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Supporting information

A comparative study of graphite electrode using co-intercalation phenomenon for rechargeable Li, Na and K batteries

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Experimental details

Electrodes were prepared by mixing graphite (90 wt%) and polyvinylidene fluoride binder (PVDF, 10 wt%) in an *N*-methyl-2-pyrrolidone (NMP) solvent. The resulting slurry was pasted onto Cu foil, dried at 120°C for 2 h, and roll-pressed. Test cells were assembled in a glove box into a two-electrode configuration with alkali metal counter electrodes. A separator of grade GF/F (Whatman, USA) was used. 1 M MCF_3SO_3 (M= Li, Na or K) in diethylene glycol dimethyl ether (DEGDME) were used as electrolytes. The crystal structure of the samples was analyzed with an X-ray diffractometer (XRD, D2PHASER) using Cu Kα radiation.

Calculation details

Density functional theory (DFT) calculations were performed to investigate the charge distribution in the co-intercalated graphite structures and the repulsion energy between negatively charged graphene layers. We used a projector-augmented wave (PAW) pseudopotentials[1, 2] with a plane-wave basis set as implemented in Vienna *Ab initio* Simulation Package (VASP)[3]. Generalized gradient approximation (GGA)[4] was used for exchange-correlation energy. We used an energy cutoff of 500 eV and all geometric relaxations were performed until the energy change between ionic steps becomes less than 0.01 eV per unit cell to ensure the convergence. Charge distribution in the ternary GICs is calculated by Bader charge analysis[5]. The amount of injected charge is one per 21 C atoms, which corresponds to the amount of electron transfer for the capacity delivered (~106 mAh g^{-1}). The free energy was also normalized to the energy per 21 C atoms.

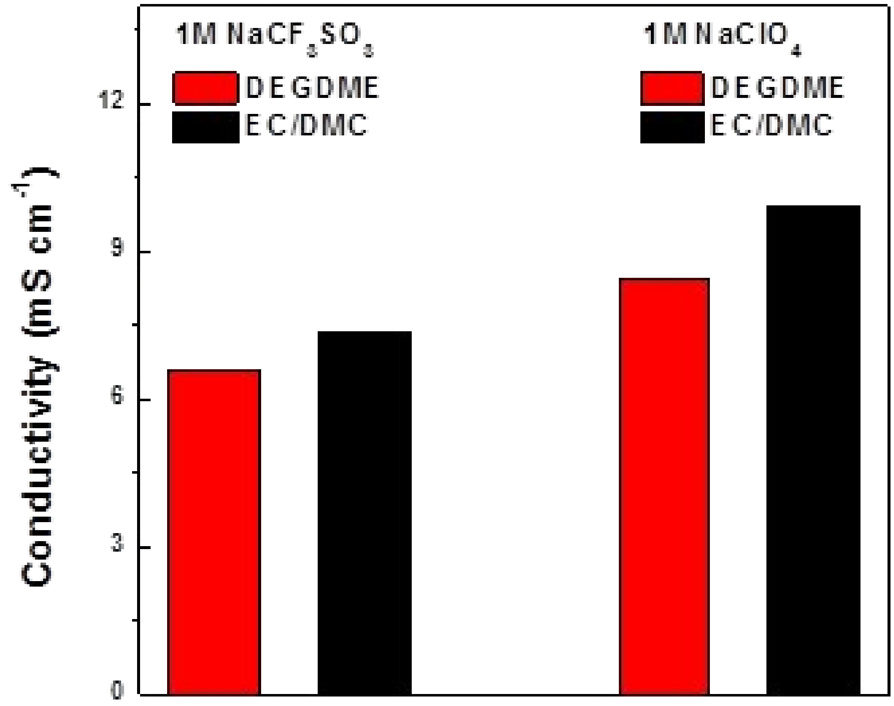

Figure S1. Ionic conductivity of DEGDME and EC/DMC electrolytes.

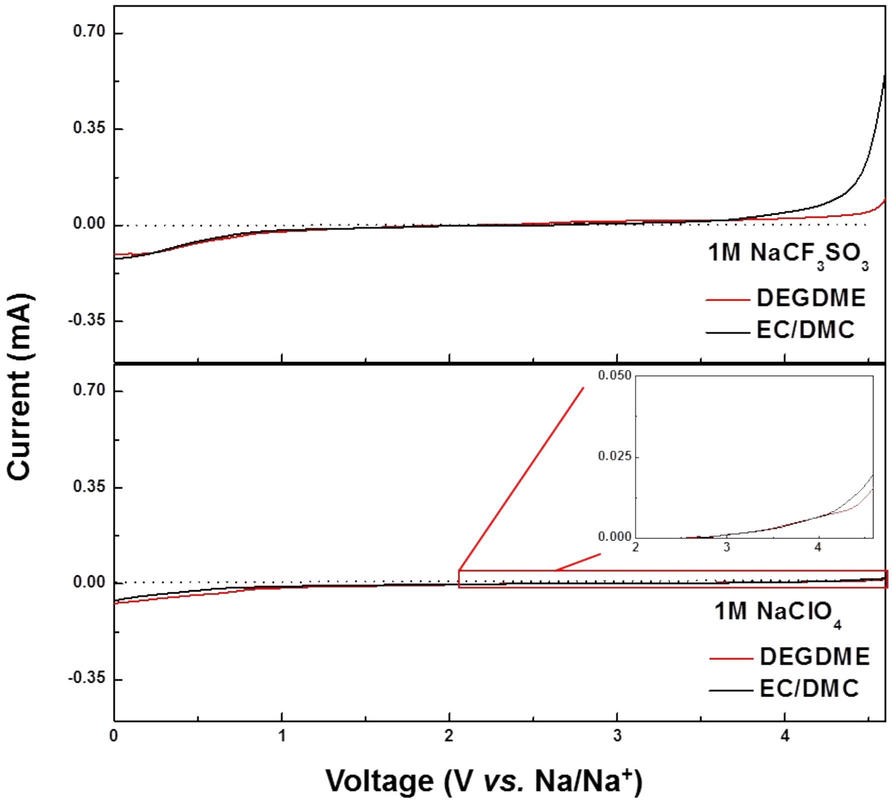

Figure S2. Stability of DEGDME and EC/DMC electrolytes.

References

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