Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting information

$EMIMBF_4$–GBL binary electrolyte working at -70 ^{o}C and 3.7 V for a high performance graphene-based capacitor

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SI-1. Characterization methods

All NMR measurements were performed on a JNM-ECA600, Sodium 2,2-Dimethyl-2-silapentane-5-sulfonate (DSS) was dissolved in D_2O as external standard (δ_{DSS}=0). ATR-IR spectra were recorded at room temperature on a Nicolet 6700FTIR (resolution: 0.35 cm^{-1}) equipped with an ATR single reflection cell. The high-resolution scanning electron microscopy (SEM) observation was conducted on a JEOL, JSM-7401 at 3.0 kV. TEM experiment was performed on a high-resolution transmission electron microscope (JEOL, JEM-2010, exited at 120 kV). Brunauer–Emmett–Teller (BET) surface area was recorded in a Quantachrome automated surface area and porosity analyzer with N_2 as the adsorption gas. Differential scanning calorimetry (DSC) was obtained using a differential scanning calorimeter (DSC-60), from -80 to 35 °C at a heating rate of 10 °C min^{-1}. The viscosities of binary electrolytes were measured by an MCR301 rheometer (Anton Paar) using a conical geometry, shear rate was set at 100 s^{-1}. An FE28 conductivity meter (Mettler-Toledo) was used for conductivity measurement of the electrolytes.

SI-2. Relationship between the viscosity and ionic conductivity.

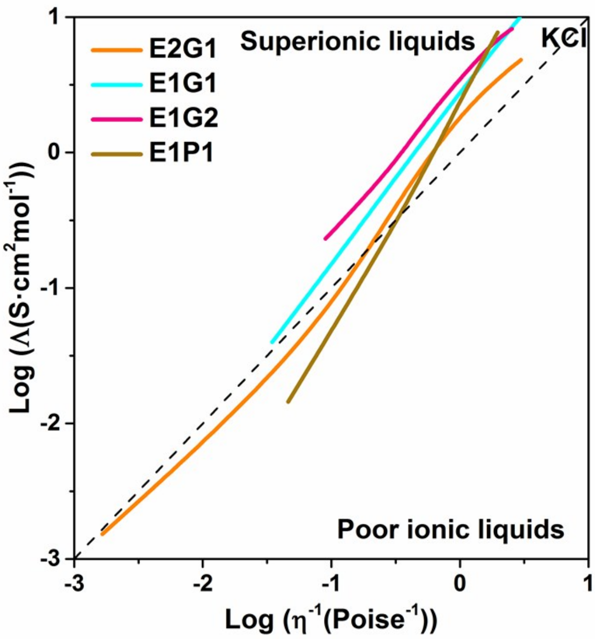

Figure S1. Walden plots of different binary electrolytes.

