# Supplementary information

## Capacitive deionization of divalent cations for water softening using functionalized carbon electrodes

Zhi Yi Leong and Hui Ying Yang*

Pillar of Engineering Product Development (EPD), Singapore University of Technology and Design, 8 Somapah Road, 487372, Singapore

*E-mail: yanghuiying@sutd.edu.sg

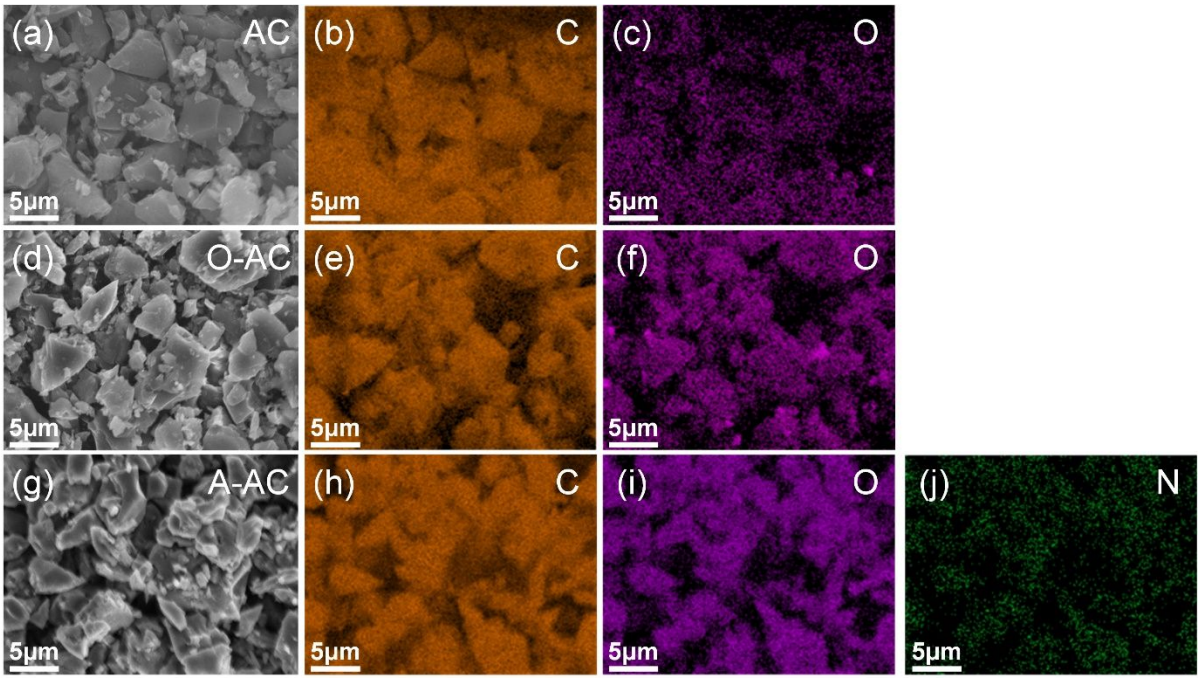


**Figure S1** SEM and EDS images of (a, b, c) AC, (d, e, f) O-AC and (g, h, i, j) A-AC.

| | B-AC (at%) | O-AC (at%) | A-AC (at%) |
|---|---|---|---|
| **C=C** | 80.5 | 93.6 | 74.54 |
| **C-C** | 17.5 | 0.73 | 8.45 |
| **C-OH, C-O-C** | 2 | 1.39 | - |
| **C=O** | - | 4.28 | - |
| **C-N** | - | - | 17.02 |

