# Supplementary information

## Electric Field Modulated Ion-sieving Effects of Graphene Oxide Membranes

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| Feed solution | $K^+$ (mM) | $Na^+$ (mM) | $Ca^{2+}$ (mM) | $Mg^{2+}$ (mM) |
|---|---|---|---|---|
| F1 | 1 | - | 1 | - |
| F2 | 1 | - | - | 1 |
| F3 | - | 1 | 1 | - |
| F4 | - | 1 | - | 1 |

**Table. S1** Description of feed solutions used.

| Ion | Valence | Ionic mass (amu)[1] | Hydrated radius (Å)[2] | Hydration energy (kJ mol$^{-1}$)[3] |
|---|---|---|---|---|
| $K^+$ | +1 | 39.098 | 3.31 | -295 |
| $Na^+$ | +1 | 22.99 | 3.58 | -365 |
| $Ca^{2+}$ | +2 | 40.078 | 4.12 | -1505 |
| $Mg^{2+}$ | +2 | 24.305 | 4.28 | -1830 |

**Table. S2** Ion properties of cations used[1-3].

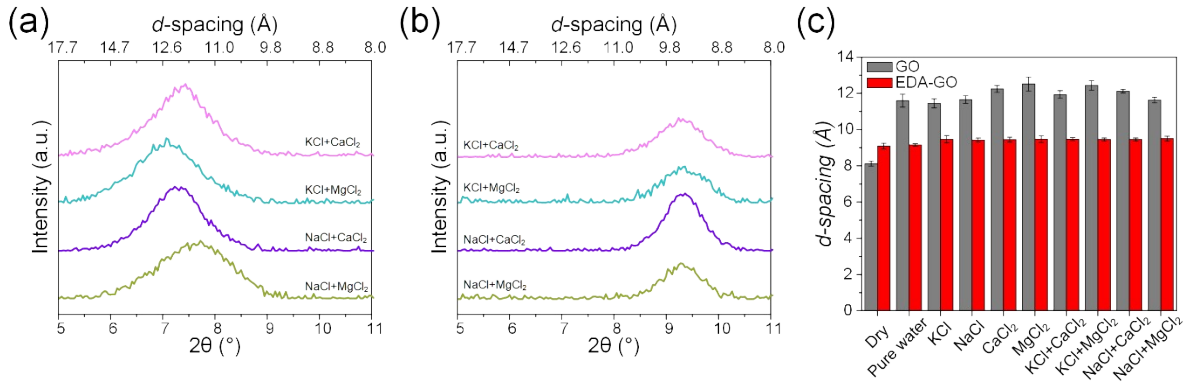


**Fig. S1** XRD results of (a) GO and (b) EDA-GO membrane immersed in mixed 1 mmol monovalent and 1 mmol divalent salt solutions as indicated. (c) *d*-spacings of GO and EDA-GO membranes under various conditions as calculated from (001) XRD peaks.

