

## **Energy & Environmental Science**

## COMMUNICATION

Supporting Information

# A novel three-dimensional desalination system utilizing honeycomb-shaped lattice structures for flow-electrode capacitive deionization

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#### **Experimental Section**

Cordierite structures ((Mg<sub>2</sub>Al<sub>4</sub>Si<sub>5</sub>O<sub>18</sub>; 150 × 150 × 450 mm) with pore size of  $\frac{120}{30}$  µm were purchased from Ceracom (Busan, Korea), as shown in Fig. S1. They were cut to the desired sizes for 1 × 3 and 3 × 3 cells, and thoroughly rinsed with deionized water. Sodium chloride and activated carbon materials (Maxsorb MSC-30) were purchased from Sigma-Aldrich Corporation (St. Louis, USA) and Kansai Coke & Chemicals Co., Ltd. (Japan), respectively. Anionic (17 wt%, poly (phenylene oxide) in N-methyl-2-pyrrolidone) and cationic (20 wt%, poly (phenylene oxide) in dimethylacetamide) membrane solution were used to coat the channels by dripping the solutions in using a pipette. Subsequently, the samples were heated at 70 °C for 1 h in the heating oven. Then, graphene solution (20 wt% in isopropanol) was coated over the ion exchange membrane deposited channels and dried at room temperature for 24 h. Pt wire was connected to the outmost graphene layer to apply external voltage. In order to circulate flow-electrode slurry into the desalination cells, the inlets and outlets of latex tubes were inserted into graphene-coated channels and sealed perfectly with silicone adhesive. For preparation of the flow-electrodes, NaCl (0.1 M) and activated carbon (9.1 wt%) were added to deionized water and stirred for 24 h. For the batch mode desalination experiment, the prepared desalination cell was located in the middle of a hand-made polystyrene (PS) chamber. This was followed by complete sealing of both ends with silicone adhesive (Fig. S2b). The saline feed water at a concentration of 35 g/L was continuously recirculated in the PS chamber at flow rates of 1.0 and 3.0 mL/min and the flow-electrodes were circulated within the system using a peristaltic pump (Miniplus 3, Gilson Inc., USA) with the flow-rate of 10 mL/min. The cell voltage of 1.2 V was applied to the Pt wires for 100 min using a potentiostat (ZIVE SPS, Wonatech, Korea). The salt concentration of the desalinated water was obtained by measuring the conductivity (DS-70 Laqua, Horiba, Japan) of the effluent and then converting it using the NaCl concentration vs. the conductivity curve (Fig. S3). For the continuous desalination experiment using the  $3 \times 3$  cell configuration, the prepared  $3 \times 3$  desalination system was placed in the middle of the PS cell and both ends were completely closed with silicone adhesive. Concentration of electrodes and voltage potential corresponded with those used in batch mode. Then, inlet and outlet tubes were connected to the PS cell by penetrating the top and bottom of the silicon adhesives with needles. Subsequently, feed water (1.1 and 35 g/L) was pumped into the PS cell at a flow rate of 0.2 mL/min and drained through the tube located on the opposite side when the feed water completely filled the PS cell. The variation in the salt concentration was obtained from the conductivity of effluent obtained over the entire time of operation.



Fig. S1 Photographs of the pristine cordierite structures utilized in the present study.



Fig. S2 Photographs of (a) prepared capacitive deionization cell and (b) setup for the continuous mode desalination experiment.



Fig. S3 Variation in conductivity as a function of NaCl concentration measured at room temperature.



Fig. S4 Variation in current as flow-electrodes are introduced to the 3 × 3-channel cells operating in batch mode.

#### Journal Name

### Fig. S5 Energy consumption and charge efficiency in honeycomb cell.

	Energy per Ion Removed (kT)	Charge Efficiency
Honeycomb Cell (35 g/L, 3 X 3)	67.9	0.81