**Table. S1** Atomic percent concentration of peaks in C1s spectrum.

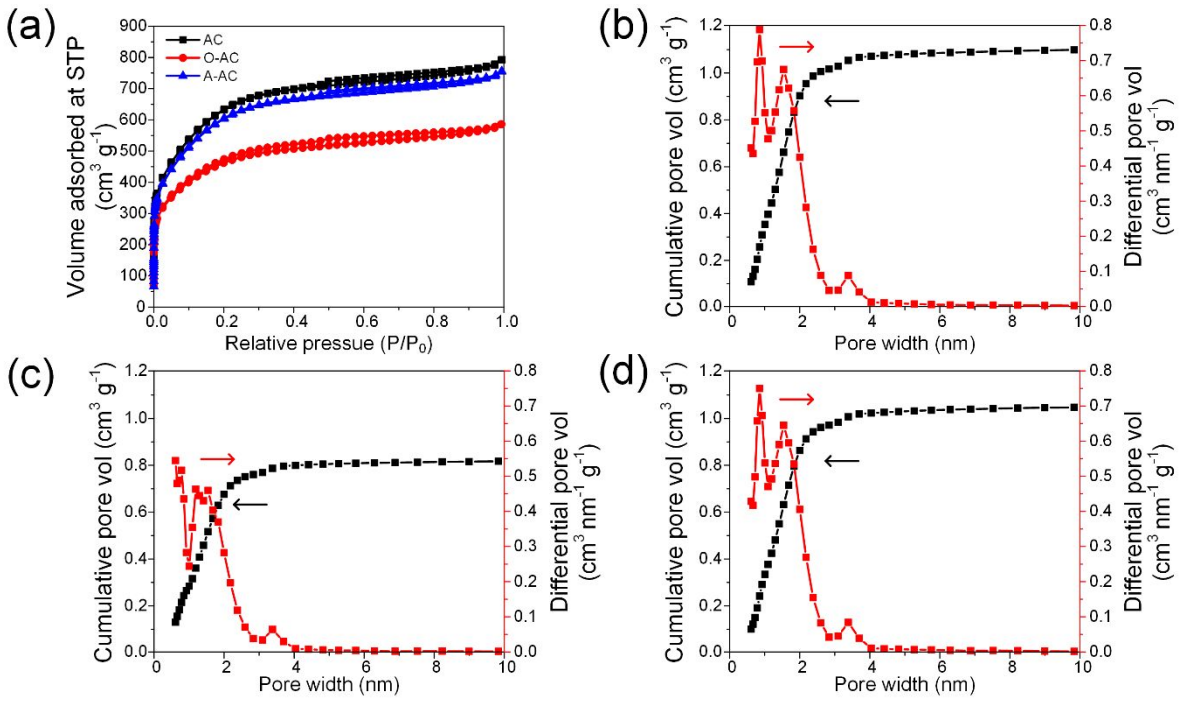


**Figure S2** (a) $N_2$ adsorption-desorption isotherms of AC, O-AC and A-AC. Cumulative pore volume and pore size distribution of (b) AC, (c) O-AC and (d) A-AC.

| Sample | SSA ($m^2$ $g^{-1}$) | Pore volume ($cm^3$ $g^{-1}$) | Total pore volume ($cm^3$ $g^{-1}$) |
|---|---|---|---|
| AC | 1862 | 1.12 | 1.23 |
| O-AC | 1503 | 0.83 | 0.91 |
| A-AC | 1768 | 1.067 | 1.17 |