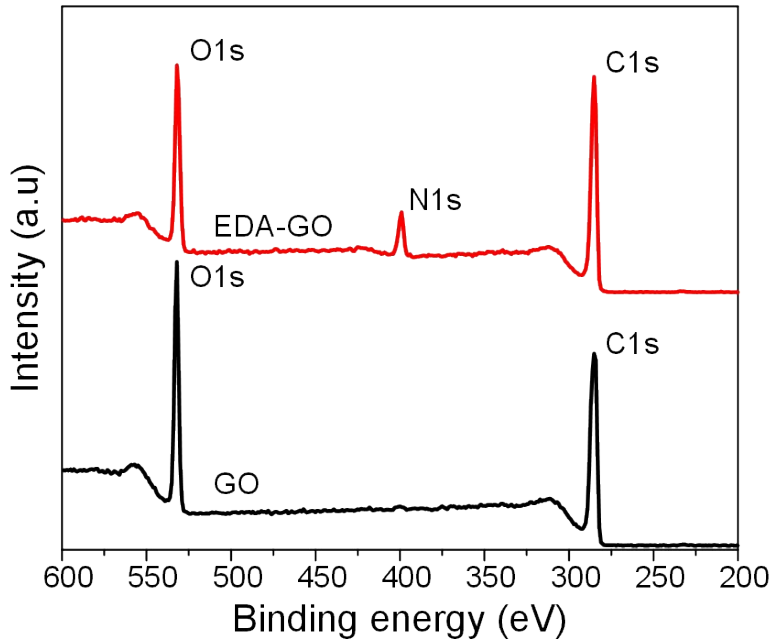


**Fig. S2** Full XPS spectra of GO and EDA-GO membranes.

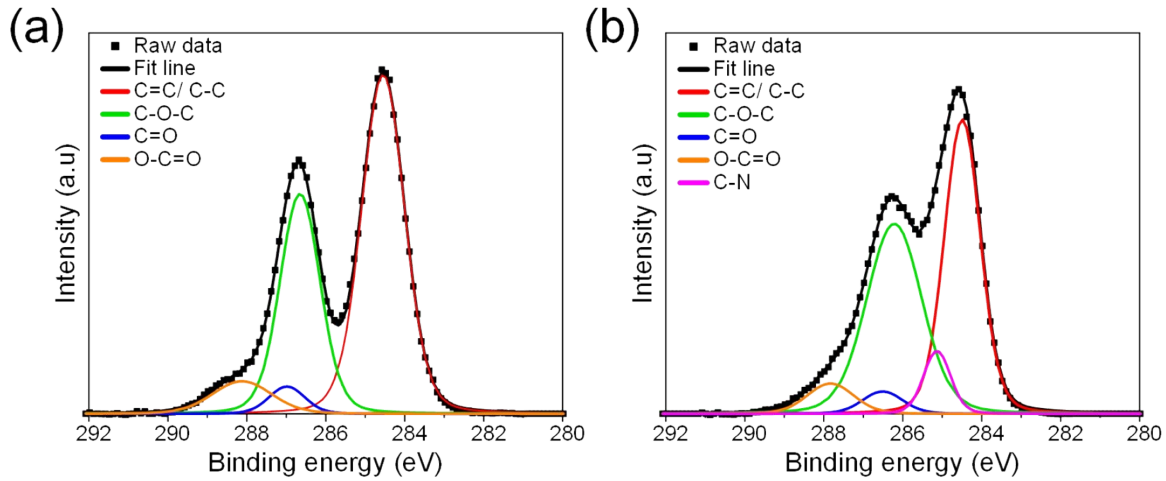


**Fig. S3** C1s XPS spectra of (a) GO and (b) EDA-GO membranes.

| | GO (at%) | EDA-GO (at%) |
|---|---|---|
| **C=C/C-C** | 55.4 | 42.3 |
| **C-O-C** | 33.6 | 41.7 |
| **C=O** | 3.72 | 3.59 |
| **O-C=O** | 7.28 | 5.6 |
| **C-N** | - | 6.81 |