SI-3. NMR characterization

3.1 NMR spectra of different electrolytes

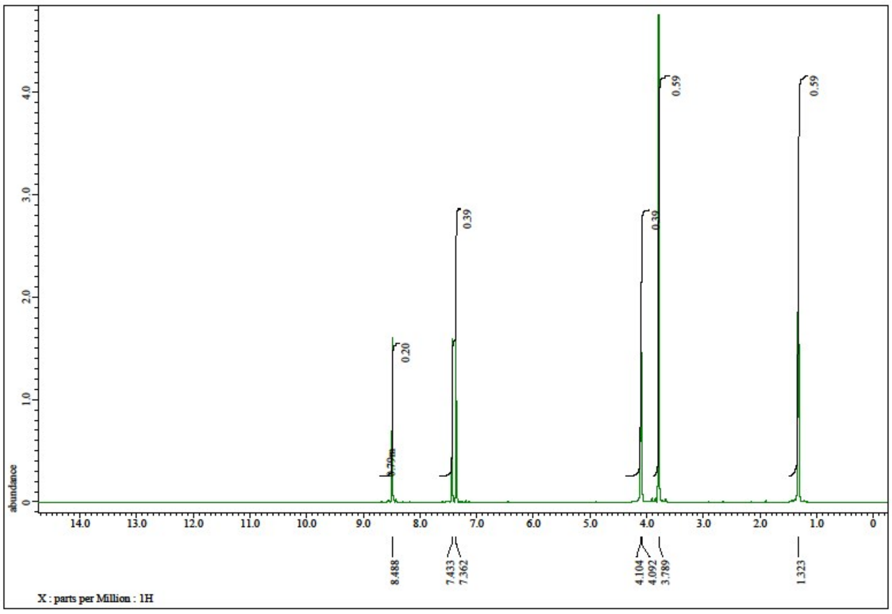

Figure S2-1. NMR pattern of pure $EMIMBF_4$(E).

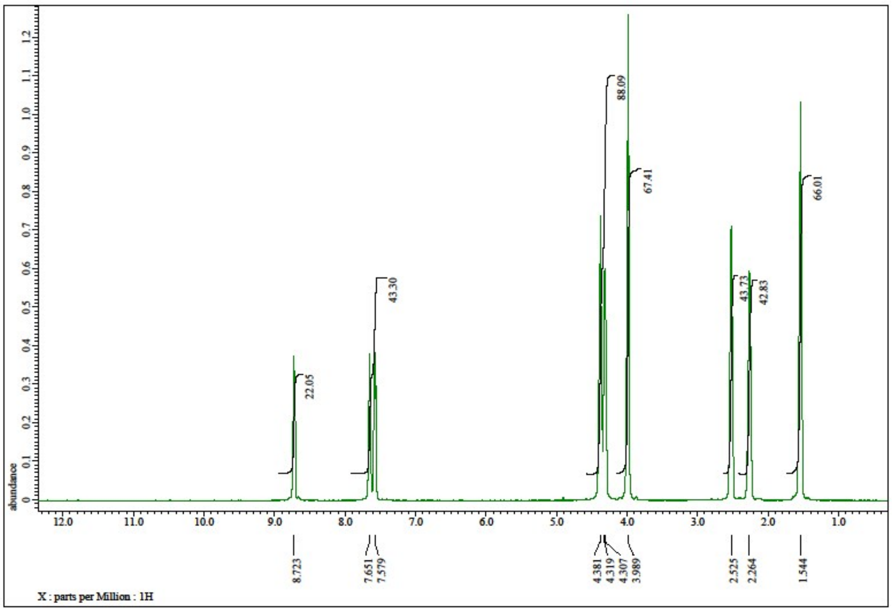

Figure S2-2. NMR pattern of E2G1.