**Table. S2** Summary of specific surface areas and porosity characteristics of AC, O-AC and A-AC.

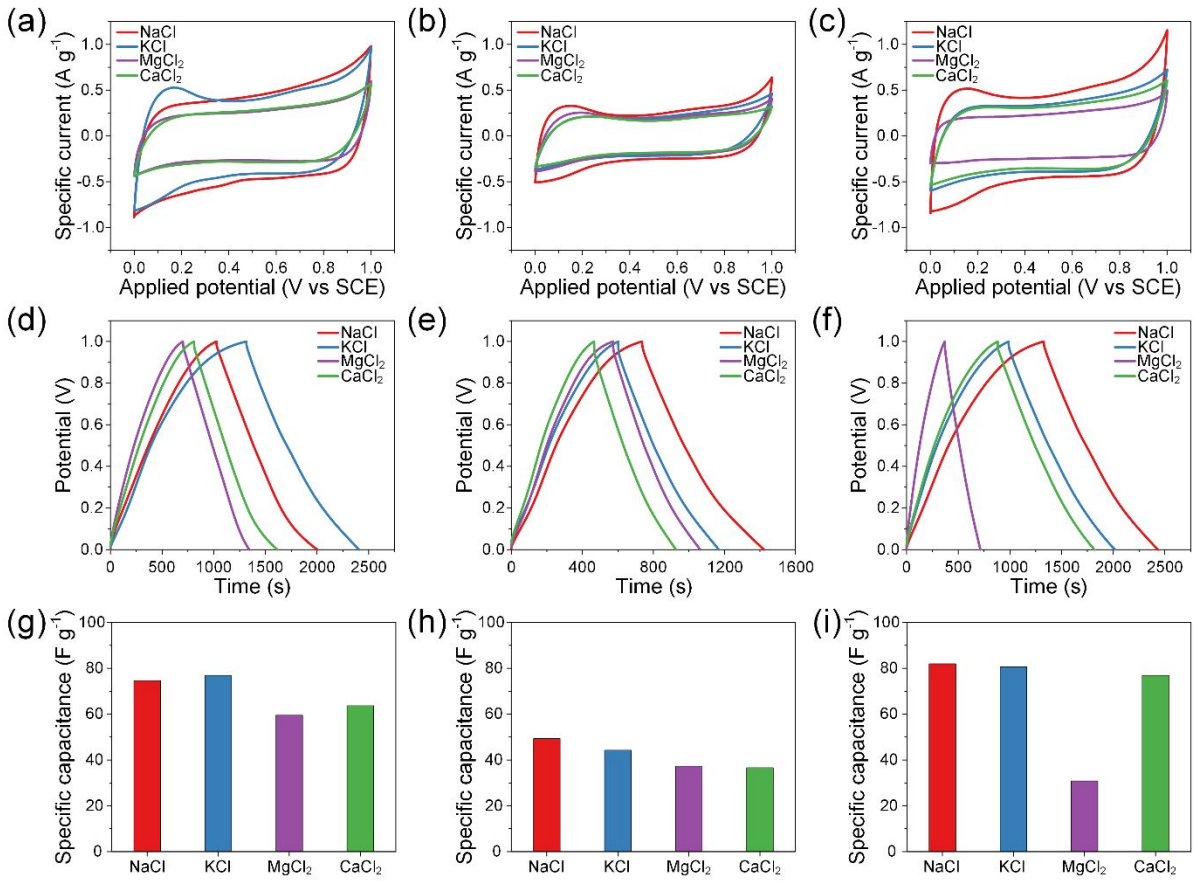


**Figure S3** CV curves of (a) AC, (b) O-AC and (c) A-AC in 0.5 M NaCl, KCl, $MgCl_2$ or $CaCl_2$ solution at 5 mV $s^{-1}$. GCD curves of (d) AC, (e) O-AC and (f) A-AC in 0.5 M NaCl, KCl, $MgCl_2$ or $CaCl_2$ solution at charge and discharge current of 0.1 A $g^{-1}$. Specific capacitances of (g) AC, (h) O-AC and (i) A-AC in different salt solutions.

**Supplementary note S1: Electrochemical characterization**

The results of our CV experiments are shown in Figure S3a – c. All samples exhibited relatively rectangular CV curves with slight peaks the ends of the potential window (0 and 1.0 V). The elevated current density is likely due to potential switching which caused some side-reactions to occur. Overall, CV curves show no significant redox peaks and the following GCD curves (Figure S3d – e) are linear which confirms the capacitive nature of the materials. According to capacitances calculated based on equation (1), O-AC possessed the least capacitance among all three samples in all solutions whereas AC and A-AC showed similar capacitances save for one in $MgCl_2$. These results correspond well with their individual surface areas which implies that ion adsorption is positively correlated to surface area. From the capacitance summary given in Figure S3g – i, monovalent salt solutions provide the largest capacitances and according to a capacitive model of adsorption, should also result in the largest adsorption capacities. Specific capacitances of AC in different salt solutions follow a decreasing order of KCl > NaCl > $CaCl_2$ > $MgCl_2$ while O-AC is NaCl > KCl > $MgCl_2$ > $CaCl_2$ and A-AC is NaCl > KCl > $CaCl_2$ > $MgCl_2$. It should be noted that electrodes fabricated for electrosorption experiments below are capacitance matched to provide the best performance.

