Electronic Supplementary Material (ESI) for RSC Advances.


# Supporting Information

# A Low-Cost and High-Loading Viologen-Based Organic Electrode for Rechargeable Lithium Batteries

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**Materials**

Lithium metal was received from China Energy Lithium Co. Ltd. Ketjen Black EC-600JD (KB) was received from AzkoNobel. Lithium hexafluorophosphate ($LiPF_6$), Lithium perchlorate ($LiClO_4$), Methyl-2-pyrrolidinone (NMP), polytetrafluoroethylene (PTFE) solvent (60 wt.%) and Propylene carbonate (PC) were received from Sigma-Aldrich, ethylene carbonate (EC) and diethyl carbonate (DEC) were received from Guangdong Zhuguang New Energy Technology Co.,Ltd. 4,4'-Bipyridine, bromoethane, potassium hexafluorophosphate ($KPF_6$), acetonitrile (ACN), acetone and ethanol were received from Shanghai Aladdin Reagent Co. Ltd. Aluminum foil and coin cell (CR2032) were received from Shenzhen Kejing Zhida Technology Co. Ltd. Carbon paper (SGL 39AA) was received from SGL Carbon Group. The electrolyte used in this work was 1.0 M $LiClO_4$ PC or 1.0 M $LiPF_6$ EC/DEC (v/v~1:1).

