Electronic Supplementary Information

Towards thermally stable high performance lithium-ion batteries: the combination of a phosphonium cation ionic liquid and a 3D porous molybdenum disulfide/graphene electrode

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Experimental section

Materials

Ionic liquid trimethyl(isobutyl)phosphonium bis(fluorosulfonyl)imide (P\textsubscript{1111}FSI, purity > 99.5\%) was provided by Cytec Canada Inc.. LiFSI (Solvionic, France) was used without further purification. Graphite powder was sourced from Qingdao Haida Corporation. All other chemicals were purchased from Sigma-Aldrich.

Preparation of IL-based electrolyte

The IL electrolyte mixture with LiFSI (3.2 mol kg\textsuperscript{-1}) was prepared by an adding-dissolving process, followed by drying under vacuum for 48 hours at 80 °C in the presence of sodium hydride (NaH) until the water content was below 50 ppm (determined by Karl Fischer titration analysis).

Synthesis of graphene oxide (GO)

The GO was synthesized via a modified Hummers method.\textsuperscript{1} Typically, graphite powder (2 g) and NaNO\textsubscript{3} (1 g) were added into the concentrated H\textsubscript{2}SO\textsubscript{4} (75 mL) in an ice bath,
followed by a gradual addition of KMnO$_4$ (5 g) under stirring. This mixture was stirred at room temperature for 6 days, and then diluted with 5% H$_2$SO$_4$ (140 mL). It was stirred for another 2 h at 90 °C, followed by an addition of 30% H$_2$O$_2$ (5 mL). The resulting precipitate, graphite oxide, was rinsed with HCl aqueous solution (1:10) and water. Graphite oxide powder was sonicated into water to form graphene oxide dispersion, which was dialyzed for 1 week prior to use.

*Synthesis of MoS$_x$ and MoS$_x$/rGO*: The MoS$_x$ and MoS$_x$/rGO was synthesized using potentiostatic deposition. Briefly, the electrodeposition was performed at a potential of -1.2 V (vs. Ag/AgCl) for 60 min on a stainless steel mesh from a solution containing 5 mM (NH$_4$)$_2$MoS$_4$, 0.1 M KCl with or without 0.25 mg mL$^{-1}$ GO.

*Material characterization*: The morphology of samples were characterized with field emission scanning electron microscopy (FE-SEM) (JEOL JSM-7500FA) and transmission electron microscopy (TEM) (JEOL JEM-2200FS). X-ray photoelectron spectroscopy (XPS) spectra were collected by illuminating the samples with a non-monochromatic x-ray source (Omnivac) using Al Kα (1486.6 eV) radiation, and the photoemission collected by an SES2002 analyser (Scienta). Raman spectra were obtained with a confocal Raman spectrometer (Jobin Yvon HR800, Horiba) using a 632.8 nm diode laser. Thermo-gravimetric analysis (TGA) was performed by Q500 (TA instruments) in air between 100 °C to 700°C at a ramp rate of 10 °C min$^{-1}$.

*Electrochemical characterization*: The LR 2032 type coin cell was assembled with an electrode (0.8 × 0.8 cm) coupled with a lithium foil. The electrolyte used was either IL electrolyte or 1 M LiPF$_6$ in 1:1 (v/v) ethylene carbonate/dimethyl carbonate. A piece of glass fiber was used as separator. Cyclic voltammetry (CV) was performed on a Solartron SI 1287. The galvanostatic charge/discharge tests were performed on a LAND CT2001A battery test system. Electrochemical impedance spectroscopy (EIS) measurements were carried out on a Bio-logic workstation (VSP model) at an open circuit potential over the frequency range of 0.01 Hz to 100 kHz. The charge/discharge tests at 50 °C were performed on BioLogic 810 Battery Cycler combined with AISET Laboratory incubator.
Fig. S1 Chemical structure of P_{1114}FSI.

Fig. S2 EDS spectra of the MoS$_2$ (a) and MoS$_2$/rGO (b).
Fig. S3 TGA curves of MoS$_2$ and MoS$_2$/rGO electrodes.

Fig. S4 XPS C 1s spectra of GO (a) and MoS$_2$/rGO (b).
Fig. S5 The first three cyclic voltammograms of the cells over a potential range of 0.0-3.0 V vs. Li/Li$^+$ at 0.2 mV s$^{-1}$ and charge/discharge curves at 0.1 A g$^{-1}$ for MG-CE (a, b), MoS$_2$-CE (c, d), and MoS$_2$-IL (e, f).
Fig. S6 Nyquist plots of the (a) MG-CE and MG-IL cells and (b) MoS$_2$-CE and MoS$_2$-IL cells (solid line: fitted curves; Inset shows the equivalent circuit).