

*Supplementary Information*

## All Solid Flexible Supercapacitor Operating at 4V with Cross-linked Polymer-Ionic Liquid Electrolyte

Yong-keon Ahn<sup>†a</sup>, Bokyung Kim<sup>†a</sup>, Jieun Ko<sup>a</sup>, Duck-Jea You<sup>a</sup>, Zhenxing Yin<sup>a</sup>, Hyunjin Kim<sup>a</sup>, Dalwoo Shin<sup>b</sup>, Sanghun Cho<sup>a</sup>, Jeeyoung Yoo<sup>\*a</sup> and Youn Sang Kim<sup>\*a,c</sup>

a. *Program in Nano Science and Technology, Graduate School of Convergence Science and Technology, Seoul National University, Seoul 151-744, Republic of Korea. E-mail:younskim@snu.ac.kr, jyoo78@snu.ac.kr*

b. *KOREA JCC CO. LTD. R&D Center 1163, Chungcheong-daero, Bugi-myeon,Cheongwon-gu, Chungcheongbuk-do, Republic of Korea*

c. *Advanced Institutes of Convergence Technology,864-1 Iui-dong, Yeongtong-gu, Suwon-si, Gyeonggi-do 443-270, Republic of Korea*

<sup>\*</sup> These authors contributed equally to this work.

## Abbreviations

Name	Abbreviation
poly-4-vninyphenol	P <sub>4</sub> VPh
cross-linked poly-4-vninyphenol	c-P <sub>4</sub> VPh
1-ethyl-3-methyl imidazolium bis(trifluoromethylsulfonyl)imide	EMITFSI
composite electrolyte consisting of EMITFSI and c-P <sub>4</sub> VPh	IL-CPX*
N-Methyl-2-pyrrolidone	NMP
Supercapacitor	SC
Symmetrical Supercapacitor with ILCPX	IL-CPX SC