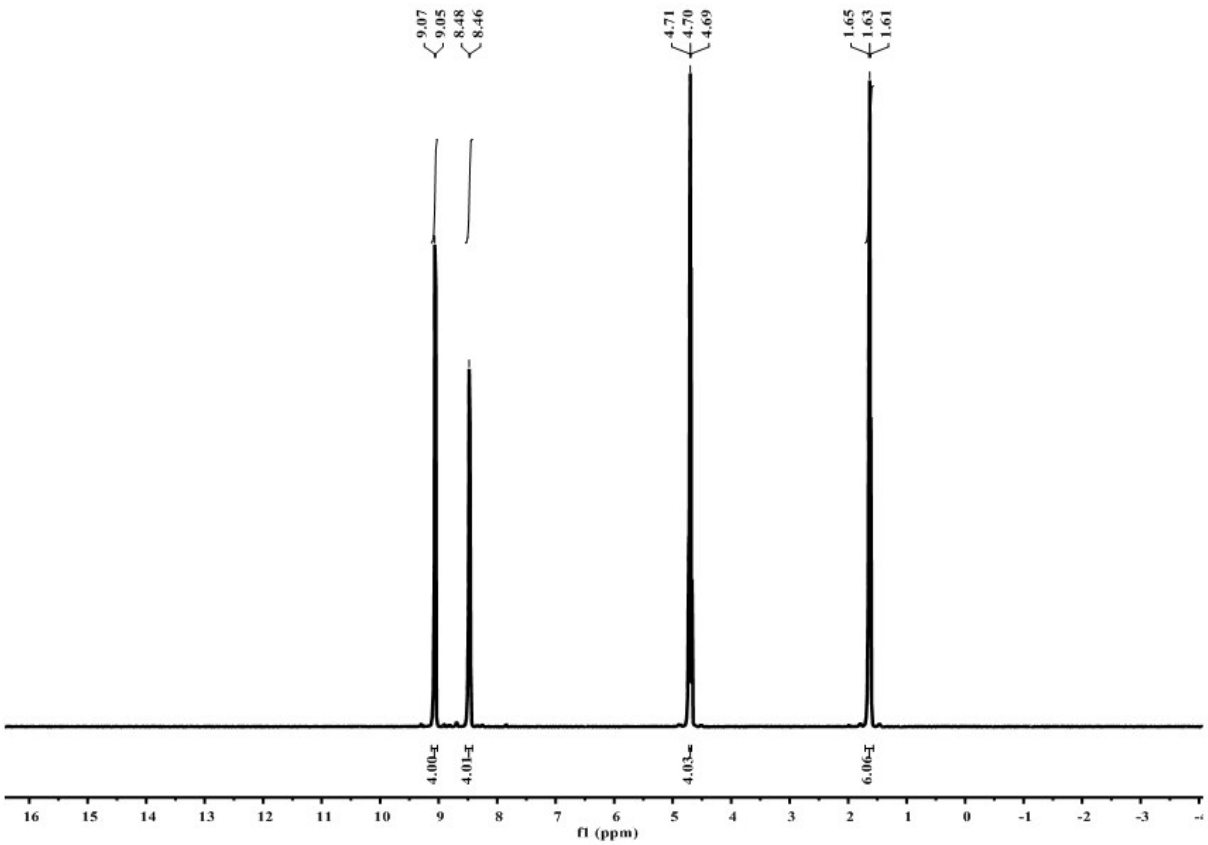


**Figure S1.** $^{1}$H NMR of EV in $D_2O$. $^{1}$H NMR (500 MHz, D2O) $\delta$ = 9.06 (d, 4 H, $J$ = 6.8 Hz), 8.47 (d, 4 H, $J$ = 6.0 Hz), 4.70 (d, 4 H, $J$ = 3.2 Hz), 1.63 (t, 6 H, $J$ = 7.2 Hz).