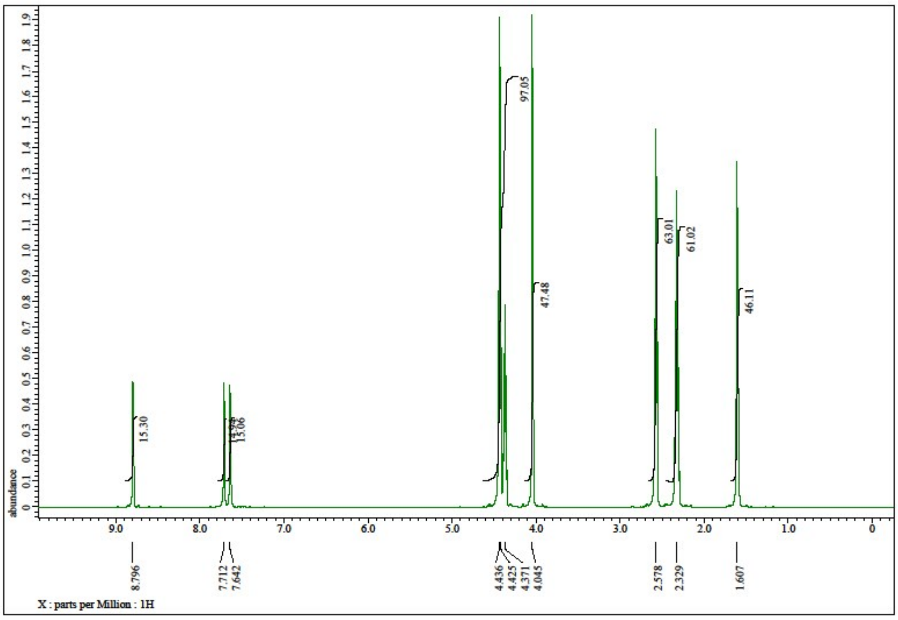

Figure S2-3. NMR pattern of E1G1.

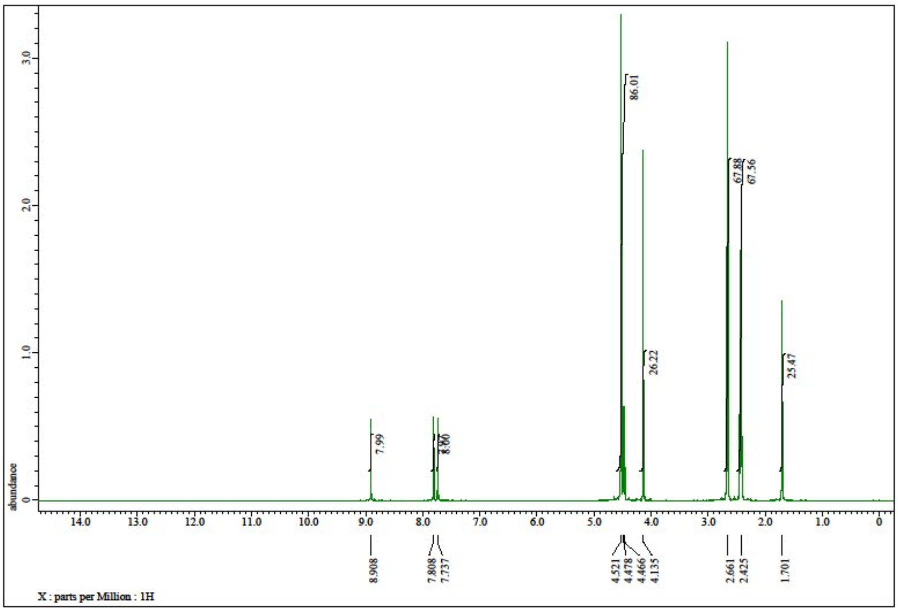

Figure S2-4. NMR pattern of E1G2.

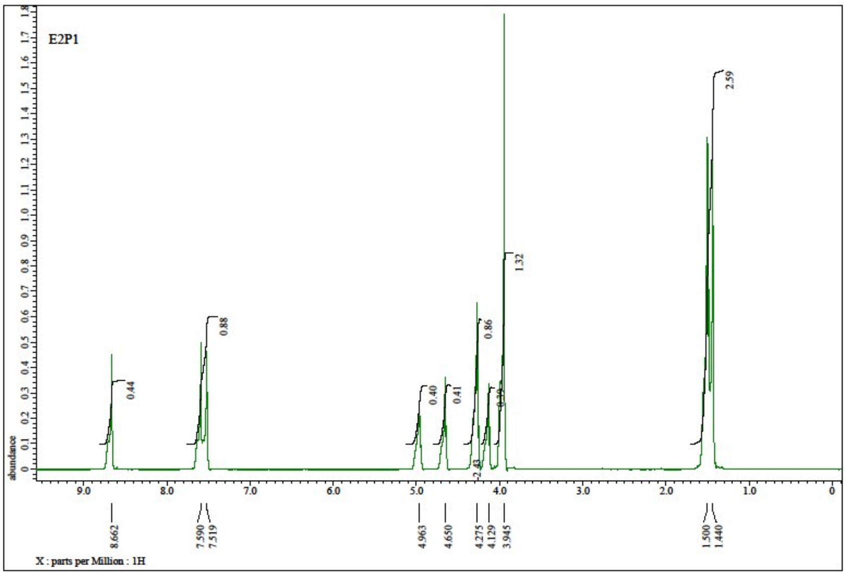

Figure S2-5. NMR pattern of E2P1.