\* x: weight ratio of EMITFSI based P<sub>4</sub>VPh weight

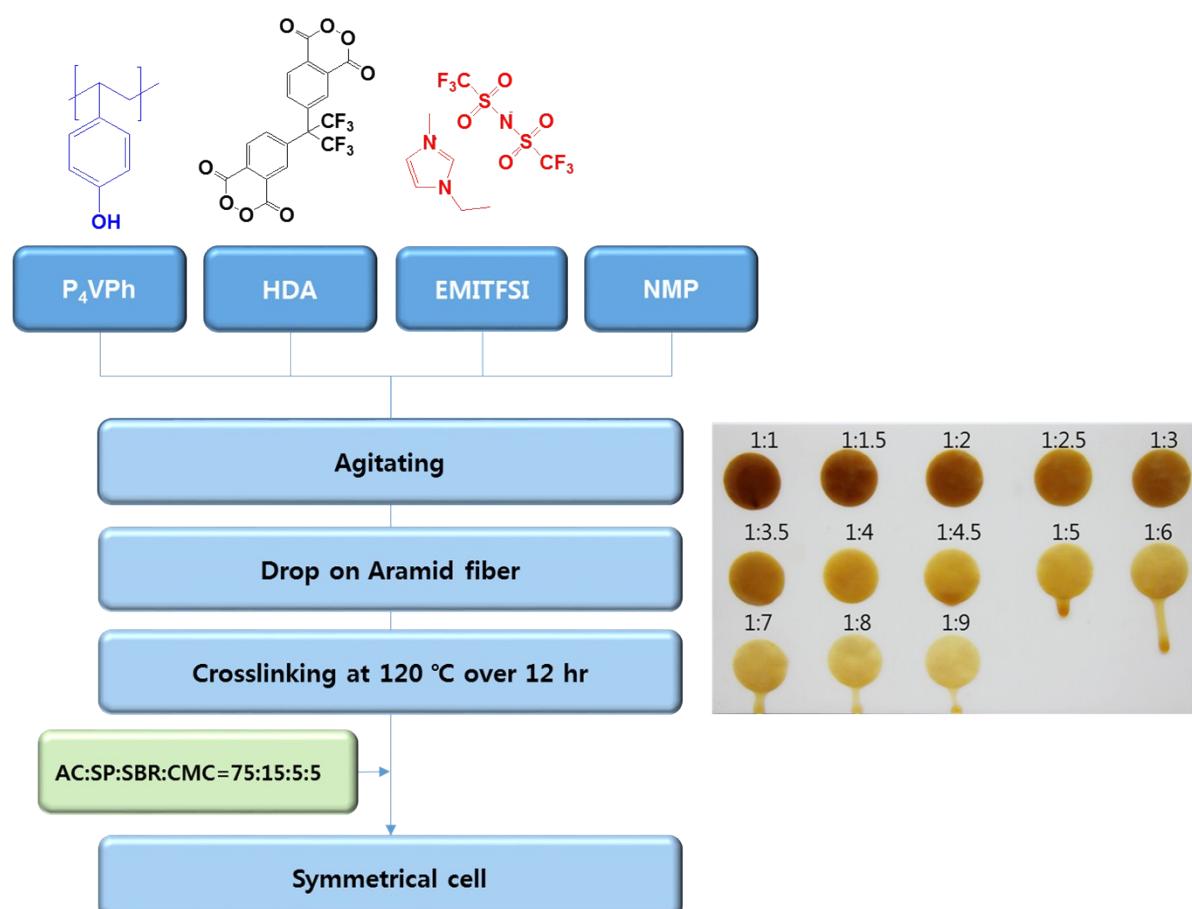


Figure S1. Preparation procedure all-solid state supercapacitor and features of synthesized IL-CPs with various EMITFSI composition.

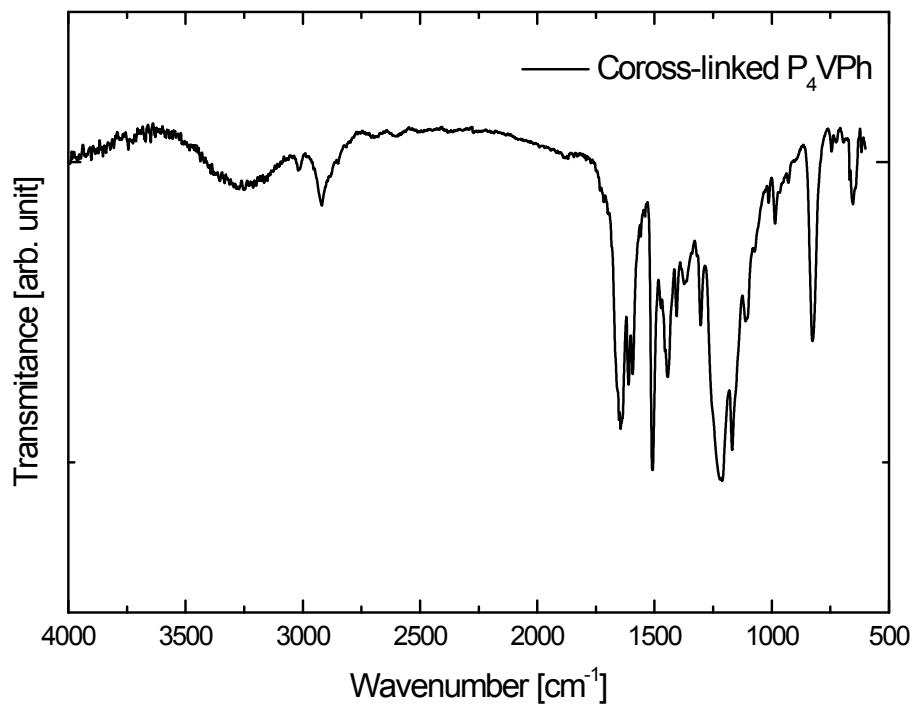


Figure S2. ATR-IR spectroscopy for cross-linked poly-4-vinylphenol

Table S1. Absorption peaks table of ATR FT-IR.

Functional groups of cation		Functional groups of anion	
<b>N=C-N stretching vibration</b>	1560-1520 cm <sup>-1</sup>	-CF <sub>3</sub> stretching	1350-1120 cm <sup>-1</sup>
<b>ring C=C vibration</b>	1605-1585 cm <sup>-1</sup>	asymmetric vibration of CF <sub>3</sub>	680-590 cm <sup>-1</sup> 555-505 cm <sup>-1</sup>
<b>C-H vibration for cyclic cations</b>	3172-3126 cm <sup>-1</sup>	symmetric vibration of CF <sub>3</sub>	600-540 cm <sup>-1</sup>

- (1) Socrates G. *Infrared Characteristic Group Frequencies: Tables and Charts*, 3rd Edition, Wiley & Sons: New York, 2004.
- (2) Kiefer, J.; Fries, J.; Leipertz, A. *Appl. Spectrosc.* 2007, **61**, 1306.