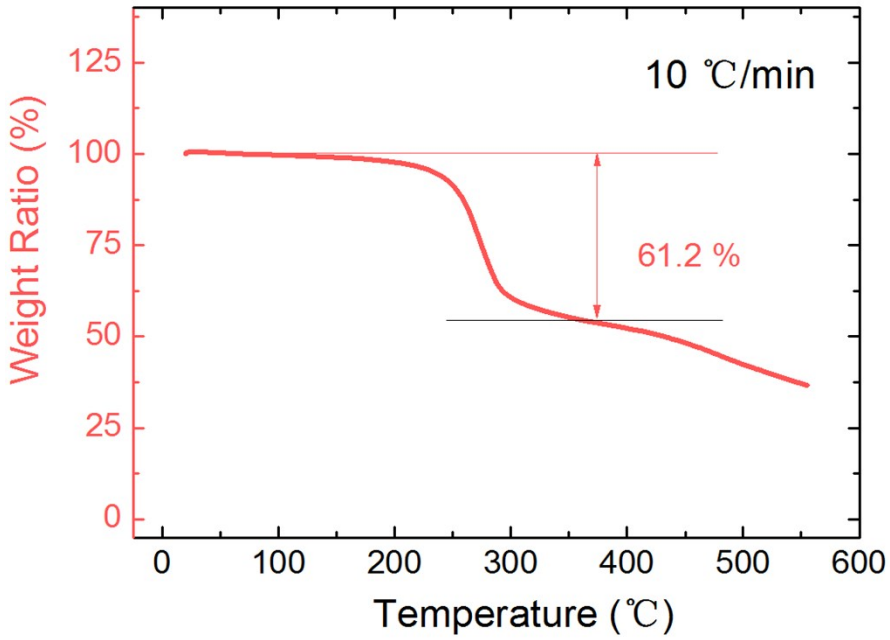


**Figure S2.** Thermal gravity analysis of as prepared EV-KB composite.

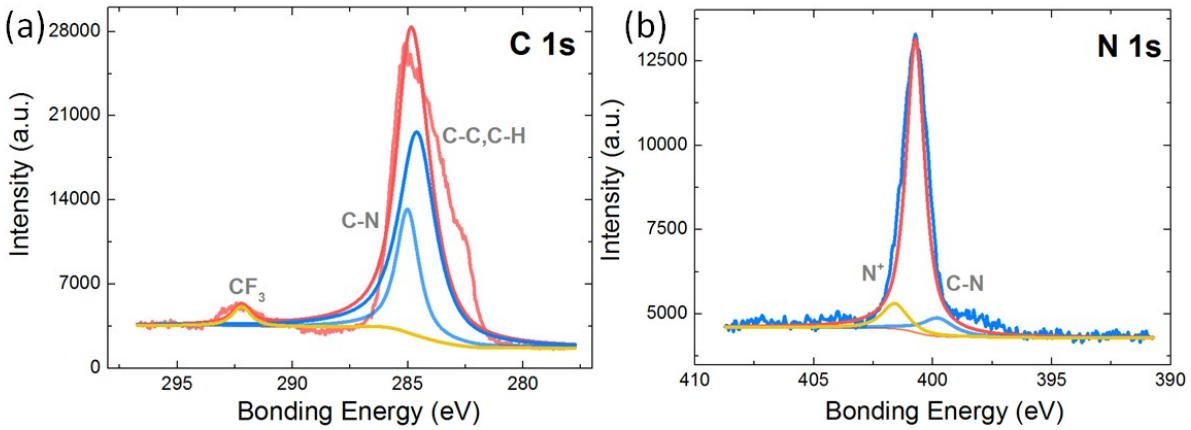


**Figure S3.** XPS spectra of EV-KB composite in (a) C 1s and (b) N 1s regions.

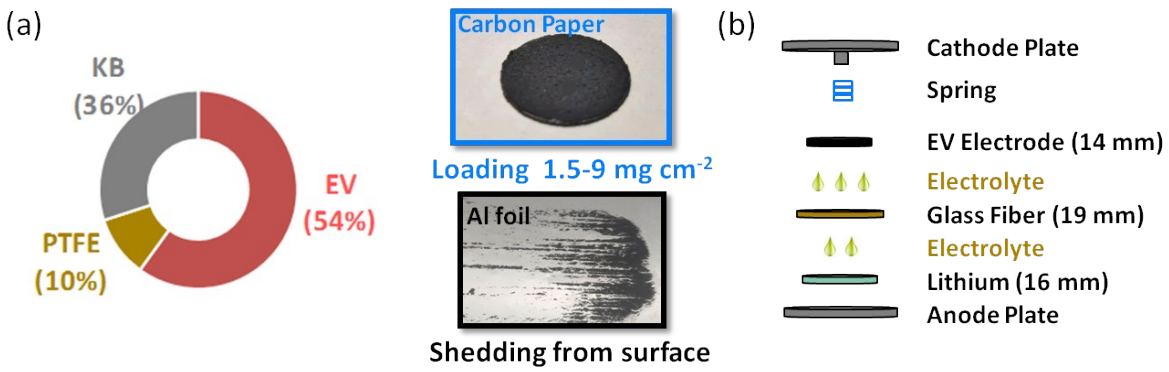


**Figure S4.** (a) EV-KB composite electrode composition and coating comparison on carbon paper and Al foil. (b) Schematic illustration of Li-EV based cell configuration.

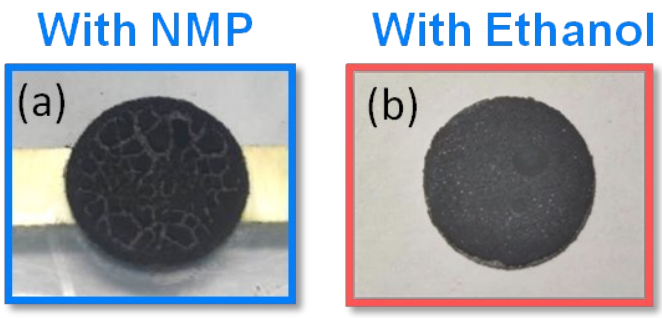


**Figure S5.** EV-KB electrode coating comparison with solvents of (a) N-methyl-2-pyrrolidone (NMP) and (b) Ethanol.

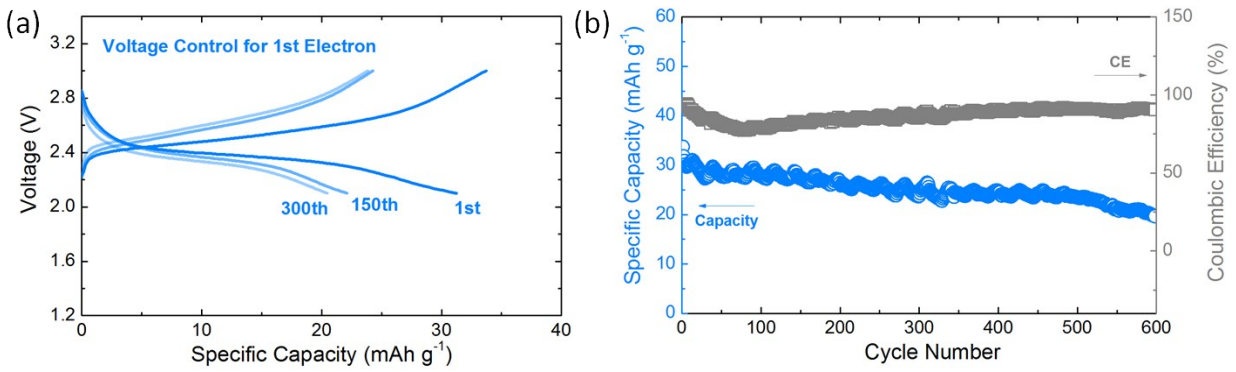


**Figure S6.** (a) Galvanostatic discharge-charge profiles of EV-KB composite electrode for 1st electron at 0.2 mA $cm^{-2}$. (b) Capacity retention and Coulombic efficiency of EV-KB composite electrode of 1st electron at 0.2 mA $cm^{-2}$.