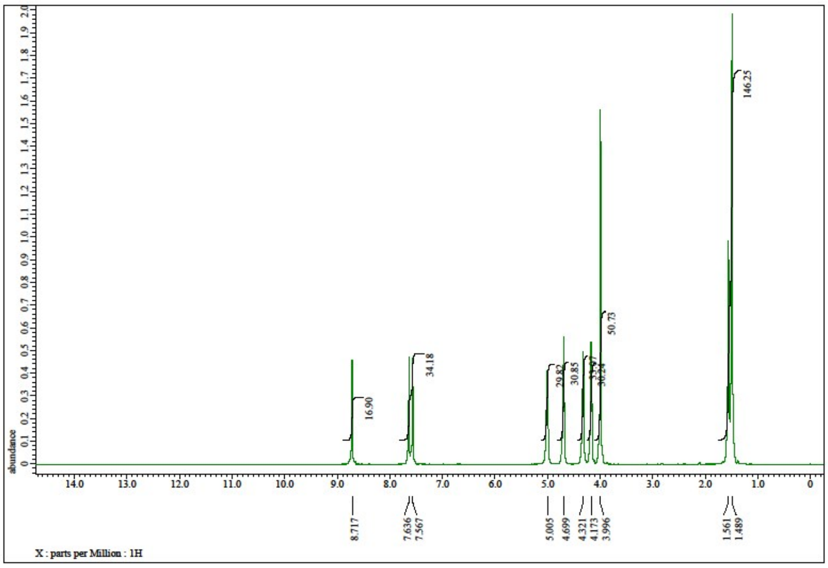

Figure S2-6. NMR pattern of E1P1.

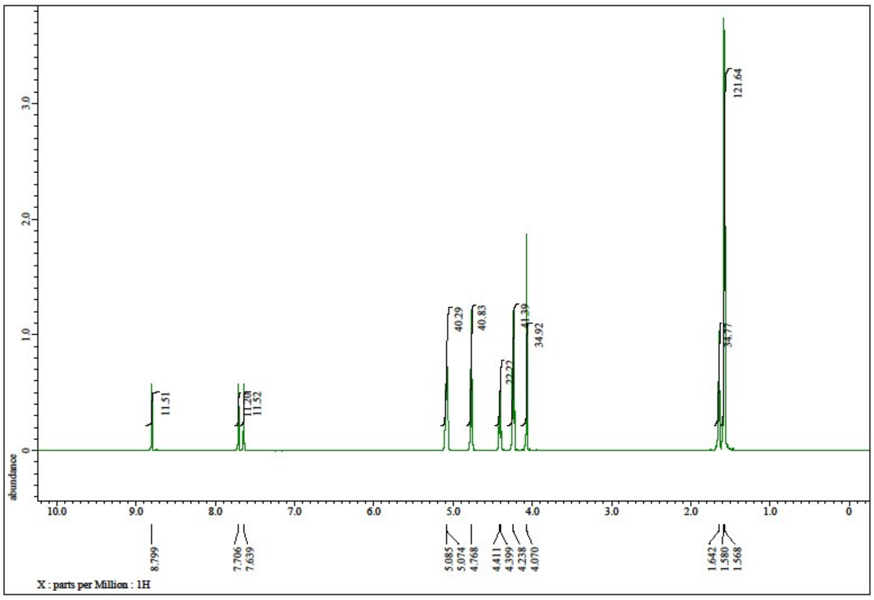

Figure S2-7. NMR pattern of E1P2.