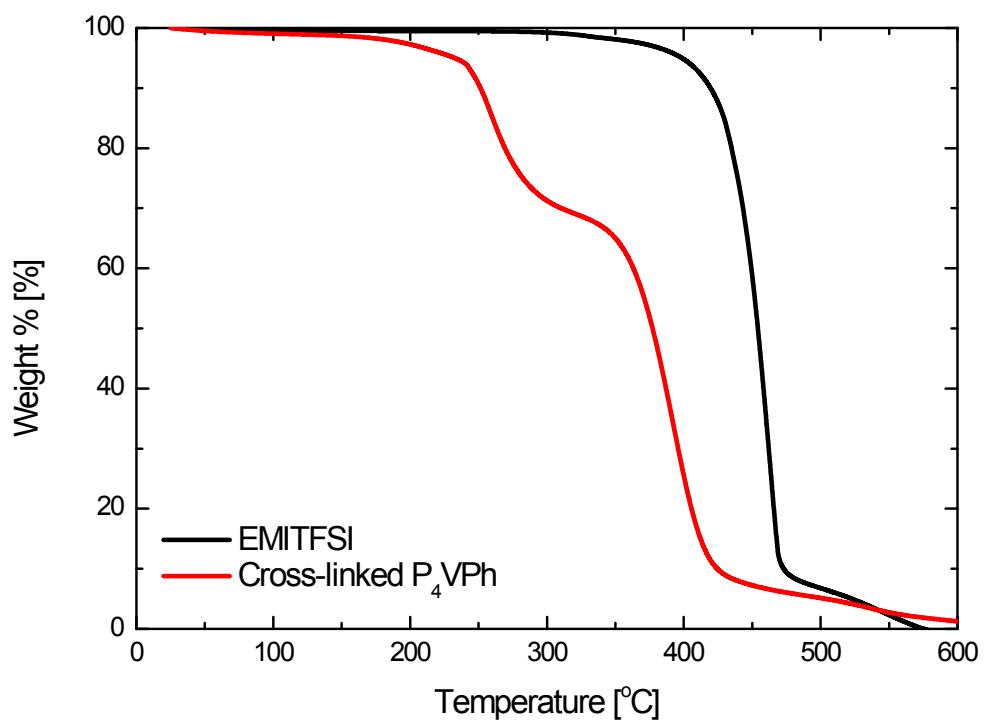


Figure S3. TGA for EMITFSI and  $P_4VPh$

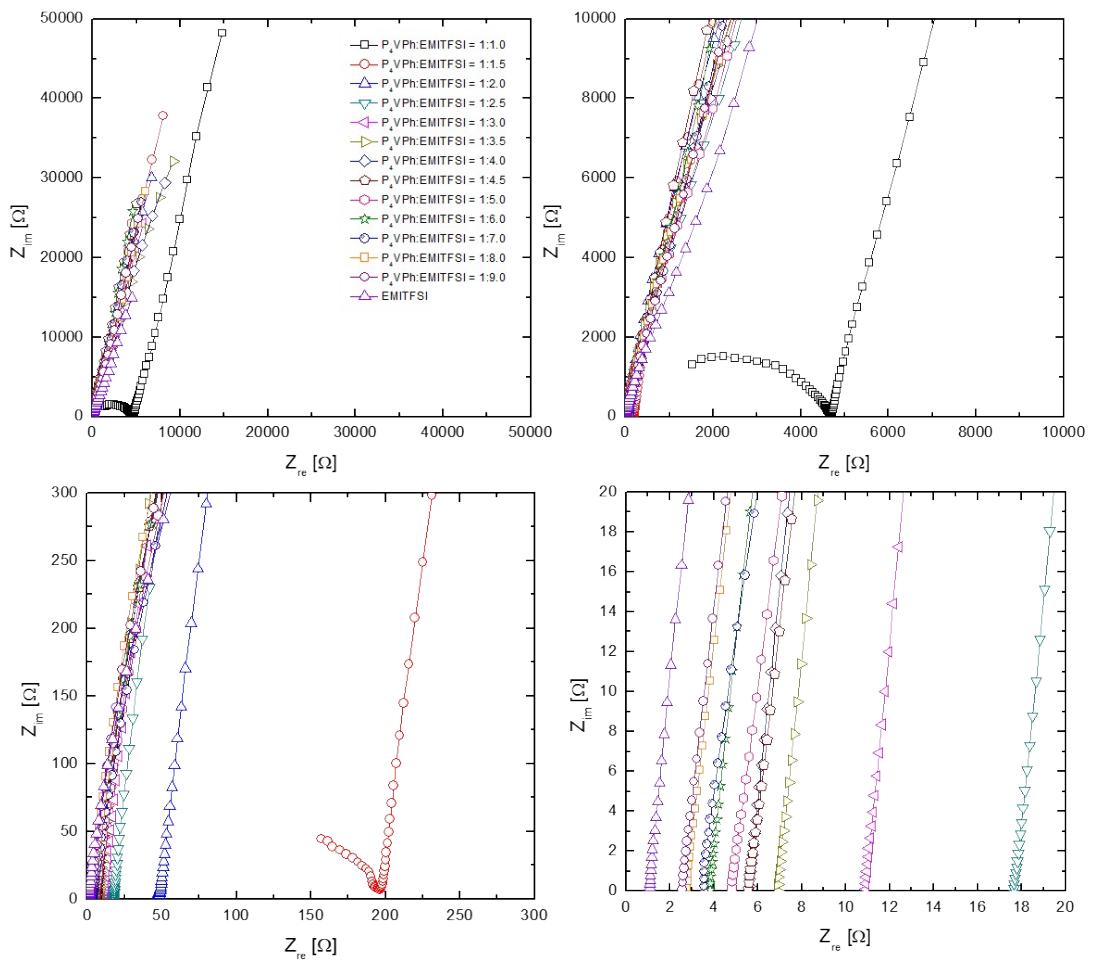


Figure S4. Electrochemical impedance spectroscopy of IL-CPs with stainless steel electrode.