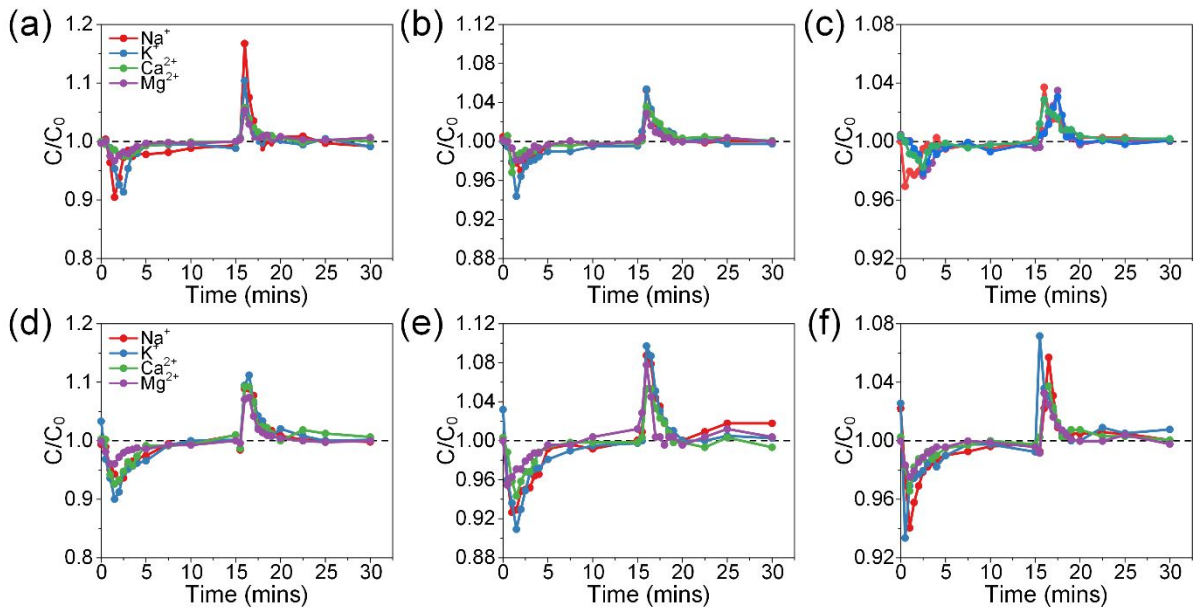


**Figure S4** Cation adsorption using AC as the cathode in (a) 2.5 mM, (b) 5 mM and (c) 10 mM single-salt solution experiments. Cation adsorption using O-AC as the cathode in (d) 2.5 mM, (e) 5 mM and (f) 10 mM single-salt solution experiments. All results were obtained during the third cycle.