SI-4. ATR-IR results

4.1 ATR-IR spectra

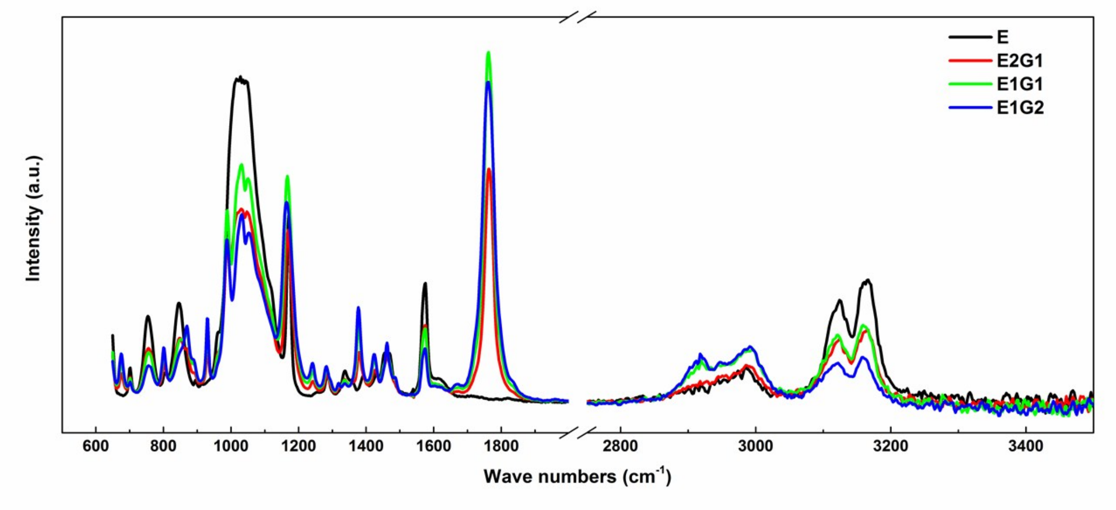

Figure S3-1. ATR-IR of $EMIMBF_4$ and binary electrolytes of $EMIMBF_4$-GBL.

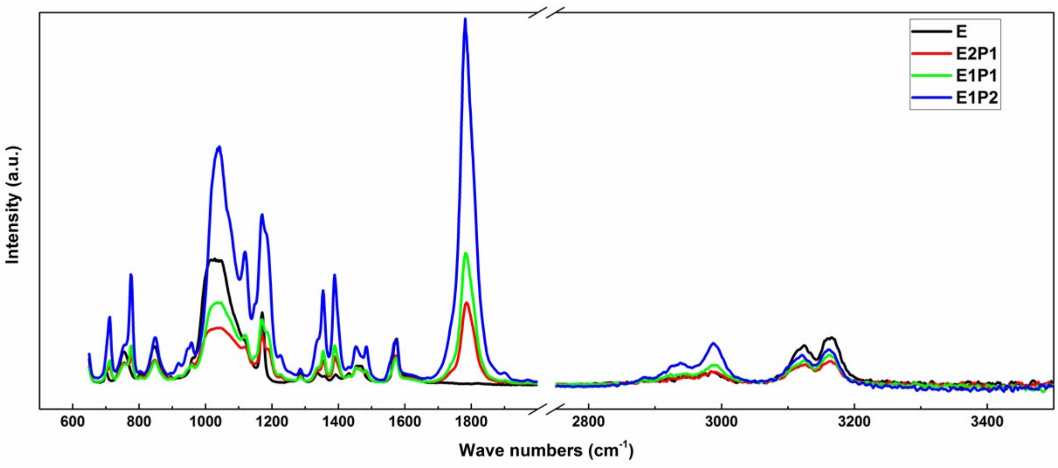

Figure S3-2. ATR-IR of $EMIMBF_4$ and binary electrolytes of $EMIMBF_4$-PC.