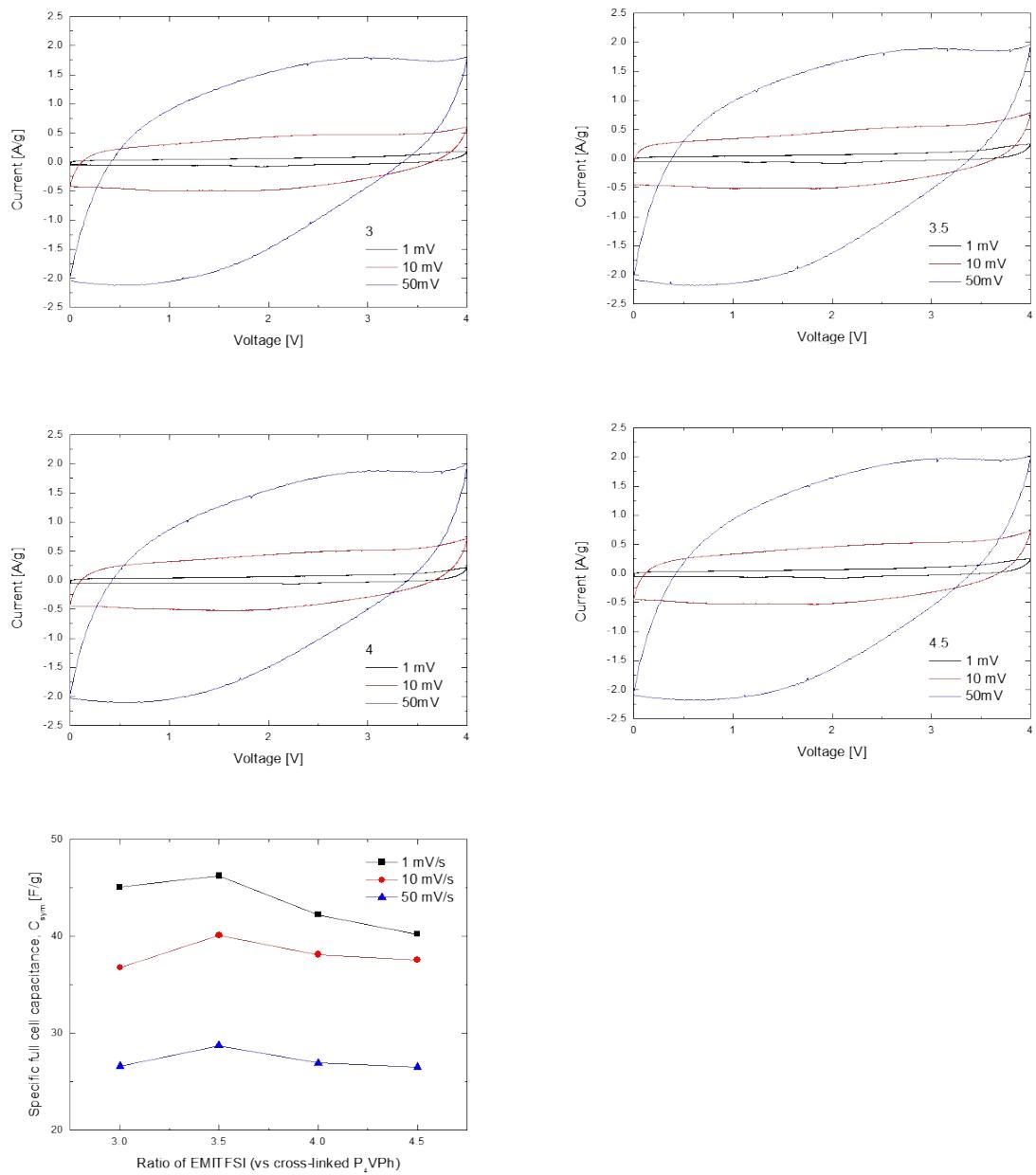


Figure S5. Cyclo-voltammograms and full cell capacitance of IC-CPx SCs with porous carbon symmetrical full cell ( $x=3, 3.5, 4$  and  $4.5$ )

