

Supporting Information

Exploring Stability of Nonaqueous Electrolytes for Potassium-Ion Batteries

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Author Contributions

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■ EXPERIMENTAL SECTION

Graphite powder, acetylene black and polyvinylidene fluoride (PVDF) were mixed at a mass ratio of 92:4:4 and dissolved in N-Methyl pyrrolidone. The slurry was coated onto the Cu foil as the current collector. The coated Cu foil was then dried under vacuum at 110 °C for 24 h, rolled-pressed, and punched in required size (~ 1.1 cm²). The loading mass of graphite is ~ 4.0 mg/cm². All the test cells were assembled in glove box filled with Ar. Besides Na-K/graphite cells employing the home-made configuration, the other cells were standard coin cells (CR2032). The separator was the glass fiber (Grade GF/A, Whatman). The electrolytes contained 0.5 M KPF₆ in EC/DEC and 0.5 M KPF₆ in DEGDME, and the water content was below 10 ppm by Karl Fischer titration (Metrohm 831).

All galvanostatic discharge/charge tests were performed using a Land 2001A battery testing system at 298 K. The lower and upper cutoff voltages were 0.01 and 2.0 V (vs. K⁺/K), respectively. ¹H nuclear magnetic resonance (NMR) spectrums of EC/DEC and DEGDME electrolytes before and after electrochemical tests were collected by a Bruker AVANCE III 400 instrument. The residual electrolyte from the separator was dissolved in CDCl₃. Powder X-ray diffraction (XRD) pattern of the graphite electrode was obtained by using a diffractometer (Rigaku D/max 2500/PC) in which Cu-Ka ($\lambda = 0.154$ nm) was used as the radiation source. X-ray photoelectron

spectroscopy (XPS) spectra were carried out on a PHI 5000 VersaProbe II spectrometer using monochromatic Al K α X-ray radiation. All specimens were washed by pure DMC for several times followed by vacuum drying.

In Figure 3, the K foil with the mass of 30 mg in K/K cell can afford a capacity of 20.6 mAh based on the theoretical capacity (687 mAh/g) of the metal K, and discharge/charge (plating/stripping) test is conducted for a constant time of 10 h (600 min). For the G/G cell the K-GIC electrode only has a capacity of less than 1.0 mAh, around equal to 1/20 of the K foil in K/K cell (Figure S4 in the Supporting Information). In addition, the shorter potassiation/depotassiation time can avoid the the increasing of the voltage which would lead to the possible decomposition of EC/DEC electrolyte. Thus, the constant time of the discharge/charge (potassiation/depotassiation) test for G/G cell is set 20 min.

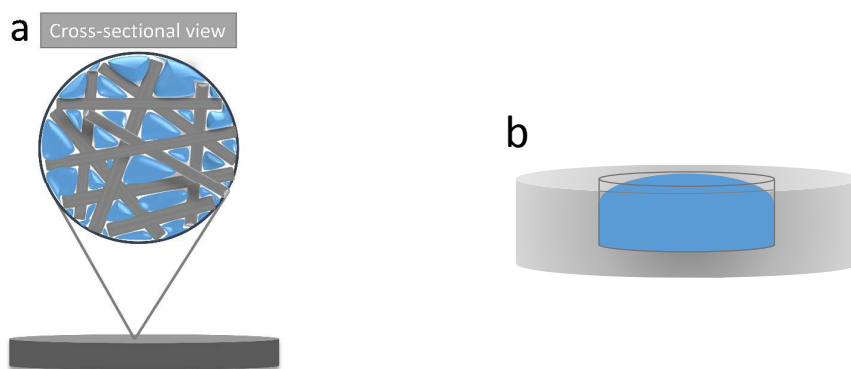


Figure S1. Schematic of the Na-K alloy (a) immobilized in carbon paper and (b) placed in home-made configuration.

Scheme S1. The KIB configuration and assembly based on Na-K liquid alloy as the anode.

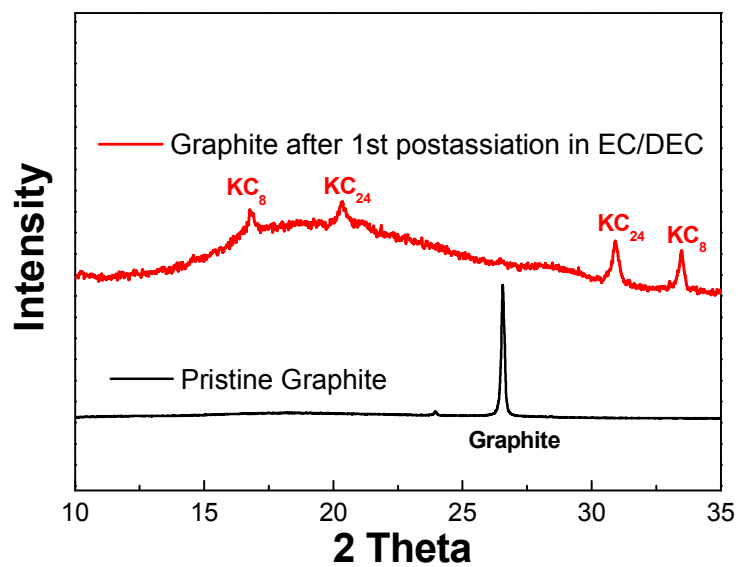
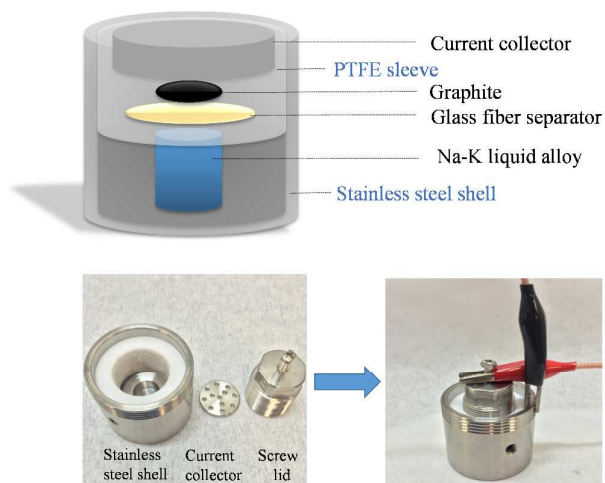