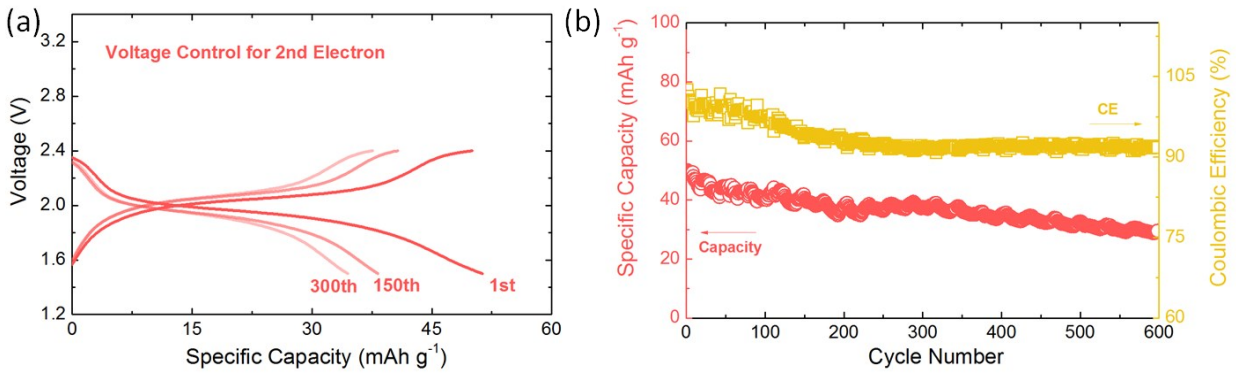


**Figure S7.** (a) Galvanostatic discharge-charge profiles of EV-KB composite electrode for 2nd electron at 0.2 mA $cm^{-2}$. (b) Capacity retention and Coulombic efficiency of EV-KB composite electrode of 2nd electron at 0.2 mA $cm^{-2}$.

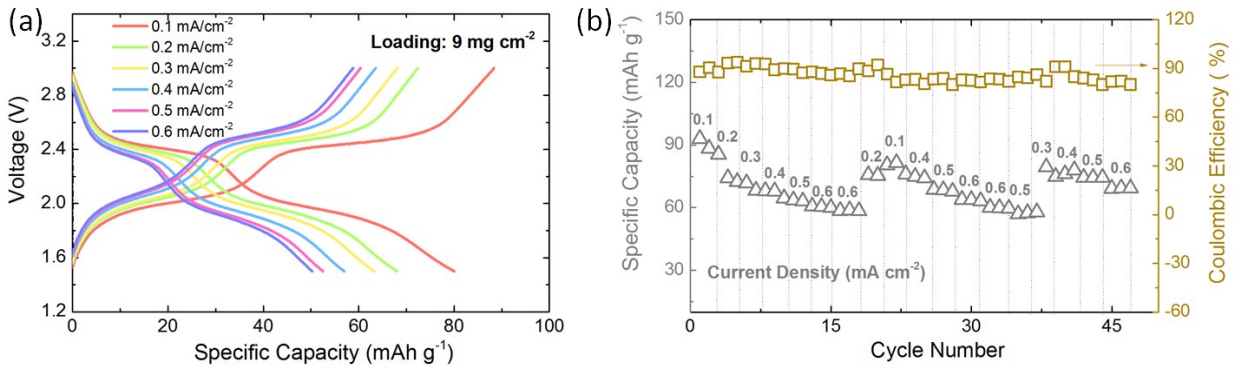


**Figure S8.** (a) First galvanostatic discharge-charge profiles of EV-KB composite electrode of mass loading ~9 mg $cm^{-2}$ between 0.1 and 0.6 mA $cm^{-2}$. (b) Rate capability and Coulombic efficiency of the EV-KB composite electrode at various current densities from 0.1 to 0.6 mA $cm^{-2}$.