SI-5. EIS fittings by the model of Randles circuit.

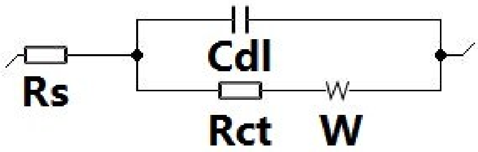

Figure S4. EIS fittings by the model of Randles circuit.

Table S1. Fitting data of resistance in different electrolytes at different temperatures.

Electrolyte	**Temperature (°C)**	**Rs (Ω)**	**Rct (Ω)**	**W ($\Omega\cdot s^{0.5}$)**
E	20	4.3	nearly disappeared	3.0
	10	10.3	0.7	10.0
	-10	18.5	0.9	15.1
E1G1	20	1.3	0.6	1.0
	-30	2.9	0.5	8.0
	-50	5.8	1.0	29.0
	-70	33.4	9.6	245.0

SI-6. Capacitance and energy density of GNF in E1G1.

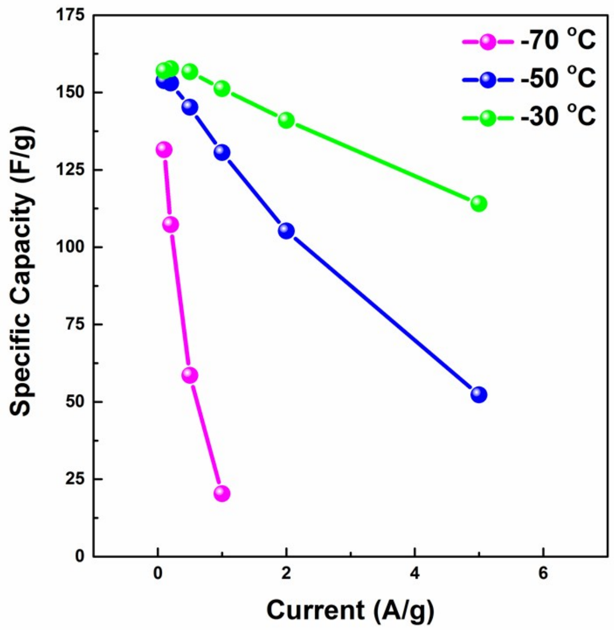

Figure S5-1. Specific capacitance of E1G1 EDLC at various current density.

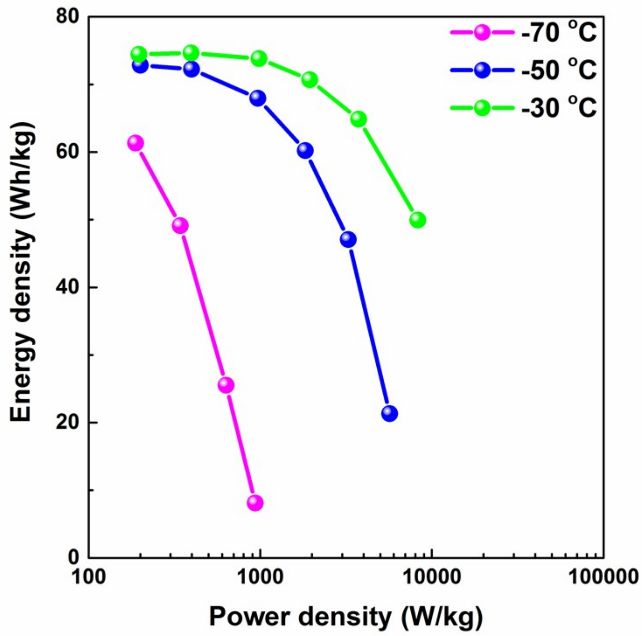

Figure S5-2. Energy density of E1G1 EDLC at different power density.