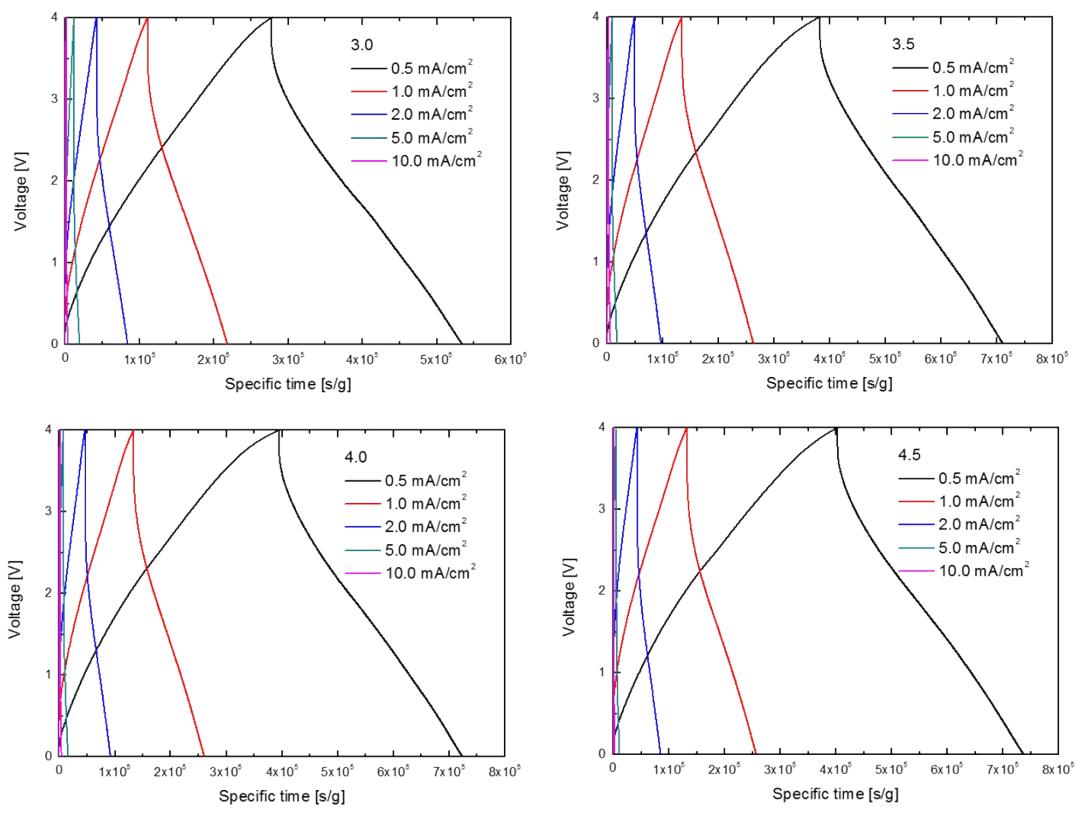


Figure S6. Galvanostatic charge-discharge profiles of IC-CP<sub>x</sub> SCs with porous carbon symmetrical full cell ( $x=3, 3.5, 4$  and  $4.5$ )

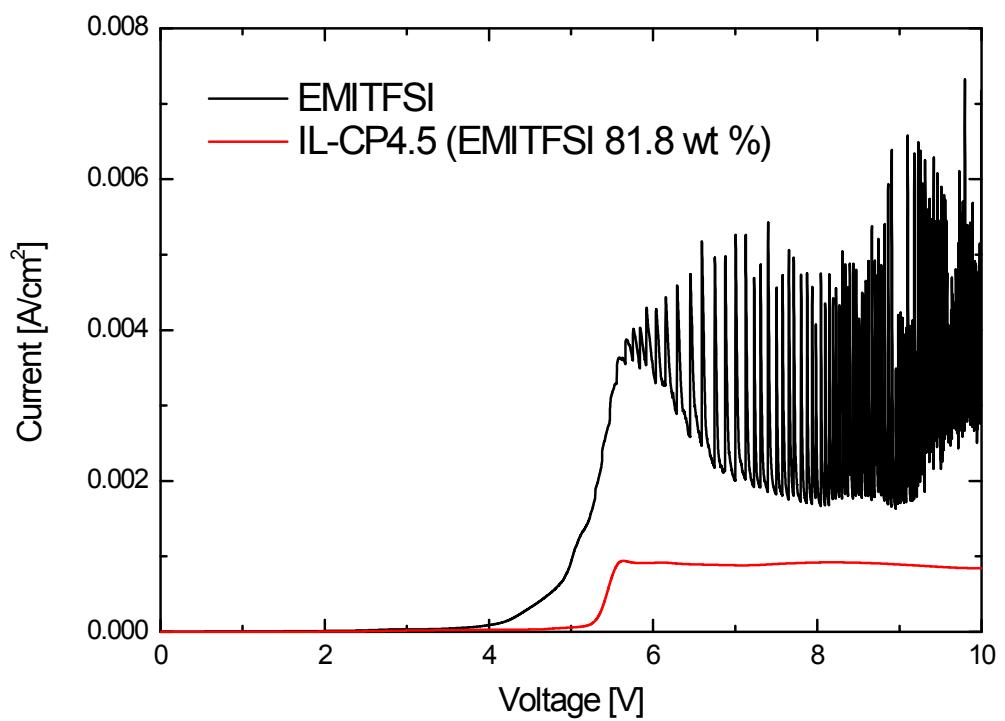


Figure S7. Linear sweep voltammogram of EMITFSI and IL-CP4.5 against Al foil in Al/Al cell (scan rate: 10 mV/s)

Table S1. Comparisons between prepared polymer electrolyte and other reported polymer electrolytes.

