Design of LiF-rich solid electrolyte interface layer through salt-

additive chemistry for boosting fast-charging phosphorus-based

lithium ion batteries performance

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Experimental Section

Synthesis of RP/CNT anode and electrolyte: Commercial RP (1.4 g), CNTs (0.6 g) and zirconium oxide balls (9, 20 mm in diameter) were put into a zirconium oxide vessel (0.1 L), which was sealed in an Ar-filled glove box and performed with a rotation rate of 400 rpm for 12 h using the high energy mechanical milling technique in a ball mill instrument. The 4.5 mol/L LiFSI/DME-FEC was prepared by dissolving a certain amount of LiFSI salt into the DME solvent, then 3 wt.% FEC was added to the electrolyte system. Similarly, the 4.5 mol/L LiFSI/DME was prepared without FEC additive. 1.0 mol/L commercial LiPF₆ in an ethylene carbonate (EC)-diethyl carbonate (DEC) solution with 5 wt.% addition of FEC was selected as a comparison as well. All chemicals and related experiments were performed in an Ar-filled glove box with less than 0.1 ppm O₂ and H₂O.

Materials characterization and electrochemical measurements: To identify the SEI composition of RP/CNT anode, X-ray photoelectron spectroscopy (XPS) was conducted on a Kratos-Axis spectrometer with monochromatic Al K α (1,486.71 eV) radiation (operating current and voltage of 10 mA and 15 kV, respectively). The battery performance was evaluated by galvanostatic cycling of coin (CR 2032) cells with the RP/CNT anode (working electrode) and Li foil (counter/reference electrode). The working electrode was made using a slurry method with RP/CNT, carbon black and polyvinylidene fluoride (PVDF) binder with a mass ratio of 7:2:1 in the N-methyl-pyrrolidone (NMP) solvent; the mass loading of RP/CNT was around 0.5-0.7 mg cm⁻² on the Cu foil. Galvanostatic discharge/charge measurements were performed on a Land battery test system in a voltage range of 0.01–3 V at room temperature. The capacity in the electrode was calculated according to the mass of composite.



Figure S1 Cycle performance for RP/CNT electrodes in 1M LiFSI/DME and 4.5 M LiFSI/DME electrolytes at discharging/charging current densities of 1 A g⁻¹.