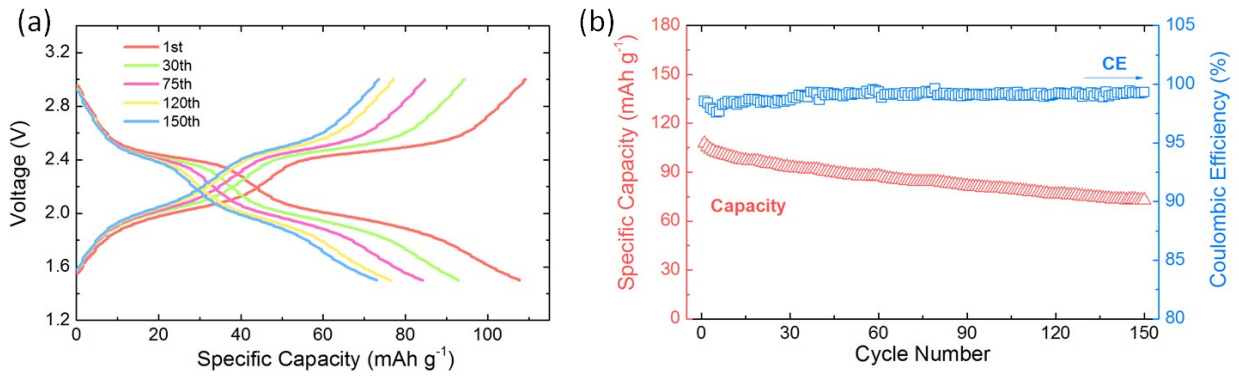


**Figure S9.** (a) Galvanostatic discharge-charge profiles of EV-KB composite electrode of mass loading ~1.5 mg $cm^{-2}$ at 0.2 mA $cm^{-2}$.(b) Capacity retention and Coulombic efficiency of EV-KB composite electrode of mass loading ~1.5 mg $cm^{-2}$ at 0.2 mA $cm^{-2}$.

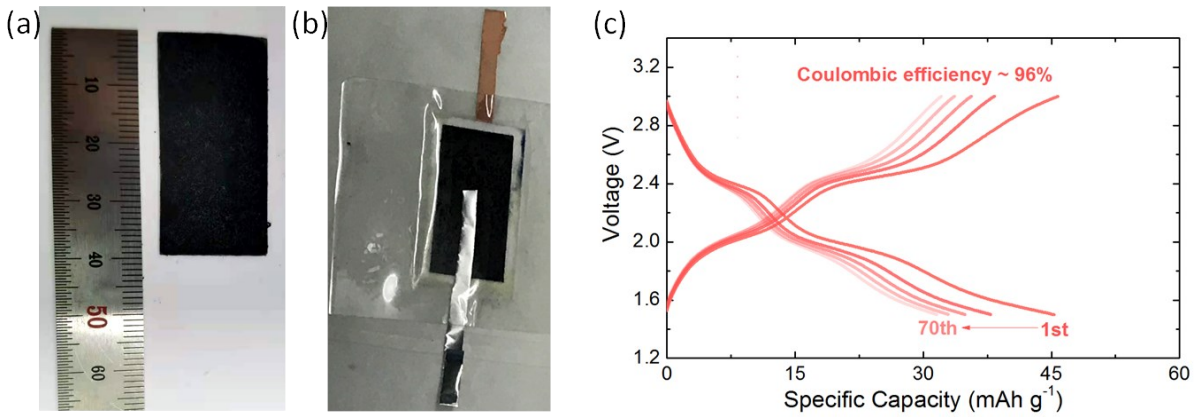


**Figure S10.** (a) Picture of EV-KB composite electrode with area of 4×2 $cm^2$. (b) Picture of EV-KB composite electrode in self-made pouch cell. (c) Galvanostatic discharge-charge profiles of EV-KB composite electrode of mass loading ~1.9 mg $cm^{-2}$ in pouch cell at 0.8 mA.

