Supporting Information for

CDI Ragone Plot as a Functional Tool to Evaluate Desalination

Performance in Capacitive Deionization

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Detailed operating parameters for each experimental condition

Carbon	Salt concentration (mM)	Flow rate (ml/min)	Electrode thickness ^a (μm)	Current load (mA)
MSP-20	2	2	300	1, 2, 3, 4
MSP-20	10	2	300	2, 5, 9, 15, 18
MSP-20	50	2	300	2, 5, 9, 15, 20, 25
MSP-20	100	2	300	2, 5, 9, 15, 20, 25
MSP-20	10	1	300	2, 5, 9, 12
MSP-20	10	4	300	2, 5, 9, 15, 18
MSP-20	10	2	200	2, 5, 9, 15, 18
MSP-20	10	2	400	2, 5, 9, 15, 18
S-51HF	10	2	300	2, 5, 9
YS-2	10	2	300	2, 5, 9, 15, 18
CEP21	10	2	300	2, 5, 9, 15, 18
MDC	10	2	300	2, 5, 9, 15

Table S1. Detailed operating parameters for each experimental condition.

^a The numbers refer to characteristic thickness of the electrode. The real electrode thickness varied depending on

samples, but its deviation was controlled not to over 10%.

Charge efficiency depending on salt concentration

Table S2 shows deionization capacity, charge, and charge efficiency obtained at different salt concentrations in Fig. 2(a). As can be seen, both deionization capacity and charge increase as salt concentration increases up to 50 mM NaCl. However, deionization capacity of 100 mM is similar to that of 50 mM, while charge increases consistently (also see Fig. S1). As a consequence, charge efficiency (ratio of deionization capacity to charge) decreases above 50 mM NaCl; no more enhanced deionization capacity can be achieved with an increase in energy consumption. Therefore, a higher salt concentration becomes disadvantageous in terms of energy use, though deionization rate can be enhanced as electrolyte resistance decreases.

Table S2. Deionization capacity, charge, and charge efficiency depending on salt concentration.

Salt concentration (mM)	Current load (mA)	Deionization capacity (mg/g)	Charge (C/g) ^a	Charge efficiency
2	4 ^b	4.3	9.2	0.77
10	5	17.4	32.9	0.87
50	5	22.6	35.9	1.04
100	5	22.7	40.0	0.94

^a Total transferred charge from a cycler to CDI cell divided by mass of both electrodes.

^b In a 2 mM NaCl solution, CDI operation was not available above 4 mA due to its high electrolyte resistance (see Fig. S1).



Fig. S1. Representative voltage profiles of different salt concentrations obtained during charging step.

Flow rate dependent charging behavior

Fig. S2 shows representative voltage profiles depending on flow rate. As can be seen, the profiles appear to be affected by flow rate; fluctuation becomes significant as flow rate increases. In particular, the profile of 1 ml/min cannot be obtained above 12 mA because charging was finished (the voltage abruptly reached to 1.2 V) in few seconds. These unstable voltage profiles would be ascribed to instantaneous depletion of ions in a spacer channel between electrodes. In order to maintain a certain current, ions need to transport from a bulk phase (a spacer channel) to double layer of electrodes. A lower current load could enable such transport continuously because influent solution promptly replaces ion-depleted solution. As current load increases, however, the replacement becomes retarded and thus the ion-depleted solution results in rise of resistance and subsequent voltage increase. Afterwards, the depletion is compensated by the influent solution (decrease in voltage), thus the resulting voltage profile increases with fluctuation. A lower flow rate seems to worsen the supply of ions, thus the fluctuation becomes severe. This effect is well matched to the voltage profile of a low salt concentration (2 mM NaCl) in Fig. S1.



Fig. S2. Representative voltage profiles of different flow rates during charging step.

Characterizations of CEP21 and MSP-20

Fig. S3 shows representative contact angle images of CEP21 and MSP-20. To obtain these images and contact angle values, a contact angle analyzer (KRÜSS, DSA100, Germany) and DSA100 software were used. The electrode was dried prior to the measurement and installed to the analyzer. Then, 6 µL of deionized (DI) water attached at the tip of a needle was smoothly transferred to the electrode surface by direct contact and following detachment. The contact angle measurement was immediately made after the transfer in order to avoid water penetration into the electrode and evaporation. The contact angle was collected from five different points of the electrode, and the resulting averaged values were 131 ± 1 and $116\pm3^{\circ}$ for CEP21 and MSP-20, inferring that the surface of MSP-20 is more hydrophilic compared to that of CEP21. Though various factors could affect contact angle measurement, oxygen functional groups developed on the carbon would play a major role. XPS analysis confirmed that atomic O/C ratio of MSP-20 was 0.20, which is two-fold higher than that of CEP21 (0.10), indicating MSP-20 contains more oxygen functional groups. BET surface area of CEP21 and MSP-20 were quite similar (2,210 and 2,266 m²/g) as reported in our previous studies (Kim and Yoon, J. Electroanal. Chem. 704 (2013) 169-174, Yang et al., Carbon 71 (2014) 294-302).



Fig. S3. Contact angle images of (a) CEP21 and (b) MSP-20.