Figure S2. XRD pattern of the graphite electrode after the initial potassiation process in EC/DEC electrolyte.

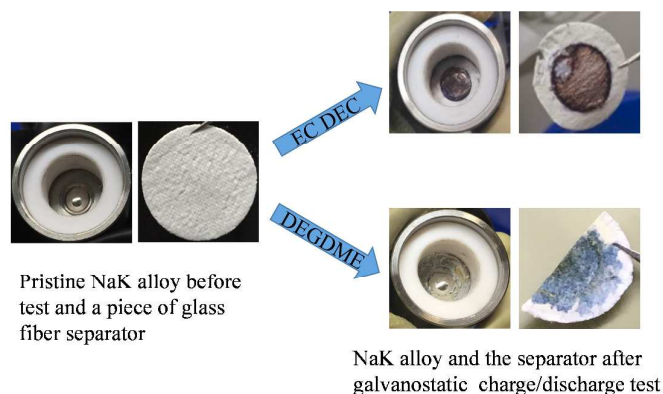


Figure S3. The Na-K alloy and the separator before and after cycling test in different electrolyte. The solubility of the Na-K alloy in ether-based solvents made the separator become blue.^{1,2}

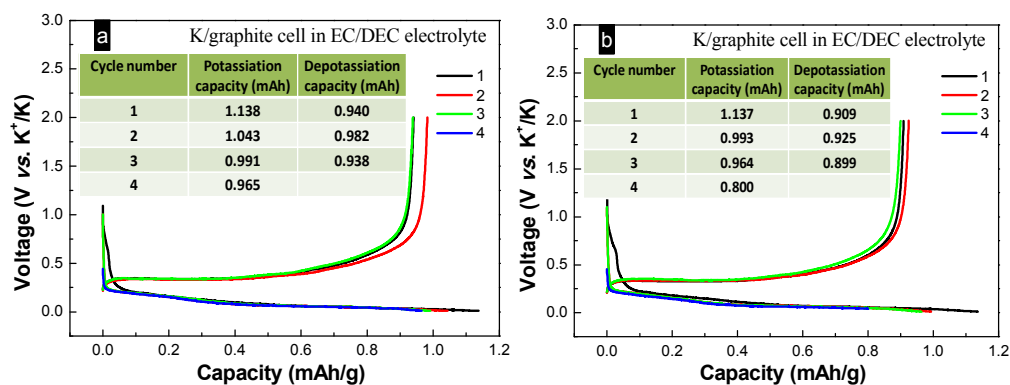


Figure S4. Discharge-charge curves of the graphite electrodes for K/G cells in EC/DEC electrolyte. Two K/graphite cells using EC/DEC electrolyte were assembled. One cell ran for three cycles and then was full discharged (potassiated) at 0.1 mA/cm² (Figure S4a), the other ran for three cycles and then was discharge to 0.8 mA/cm² (Figure S4b). Then two cells were disassembled in glove box and the graphite electrodes was washed with low boiling DMC (dimethyl carbonate) solvent. Next, a G/G cell using the electrodes with full discharged capacity and the capacity of 0.8 mAh as the anode and the cathode, respectively, was assembled.

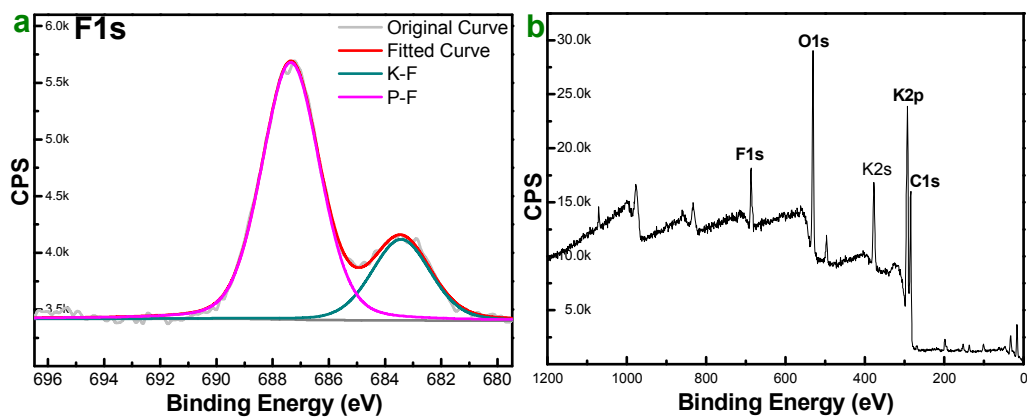


Figure S5. XPS spectra of graphite anode for K/graphite half-cell with EC/DEC based electrolyte after 10 cycles: (a) F 1s and (b) full spectrum.

■ REFERENCES

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