Ref.	electrolyte type	polymer	ionic liquid	solvent	ionic conductivity [mS cm <sup>-1</sup> ]	electrode material	rated voltage [V]	capacitance	method	energy density [Wh kg <sup>-1</sup> ]	additives
1	B	PAN	EMIBF4	none	15	AC	3	45 F g <sup>-1</sup>	CV (5 mV s <sup>-1</sup> )	-	sulpholane
2	A	PVdF-HFP	EMTf	EC-PC		AC	2	136 F g <sup>-1</sup>	Galvanostatic charge/discharge (1 mA cm <sup>-2</sup> )	18.8	-
3	B	PVdF-HFP	EMITFSI	none	1.5	AC	2~4	20 F g <sup>-1</sup>	Galvanostatic charge/discharge (10 mA cm <sup>-2</sup> )	20	zeolite
4	B	PEO	EMTf	none	0.1	MWCNT	2	3 F g <sup>-1</sup>	Galvanostatic charge/discharge (0.2 mA cm <sup>-2</sup> )	-	-
5	B	PEO	EMIHSO4	none	1.82	graphite	1.5	2 mF cm <sup>-2</sup>	CV (1 mV s <sup>-1</sup> )	-	silica
6	B	PEO	EMITri	none	16.2	AC	3	92 F g <sup>-1</sup>	Galvanostatic charge/discharge (2 mA cm <sup>-2</sup> )	0.01	-
7	B	PAN	BMIMTFSI	none	2.42	RGO	3	108 F g <sup>-1</sup>	Galvanostatic charge/discharge (2 mA cm <sup>-2</sup> )	30.51	-
8	A	PVdF-HFP	EMITf	EC-PC	1	AC	2	167 F g <sup>-1</sup>	Galvanostatic charge/discharge (1 mA g <sup>-1</sup> )	23.1	lithium triflate
9	A	pDADMATFSI	PYR <sub>14</sub> TFSI	none	0.5	AC	3.5	100 F g <sup>-1</sup>	Galvanostatic charge/discharge (1 mA cm <sup>-2</sup> )	37	-
<b>this work</b>	<b>B</b>	<b>c-P<sub>4</sub>VPh</b>	<b>ENITFSI</b>	<b>none</b>	<b>0.4</b>	porous carbon	<b>4</b>	<b>172 F g<sup>-1</sup></b>	Galvanostatic charge/discharge (1 mA cm <sup>-2</sup> )	<b>72.29</b>	-

Electrolyte type A : incorporation of an IL in a polymer matrix (The role of polymer is separator)

Electrolyte type B : polymerization of a monomer in an IL

Electrolyte type C : poly ionic liquid

Abbreviation in Table S1.

EMIBF<sub>4</sub> : 1-ethyl-3-methyl-imidazolium tetrafluoroborate

EMITf : 1-ethyl-3-methyl-imidazolium trifluoromethanesulfonate

EMHSO<sub>4</sub> : 1-ethyl-3-methylimidazolium hydrogensulfate

EMITri : 1-ethyl-3-methyl-imidazolium triflate

PYR<sub>14</sub>TFSI : N-methyl-N-butylpyrrolidinium bis(trifluoromethanesulfonyl) imide

PAN : poly acrylonitrile

PVdF-HFP : poly(vinylidenefluoride—hexafluoropropylene)

PEO : poly ethylene oxide

pDADMATFSI : poly(diallyldimethylammonium) bis(trifluoromethanesulfonyl)imide

EC : ethylene carbonate

PC : propylene carbonate

AC : activated carbon

MWCNT : multi-walled carbon nanotube

#### Reference

1. A. Lewandowski, A. Swiderska, *Solid State Ion.* 2003, **161**, 243.
2. G.P. Pandey, S.A. Hashmi, Y. Kumar, *Energy & Fuels* 2010, **24**, 6644
3. W. Lu, K. Henry, C. Turchi, J. Pellegrino, *J. Electrochem. Soc.* 2008, **155**, A361.
4. G.P. Pandey, Y. Kumar, S. a. Hashmi, *Solid State Ion.* 2011, **190**, 93.
5. S. Ketabi, X. Liu, Z. Le, K. Lian, *ECS Trans.* 2013, **50**, 411.
6. A. Lewandowski, A. Swiderska, *Appl. Phys. A Mater. Sci. Process.* 2006, **82**, 579.
7. P. Tamilarasan, S. Ramaprabhu, *Energy* 2013, **51**, 374.
8. Y. Kumar, G.P. Pandey, S. a. Hashmi, *J. Phys. Chem. C* 2012, **116**, 26118.
9. Girum Ayalneh Tiruye, David Munoz-Torrero, Jesus Palma, Marc Anderson, *Journal of Power Sources* 2015, **279**, 472