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

Yu Ge[a], Cristina Pozo-Gonzalo[b], Yong Zhao[a], Xiaoteng Jia[a], Robert Kerr[b], Caiyun Wang[a], Patrick C. Howlett*[b], and Gordon G. Wallace[a]*

[a]*ARC Centre of Excellence for Electromaterials Science, Intelligent Polymer Research Institute, AIIM Facility, University of Wollongong, NSW 2522, Australia.*

[b]*Institute for Frontier Materials (IFM), Deakin University, Burwood, Victoria 3125, Australia.*

E-mail: caiyun@uow.edu.au, patrick.howlett@deakin.edu.au

Experimental section

Materials

Ionic liquid trimethyl(isobutyl)phosphonium bis(fluorosulfonyl)imide ($P_{111i4}FSI$, 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^{-1}) was prepared by an adding-dissolving process, followed by drying under vacuum for 48 hours at 80 ^{o}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.[1] Typically, graphite powder (2 g) and $NaNO_3$ (1 g) were added into the concentrated H_2SO_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_2SO_4 (140 mL). It was stirred for another 2 h at 90 ^oC, followed by an addition of 30% H_2O_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 $(NH_4)_2MoS_4$, 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 ^oC to 700^oC at a ramp rate of 10 ^oC 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 ^oC were performed on BioLogic 810 Battery Cycler combined with AISET Laboratory incubator.

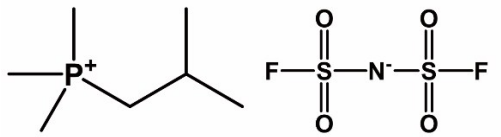

Fig. S1 Chemical structure of $P_{111i4}FSI$.