**Table. S3** Atomic percent concentration of peaks in C1s spectrum.

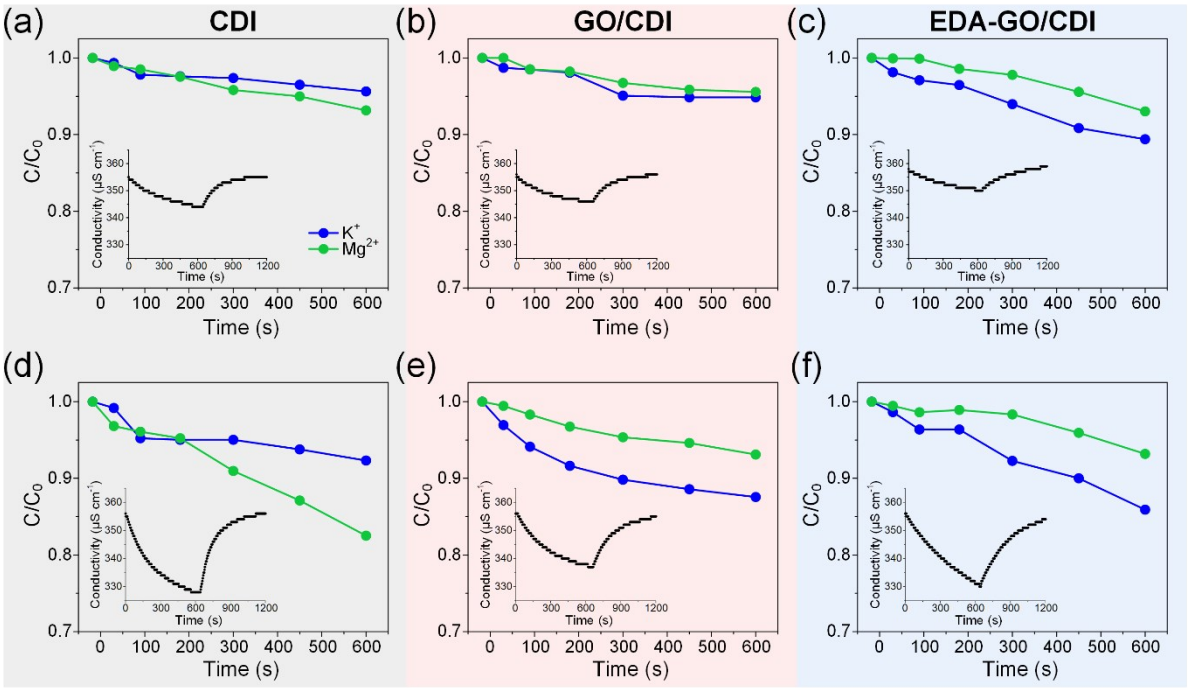


**Fig. S4** Normalised half-cycle cation adsorption curves for CDI (grey area), GO/CDI (red area) and EDA-GO/CDI (blue area) using feed solution F2. Top row (a – c): 0.8 V. Bottom row (d – f): 1.2 V. Inserts are corresponding full-cycle conductivity curves.