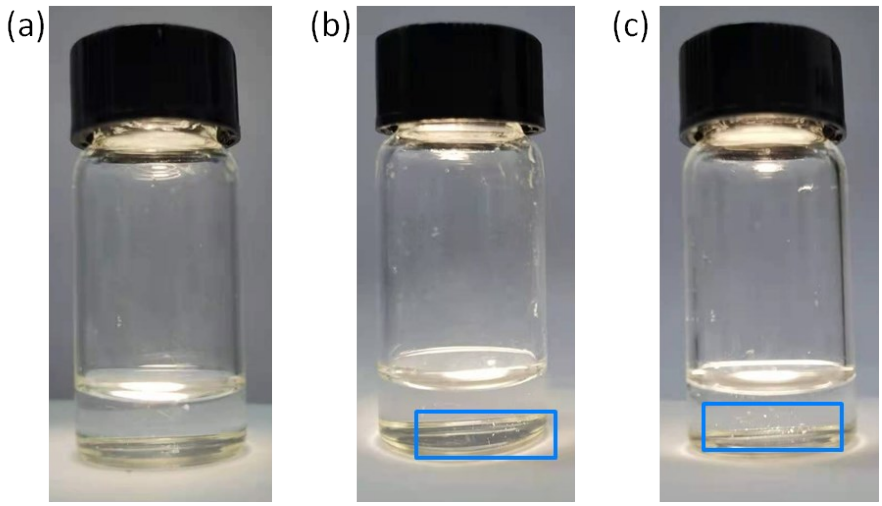


**Figure S11.** Solubility test pictures of (a) 2 mM; (b) 5 mM and (c) 10 mM $EV(PF_6)_2$ in 1 M $LiClO_4$ PC electrolyte.