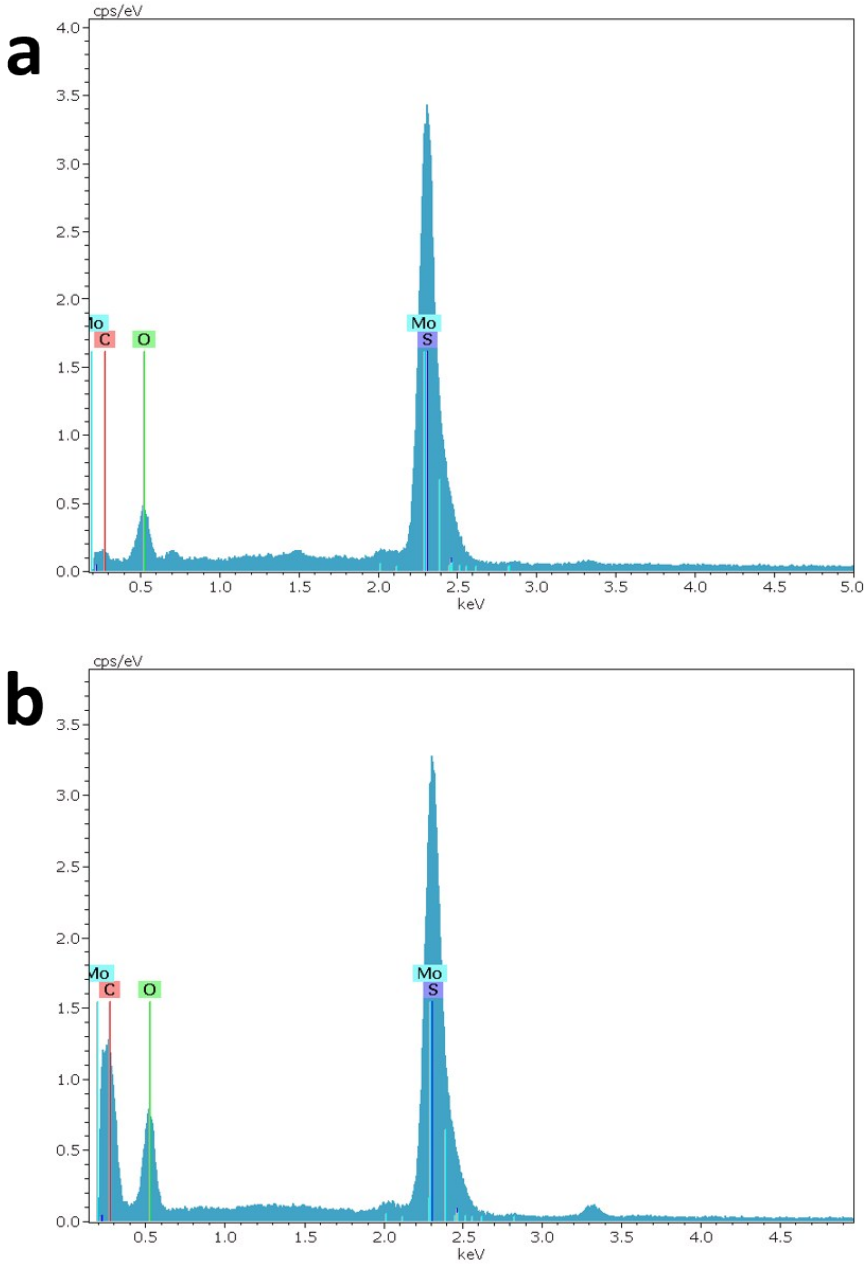

Fig. S2 EDS spectra of the MoS_2 (a) and MoS_2/rGO (b).

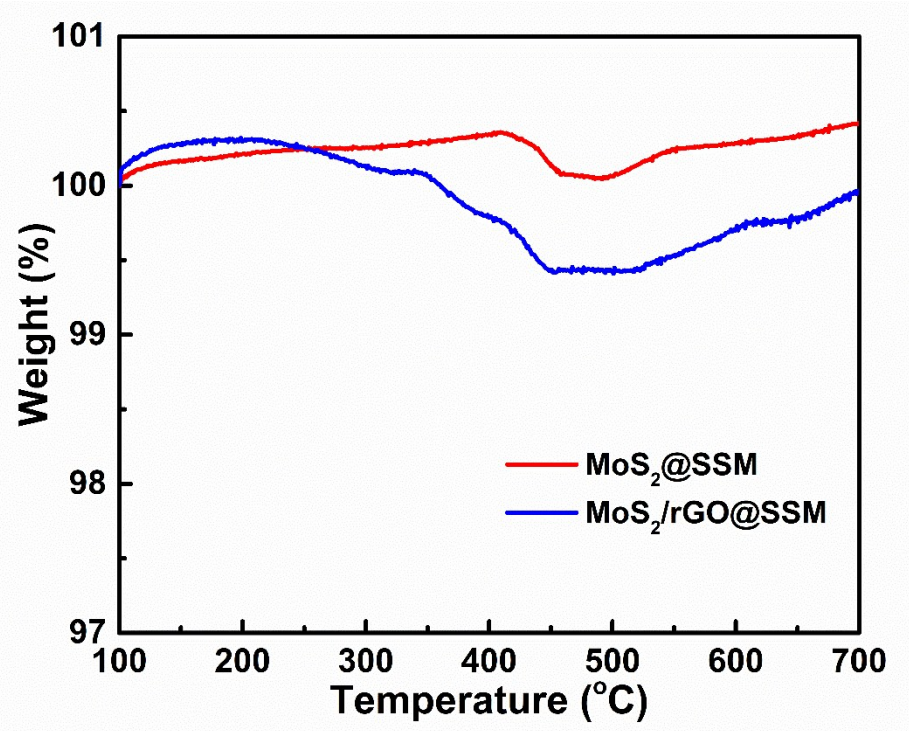

Fig. S3 TGA curves of MoS_2 and MoS_2/rGO electrodes.