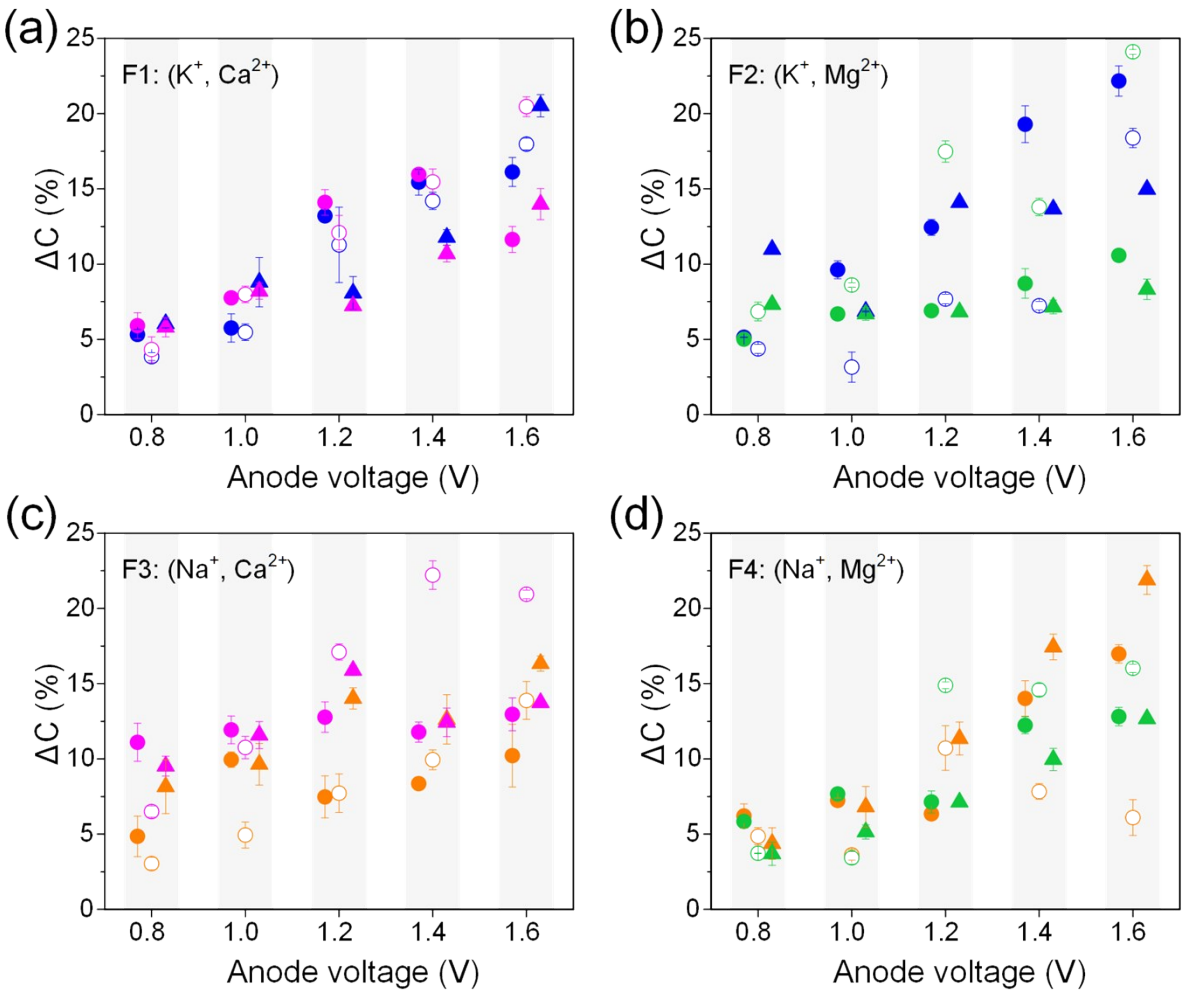


**Fig. S5** Concentration change of effluent solution under positive anode voltage for feed solution (a) F1, (b) F2, (c) F3 and (d) F4. Empty circles (○) denote CDI, filled circles (●) denote GO/CDI and filled triangles (▲) denote EDA-GO/CDI. Blue represents $K^+$, orange represents $Na^+$, magenta represents $Ca^{2+}$ and green represents $Mg^{2+}$. Data points of GO/CDI and EDA-GO/CDI were offset for clarity and error bars denote standard deviations.