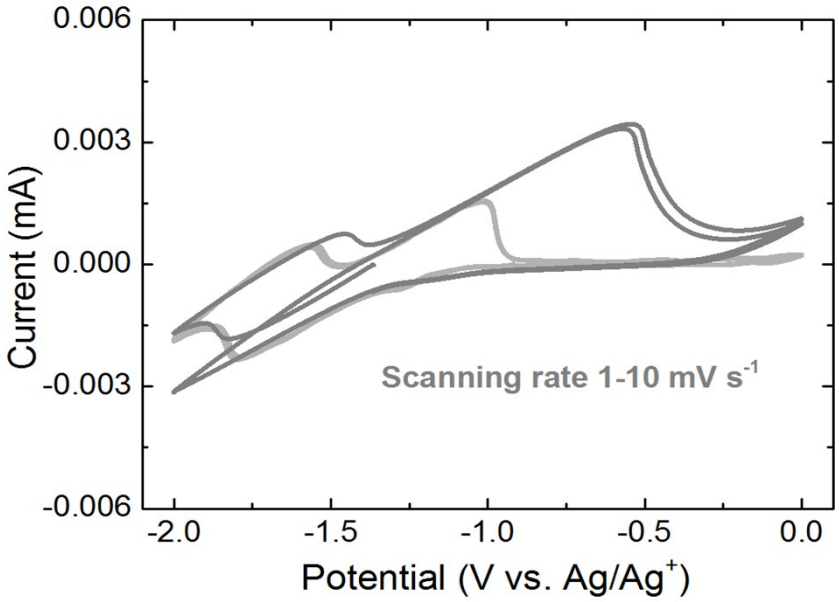


**Figure S12.** Cyclic voltammetry results of 2 mM $EV(PF_6)_2$ in 1 M $LiPF_6$ in EC/DEC at various scanning rates.

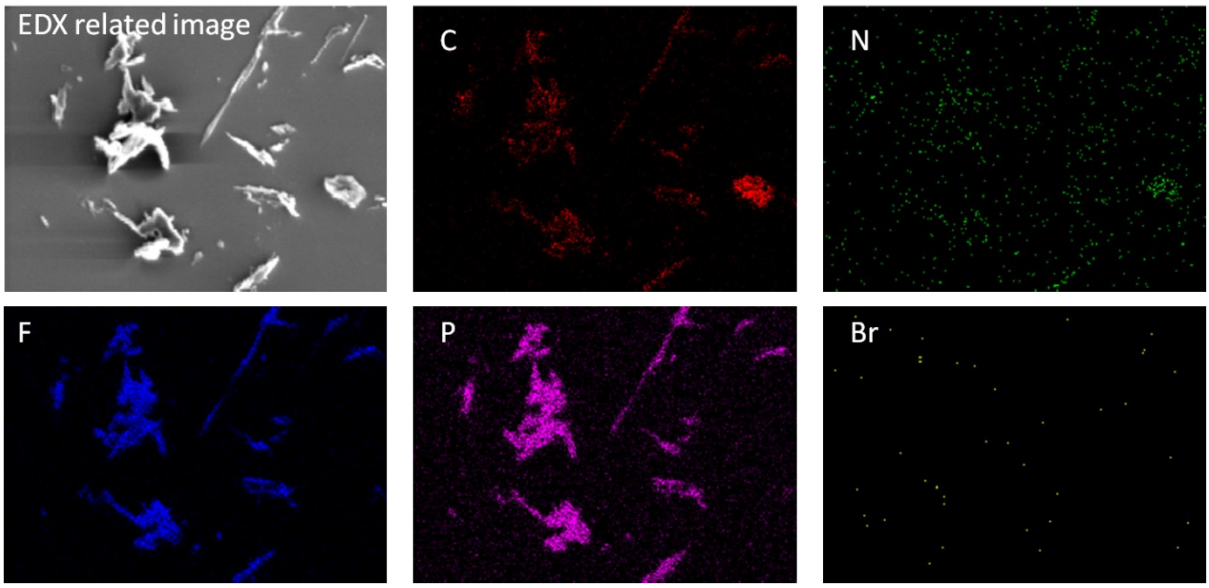


**Figure S13.** EDX images of $EV(PF_6)_2$.

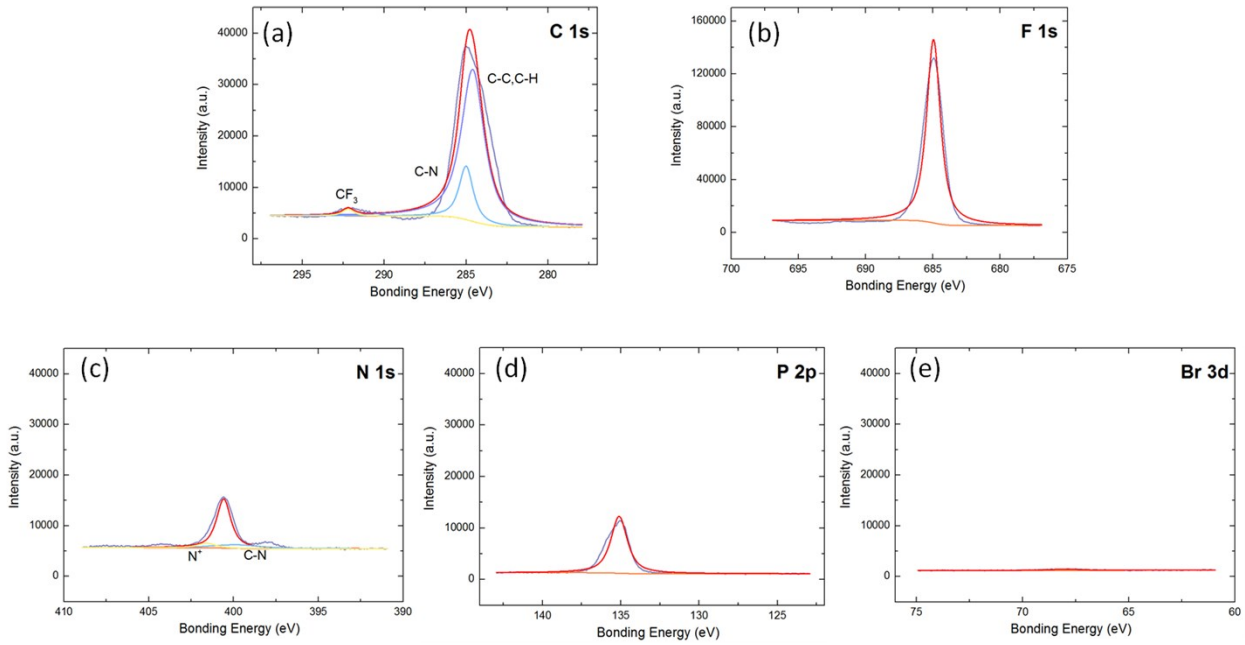


**Figure S14.** XPS spectra of $EV(PF_6)_2$ in (a) C 1s ; (b) F 1s; (c) N 1s; (d) P 2p and (e) Br 3d regions.