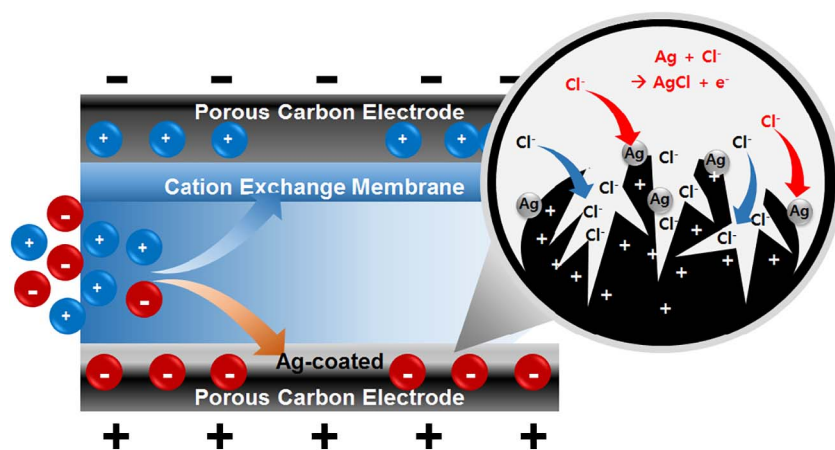


Fig. 1. Schematic of HCDI with Ag-coated carbon composite electrode (Ag coated HCDI).

2.2. Silver coated carbon composite electrode

The Ag coated carbon composite electrode was prepared by ultraviolet radiation and Ag reduction (refer to Fig. S1 for the details). The carbon electrode was radiated for 30 min with UV-C (TUV 8W G8 T5, Philips, USA) in a solution of AgNO_3 varied from 3 to 100 mM. Unless mentioned otherwise, the concentration of AgNO_3 was 50 mM. Subsequently, the electrode was immersed into 0.1 M NaOH for few seconds and was immediately rinsed with deionized water. The electrode was then dried in a vacuum oven at 120 °C.

2.3. Characterization of the Ag coated CDI carbon electrode

The surface morphologies and atomic composition of the Ag coated carbon composite electrode were obtained with a scanning electron microscope (SEM-EDX, JSM-6700F, JEOL, Japan). Pore volume and specific surface area were measured according to the Brunauer–Emmett–Teller (BET) equation (ASAP 2010, Micromeritics Instrument Corp., USA). The surface hydrophilicity of the electrode was analyzed by the sessile drop method with a contact angle analyzer (DSA 100, KRUSS, Hamburg, Germany).

Cyclic voltammetry (CV) and galvanostatic charging/discharging were measured with a potentiostat (PARSTAT 2273, Princeton Applied Research, USA) to determine the electrochemical properties of the electrodes. A characterization cell was used as previously described [8]. A pristine carbon electrode and an Ag/AgCl (sat. KCl) electrode were used as the counter electrode and the reference electrode, respectively. The scan rate (v) was set at 2 mV s^{-1} . The specific capacity (mAh g^{-1}) was measured from the galvanostatic charging/discharging profile in a 3-electrode system. The galvanostatic profile was obtained with a current density of 0.5 mA cm^{-2} . The potential range was -0.10 – 0.25 V .

In addition, the electrochemical stability of the specific capacity was evaluated in a two-electrode system. 2 M NaCl was used as an electrolyte. The current density was set at 10 mA cm^{-2} in a potential range from zero to 0.7 V.

Galvanostatic charge/discharge tests were performed, and for each cycle, the capacity retention ratio and the coulombic efficiency were calculated as follows (Eqs. (1) and (2)) [8,24]:

$$\text{Capacity retention ratio} = \frac{C_i}{C_0} \left(C_i = \frac{2I \cdot \Delta t_i}{m \cdot \Delta V_i} \right) \quad (1)$$

$$\text{Coulombic efficiency} = \frac{C_{i,d}}{C_{i,c}} \quad (2)$$

where C_i is the specific capacity at the i th cycle (mAh g^{-1}); C_0 is the specific capacity at the first cycle (mAh g^{-1}); $C_{i,c}$ and $C_{i,d}$ are the specific capacity at the charging and discharging in the i th cycle (mAh g^{-1}); I is the current (A); Δt_i is the charging time at the i th cycle

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