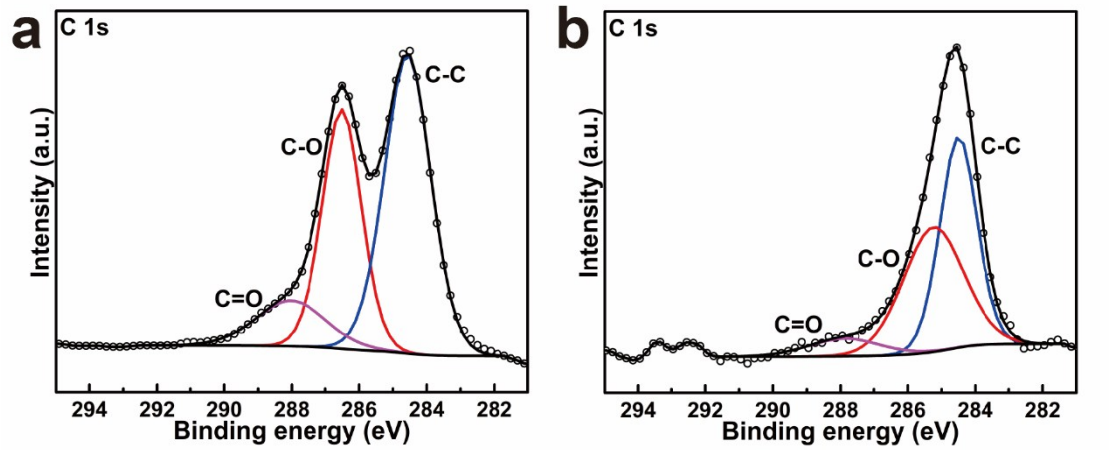

Fig. S4 XPS C 1s spectra of GO (a) and MoS_2/rGO (b).