SI-7. Linear fitting of Lnσ with 1000/(T-T_g).

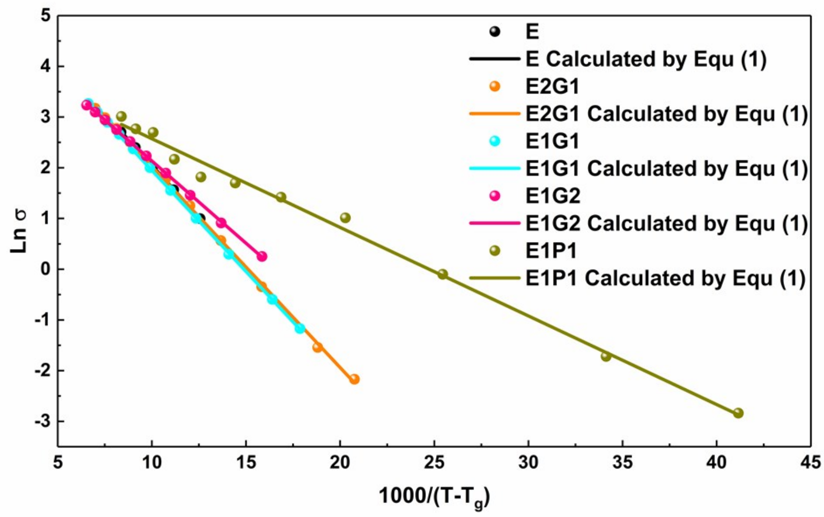

Figure S6. Fitting plot of conductivities of mixture electrolytes and pure $EMIMBF_4$.

SI-8. Electrochemical performance of previously reported low-temperature high-voltage electrolytes and EIG1。

Table S2. Electrochemical performance of previously reported low-temperature high-voltage electrolytes and EIG1.

Electrolyte	Electrode	Potential/V	$C_{electrode}$/F g^{-1}	E_{SC}/W h kg^{-1} a)	Temperature/°C	Current density/ scan rate	Ref
$EMIMBF_4$-GBL (E1G1)	GNF (1274 m^2 g^{-1})	3.7	157 153 131	73.8 72.3 61.3	-30 -50 -70	0.5 A g_{-1} 0.2 A g_{-1} 0.1 A g_{-1}	This work
$(PIP_{13}FSI)_{0.5}(PYR_{14}FSI)_{0.5}$	Reduced graphene film	3.5	135 125 96 55	57.4 53.1 40.8 23.4	0 -20 -30 -40	1 mV s^{-1}	29
$(PIP_{13}FSI)_{0.5}(PYR_{14}FSI)_{0.5}$	a-MEGO (~2000 m^2 g^{-1})	3.5 3	130 120 100	40.6 37.5 31.25	Room temperature -40 -50	20 mV s^{-1} 1 mV s^{-1}	30
$(BMIBF_4)_{0.5}(BMPBF_4)_{0.5}$	A-NPG	3.5	109 92	- 46.4	Room temperature -50	5 mV s^{-1}	31
$SBPBF_4$+AN/DBC-20%	AC	2.7	117 117 116 114	27.8 26.6 25.8 22.8	20 -40 -50 -60	0.1 A g^{-1}	32
$PYRNO_3$+GBL	AC(~1500 m^2 g^{-1})	2	129 119	17.4 16.5	20 -40	0.17 A g^{-1}	47
$DAIMBF_4$+PC	AC(~2400 m^2/g)	2.5	37 30	8.0 6.5	25 -30	0.32 A g^{-1}	48
PYR_{14}TFSI+BuCN	AC(~3000 m^2/g)	2.5	130 125	28.2 27.1	25 -20	0.5 A g^{-1} 0.2A g^{-1}	49

a)Calculated by formulas: $E_{capacitor}=C_{capacitor}U^2/2$; $C_{capacitor}=C/4$