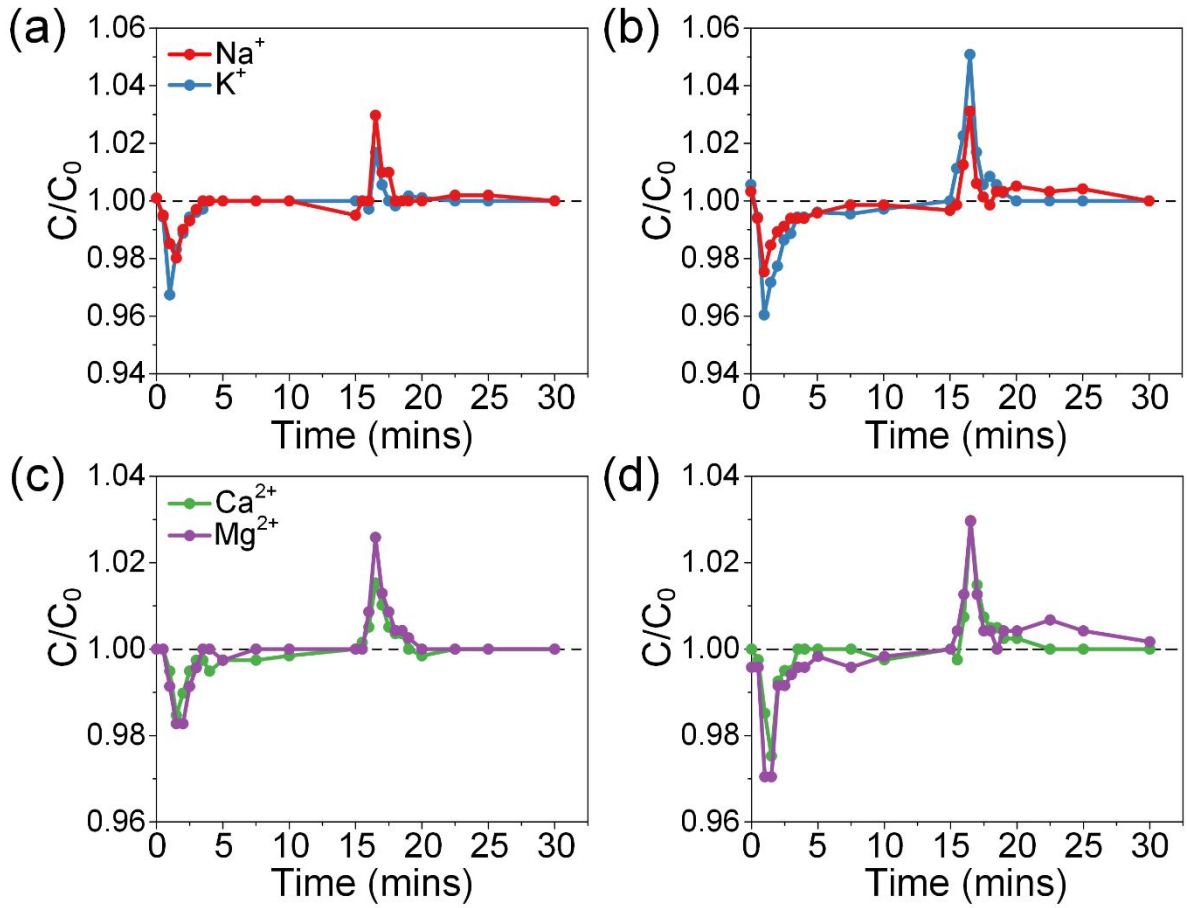


**Figure S5** Cation concentration curves of experiment 1 using either (a) AC or (b) O-AC as the cathode. Cation concentration curves of experiment 2 using either (c) AC or (d) O-AC as the cathode.

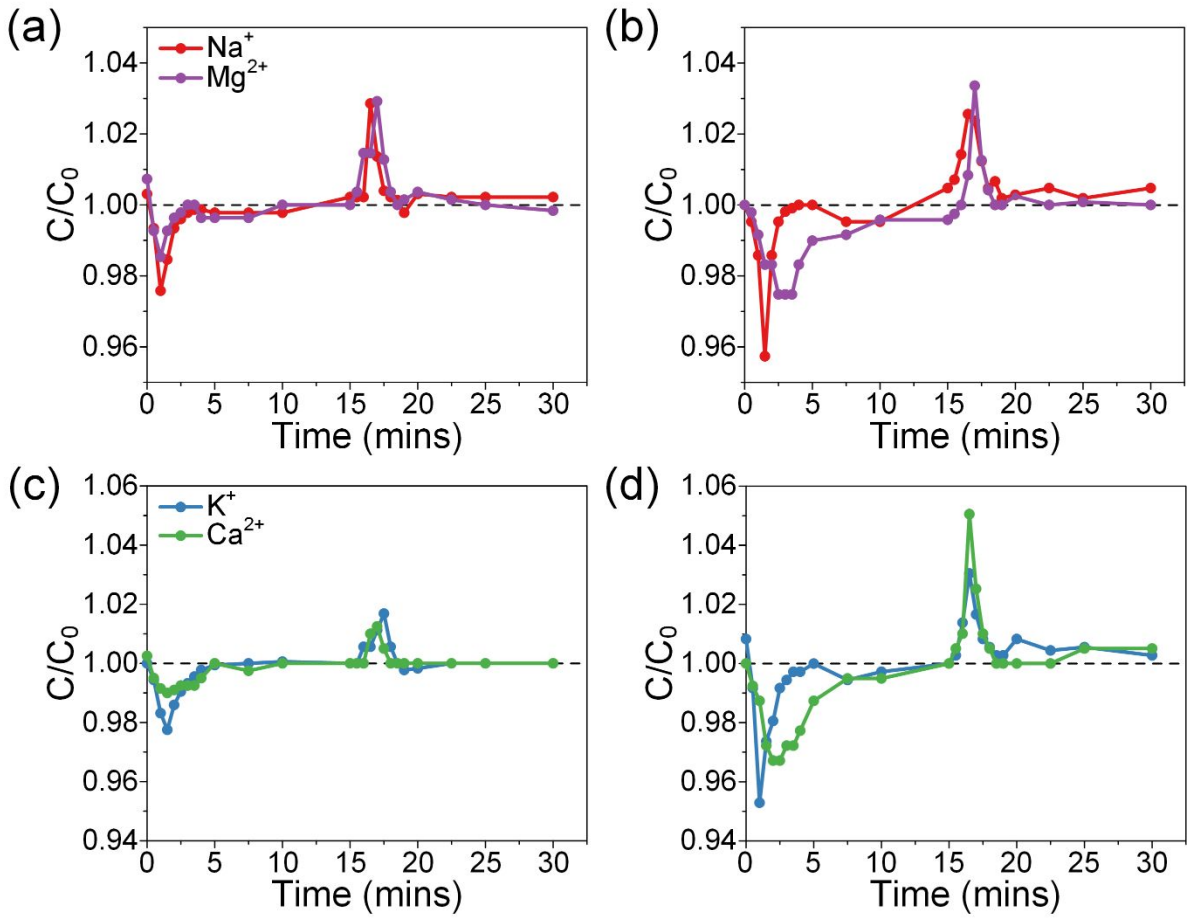


**Figure S6** Cation concentration curves of experiment 3 using either (a) AC or (b) O-AC as the cathode. Cation concentration curves of experiment 4 using either (c) AC or (d) O-AC as the cathode.