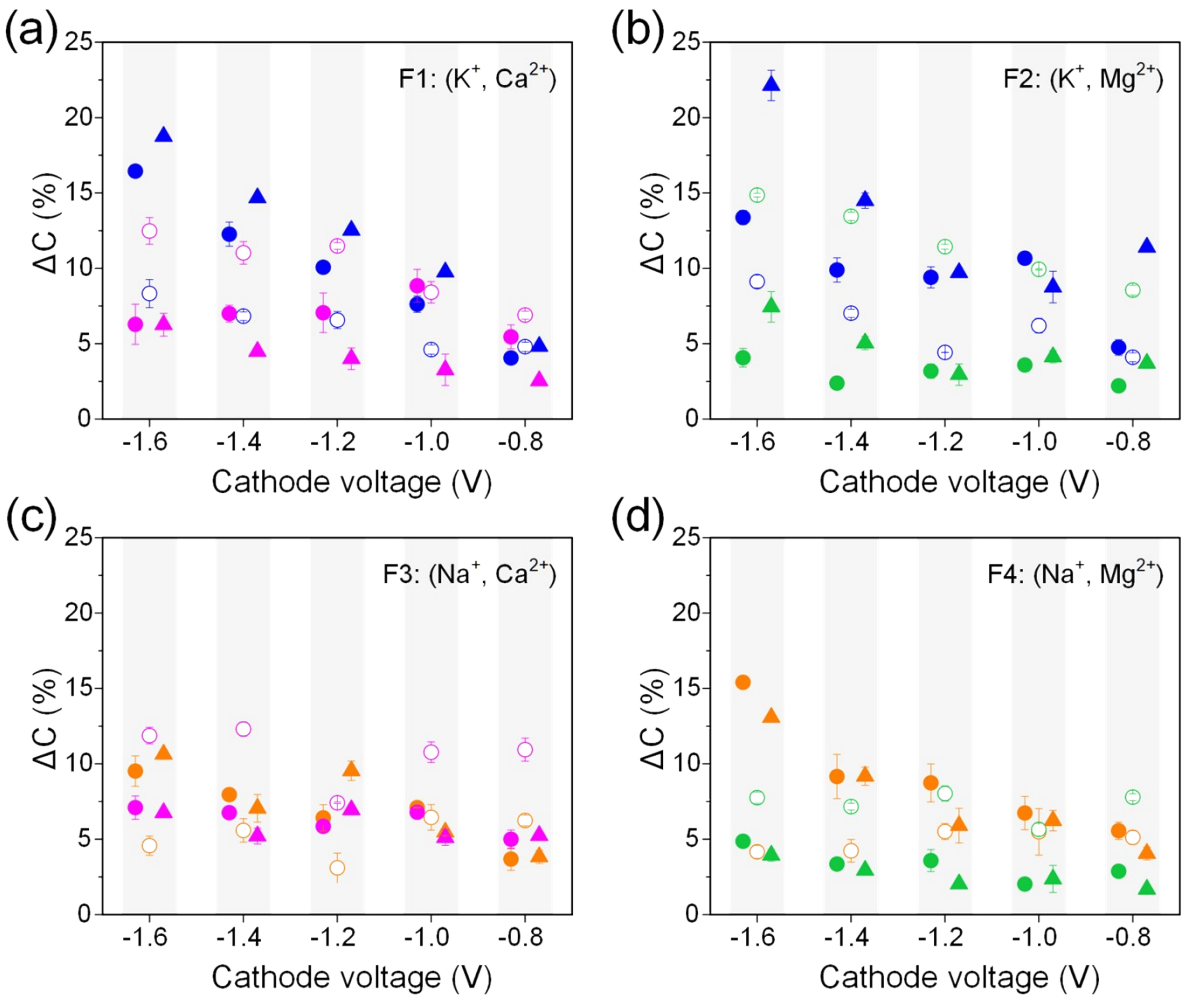


**Fig. S6** Concentration change of effluent solution under negative cathode voltage for feed solution (a) F1, (b) F2, (c) F3 and (d) F4. Empty circles (○) denote CDI, filled circles (●) denote GO/CDI and filled triangles (▲) denote EDA-GO/CDI. Blue represents $K^+$, orange represents $Na^+$, magenta represents $Ca^{2+}$ and green represents $Mg^{2+}$. Data points of GO/CDI and EDA-GO/CDI were offset for clarity and error bars denote standard deviations.

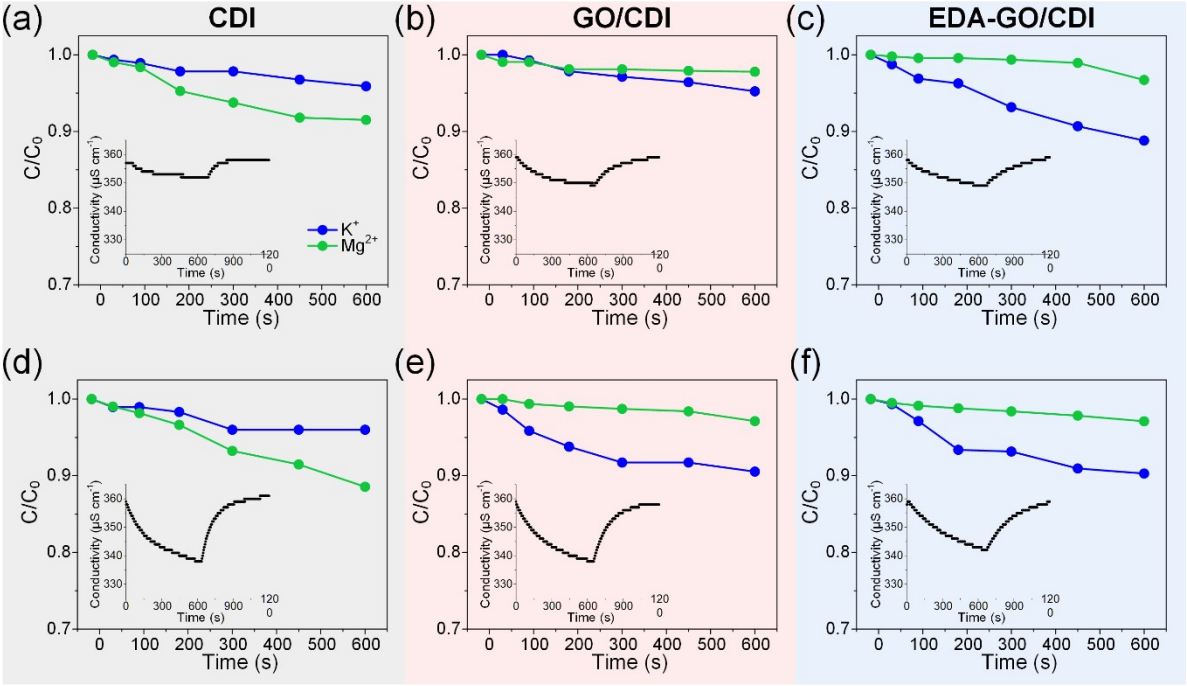


**Fig. S7** Normalised half-cycle cation adsorption curves for CDI (grey area), GO/CDI (red area) and EDA-GO/CDI (blue area) using feed solution F2. Top row (a – c): -0.8 V. Bottom row (d – f): -1.2 V. Inserts are corresponding full-cycle conductivity curves.

**References**


1. Lide DR. *CRC handbook of chemistry and physics: a ready-reference book of chemical and physical data*. CRC press (1995).

2. Nightingale Jr E. Phenomenological theory of ion solvation. Effective radii of hydrated ions. *The Journal of Physical Chemistry* **63**, 1381-1387 (1959).

3. Babu CS, Lim C. Theory of ionic hydration: Insights from molecular dynamics simulations and experiment. *The Journal of Physical Chemistry B* **103**, 7958-7968 (1999).