Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A. This journal is © The Royal Society of Chemistry 2016

Supporting Information

Reversible Anion Intercalation in a Layered Aromatic Amine: A High-Voltage Host Structure for Organic Batteries

Elise Deunf^a, Philippe Moreau^a, Eric Quarez^a, Dominique Guyomard^a, Franck Dolhem^{bc}, Philippe Poizot*^{ad}

^{a.} Institut des Matériaux Jean Rouxel (IMN), UMR CNRS 6502, Université de Nantes, 2 rue de la Houssinière, B.P. 32229, 44322 Nantes Cedex 3, France.

b. Laboratoire de Glycochimie, des Antimicrobiens et des Agroressources (LG2A), FRE CNRS 3517, Université de Picardie Jules Verne, 33 rue Saint-Leu, 80039 Amiens Cedex, France.

^{c.} Réseau sur le Stockage Électrochimique de l'Énergie (RS2E), FR CNRS 3459, France.

^{d.} Institut Universitaire de France (IUF), 103 bd Saint-Michel, 75005 Paris Cedex 5, France.

Index to supporting information:

Figure S1a. ¹H and ¹³C NMR spectra of Li₂DAnT·~2THF

Figure S1b. ¹H and ¹³C NMR spectra of Li₂DAnT

Figure S2. Overlaid FT-IR spectra for Li₂DAnT·~2THF and Li₂DAnT

Figure S3. TEM image of Li₂DAnT

Figure S4. FT-IR spectra of dried Li₂DAnT-containing PC solvent and its corresponding thermal analysis

Figure S5. Potential vs differential capacity curves for Li/Li₂DAnT cells using either PC/LiClO₄ 1 M or PC/LiTFSI 1 M as electrolyte (rate: 1 electron exchanged in 2 h)

Figure S6. Discharge capacity retention curves for Li/Li_2DAnT cells cycled at a rate of 1 electron in 2 h or 20 h using PC/LiClO₄ 1 M as electrolyte

Figure S7. Electrochemical evaluation of the two-electron reaction for Li_2DAnT measured in Li half cells using different PC-based electrolytes

Figure S8. Long-term cycling of pure Li₂DAnT (400 cycles) vs. Li with post-mortem SEM investigation



Figure S1a. ¹H and ¹³C NMR spectra of Li₂DAnT·~2THF measured in (CD₃)₂SO and recorded on a 400 MHz apparatus.



Figure S1b. ¹H and ¹³C NMR spectra of Li₂DAnT measured in $(CD_3)_2SO$ and recorded on a 400 MHz apparatus.



Figure S2. Overlaid FT-IR spectra of as-prepared Li₂DAnT·~2THF (deep blue) and Li₂DAnT (light blue), respectively.



Figure S3. Typical TEM image of Li_2DAnT confirming the obtainment of a lamellar structure. White arrows correspond to crystals lying perpendicular to the electron beam while black ones correspond to those parallel.



Figure S4. (a) Overlaid FT-IR spectra of Li_2DAnT (light blue) and Li_2DAnT soaked in PC then dried (black). (b) TG-DSC traces of Li_2DAnT soaked in PC then dried measured under argon at a heating rate of 5 °C.min⁻¹.



Figure S5. Overlaid of the differential capacity vs potential curves regarding the first cycle of Li_2DAnT/Li half cells cycled in PC/LiClO₄ 1 M (red) and PC/LiTFSI 1 M (green), respectively (extracted from potential-specific capacity curves reported in Figure 5).



Figure S6. Discharge capacity retention curves for Li half cells using Li_2DAnT as active electrode material mixed with 33 wt% of carbon black and galvanostatically cycled at a rate of 1 electron exchanged in 2 h ($I_m = 37.3 \text{ mA.g}^{-1}$) or 20 h ($I_m = 3.73 \text{ mA.g}^{-1}$), respectively, using PC/LiClO₄ 1 M as electrolyte.



Figure S7. Electrochemical evaluation of the two-electron reaction for Li₂DAnT measured in Li half cells using 33 wt% of carbon black and galvanostatically cycled at a rate of 1 electron exchanged in 5 h ($I_m = 15 \text{ mA.g}^{-1}$) in different electrolytes (red: PC/LiClO₄ 1 M; orange: PC/LiPF₆ 1 M; green: PC/LiTFSI 1 M). The expected electrochemical process for Li₂DAnT is also indicated at the molecular level.



Figure S8. (a) Same discharge capacity retention curves with coulombic efficiency as reported in Figure 6b extended to 400 cycles (long-term cycling). (b) SEM imaging of the positive electrode material after 400 cycles (no binder, no carbon additive). Note that no metallization of the sample was performed for the SEM investigation.