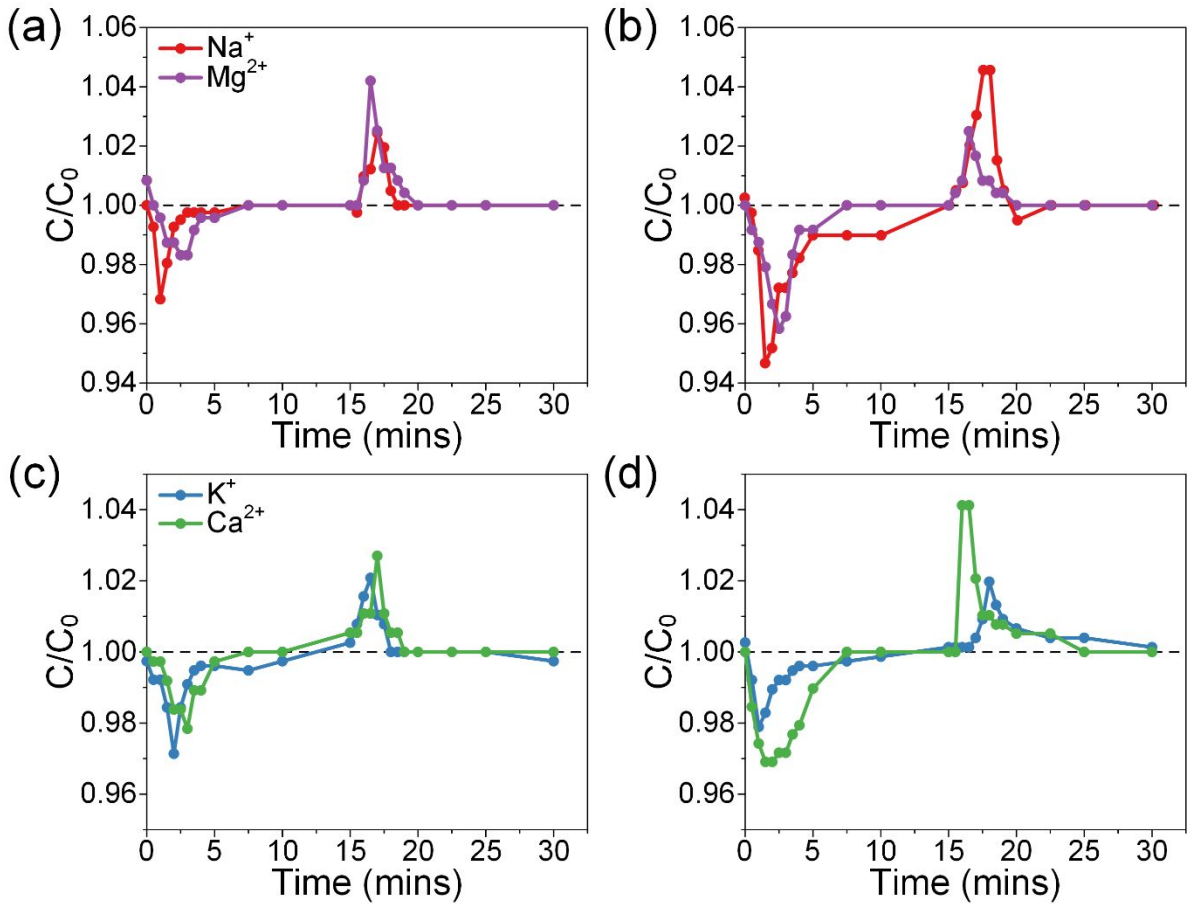


**Figure S7** Cation concentration curves of experiment 5 using either (a) AC or (b) O-AC as the cathode. Cation concentration curves of experiment 6 using either (c) AC or (d) O-AC as the cathode.