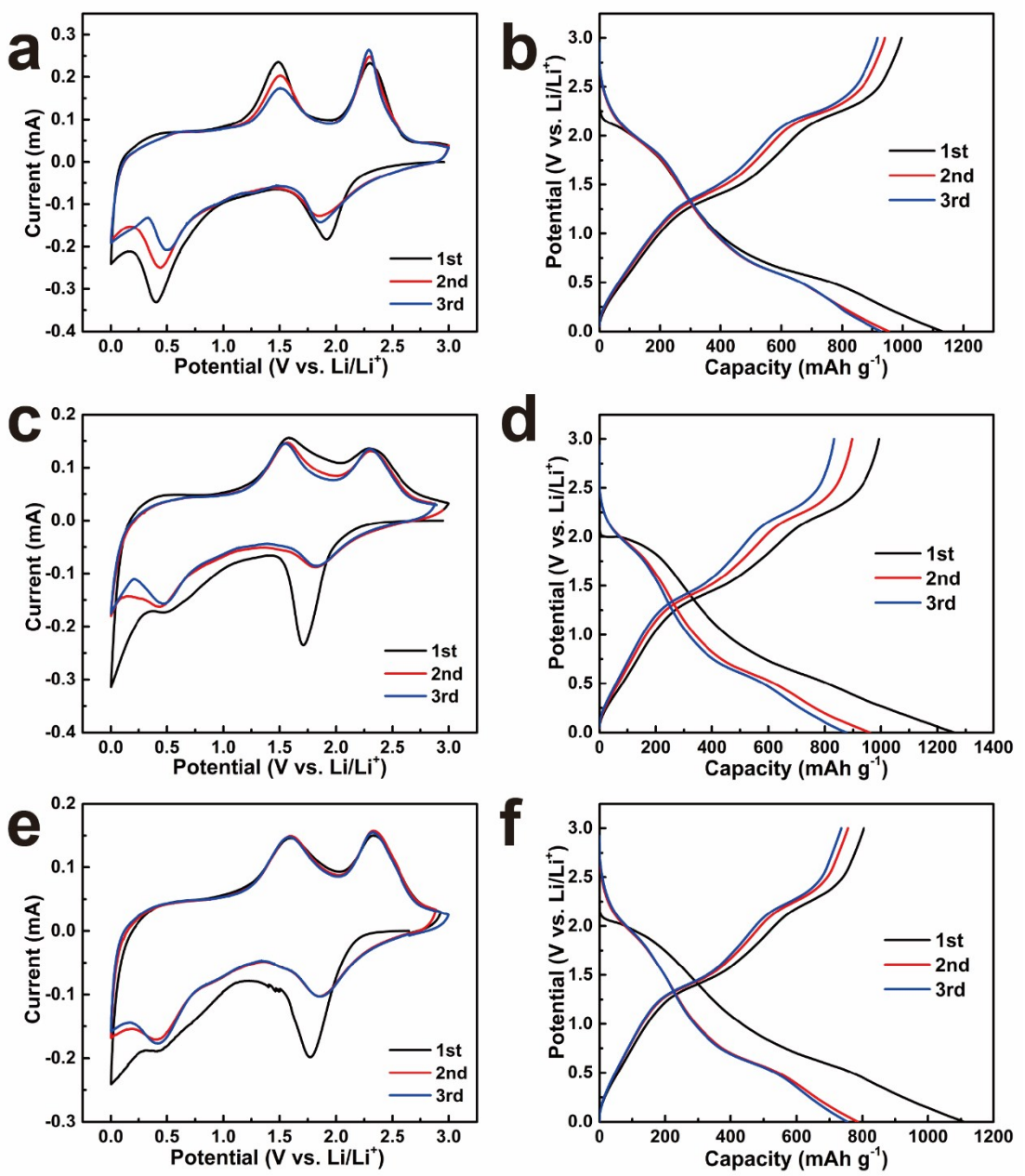

Fig. S5 The first three cyclic voltammograms of the cells over a potential range of 0.0-3.0 V vs. Li/Li^+ at 0.2 mV s^{-1} and charge/discharge curves at 0.1 A g^{-1} for MG-CE (a, b), MoS_2-CE (c, d), and MoS_2-IL (e, f).

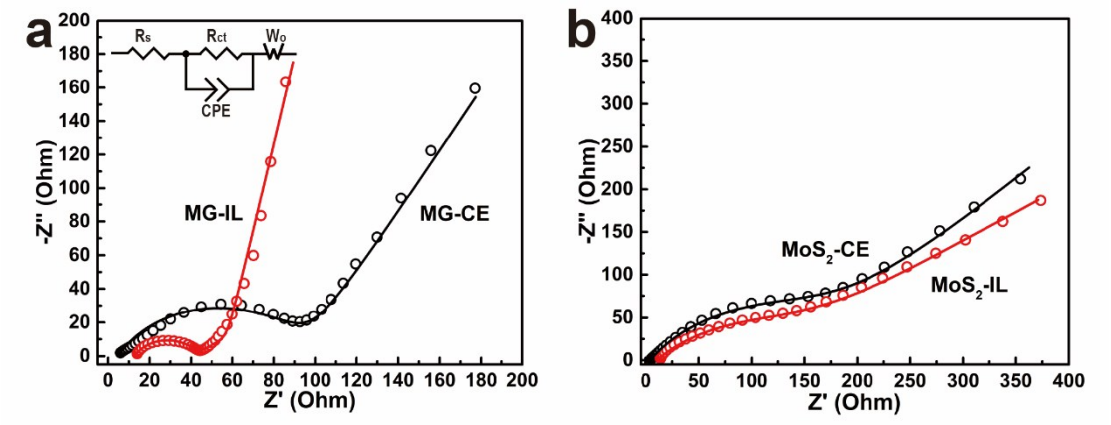

Fig. S6 Nyquist plots of the (a) MG-CE and MG-IL cells and (b) MoS_2-CE and MoS_2-IL cells (solid line: fitted curves; Inset shows the equivalent circuit).

1. Y. Meng, K. Wang, Y. Zhang and Z. Wei, *Advanced Materials*, 2013, **25**, 6985-6990.