## **Supporting Information**

## Amphiphilic Block-Graft Copolymer Electrolyte: Synthesis, Nanostructure, and Use in Solid-state Flexible Supercapacitors

Jung Yup Lim<sup>1</sup>, Jin Kyu Kim<sup>1</sup>, Jung Min Lee<sup>2</sup>, Du Yeol Ryu<sup>1,\*</sup>, Jong Hak Kim<sup>1,\*</sup>

<sup>1</sup>Department of Chemical and Biomolecular Engineering, Yonsei University, 262 Seongsanno, Seodaemun-gu, Seoul 120-749, South Korea

<sup>2</sup> The 4th R&D Institute, Agency for Defense Development, Yuseong-gu, Daejeon, 305-152, South Korea

\* To whom correspondence should be addressed

E-mail: <u>dyryu@yonsei.ac.kr</u> (D. Y. Ryu) or <u>jonghak@yonsei.ac.kr</u> (J. H. Kim)

## Calculation of Flory–Huggins interaction parameter (χ)

The  $\chi$  values of PS-toluene, PS-chloroform, PB-toluene, and PB-chloroform pairs are available from literature (Adv. Funct. Mater. 2012, 22, 1759–1767; *Journal of Polymer Science: Polymer Physics Edition* **1983**, *21*, 1993–2001).

However, the  $\chi$  values of POEM-toluene and POEM-chloroform are not available. They were predicted using the equation below.

$$\chi_{PS} = \frac{V_s (\delta_S - \delta_P)^2}{RT} + 0.34$$

where:

 $\chi_{PS}$  is the Flory–Huggins interaction parameter between the polymer and the solvent,

R is the gas constant = 8.31 J/K mol,

T is the temperature = 298 K (25 °C),

V<sub>s</sub> is the molar volume of the solvent: toluene (106.8 ml/mol) and chloroform (80.7 ml/mol),

 $\delta_8$  is the solubility parameter of the solvent: toluene (18.2 J<sup>1/2</sup>cm<sup>-2/3</sup>) and chloroform (19.0 J<sup>1/2</sup>cm<sup>-2/3</sup>), and

 $\delta_P$  of POEM is approximately 19.8 J<sup>1/2</sup>cm<sup>-2/3</sup>.

Thus,

 $\chi_{\text{POEM-toluene}} = 106.8 \times (18.2 - 19.8)^2 / 8.31 \times 298 + 0.34 = 0.4245.$ 

and

 $\chi_{\text{POEM-chloroform}} = 80.7 \times (19.0-19.8)^2/8.31 \times 298 + 0.34 = 0.3608.$ 





Figure S2. Ionic conductivity of SBS-g-POEM block-graft copolymer electrolyte with different ratios of SBS to POEM at 25 °C. The LiTFSI concentration was fixed at 1.7% and PC was not added.





Figure S3. UTM curves of neat SBS and SBS-g-POEM (1:3) block-graft copolymer.

Figure S4. Ionic conductivity of SBS-g-POEM block-graft copolymer electrolyte with different POEM chain lengths at 25 °C. The LiTFSI concentration was fixed at 1.7 % while PC was not added.



Figure S5. Ionic conductivity of SBS-g-POEM block-graft copolymer electrolyte with different PC concentrations at 25 °C.



Figure S6. Ionic conductivity of SBS-g-POEM block-graft copolymer electrolyte with different LiTFSI concentrations at 25 °C. The PC concentration was fixed at 84%.



Figure S7. Ionic conductivity of neat SBS electrolyte and SBS-g-POEM block-graft copolymer electrolyte at 25 °C.



Figure S8. (a) Photos of electrolyte solutions of SBS (left) and SBS-g-POEM (right). (b) Photos of electrolyte films of SBS (left) and SBS-g-POEM (right) after evaporation of the solvent.



Figure S9. SEM images of activated carbon/carbon black/PVDF coated carbon paper substrates.



Figure S10. (a) CV curves of supercapacitor cell fabricated with SBS-g-POEM electrolyte at various bending angles. (b) photographs of SBS-g-POEM supercapacitor cell.



Figure S11. Galvanostatic charge-discharge curves of (a) supercapacitor cells at a constant current of 1 mA, (b) cell with  $PVA/H_3PO_4$  and (c) cell with SBS-g-POEM electrolyte at different currents of 0.5 mA, 1 mA and 5 mA.





