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## **Supplemental Information**

## Ultrahigh energy storage capability of trilayer

## polyetherimide/poly(vinylidenefluoride-trifluoroethylene-

## chlorofluoroethylene) with low loading of boron nitride nanosheets

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**Figure S1.** The structural characterizations of HBPE-*g*-HFBA copolymer: (a) <sup>1</sup>H NMR spectra, and (b) GPC trace.

Table S1. The grafting characterizations and GPC results of HBPE-g-HFBA

Sample	Volume	Yields	HFBA grafting <sup>a</sup>	Branching degree <sup>b</sup>	GPC °		
	(mL)	(g)	(mol%)	(/1000C) -	$\bar{M}_{n,PS}(kD)$	$\bar{M}_{\rm w,PS}$ (kD)	PDI
HBPE-g-HFBA	20	3.88	4.3	84.2	27.8	33.4	1.2

a: HFBA grafting obtained via <sup>1</sup>H NMR spectroscopy based on end-group analysis.

b: Degree of branching determined via <sup>1</sup>H NMR spectroscopy.

c: Number-averaged molecular weight  $(M_n)$ , weight-averaged molecular weight  $(M_w)$ 

and polydispersibility index (PDI) determined via GPC with polystyrene (PS) standards.



Figure S2. Cross-sectional SEM morphology of trilayer all-polymer LNL composite and BNNSs/P(VDF-TrFE-CFE) nanocomposite.

Table S2. Layer thickness and volume fraction of trilayer all-polymer LNL

composites with	different P	(VDF-TrFE-	·CFE) loadings
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	Concentration (wt%)	P(VDF-TrFE-CFE) thickness (μm)	PEI thickness (µm)	P(VDF-TrFE- CFE) (vol%)
L-6%N-L	6	2.4		33.3
L-8%N-L	8	4.7	2.4	49.5
L-10%N-L	10	7.2	2.4	60
L-12%N-L	12	9.4		66.2



Figure S3. (a)XRD patterns, and (b) TGA curves of BNNSs.



Figure S4. FTIR spectra of BNNSs/P(VDF-TrFE-CFE) nanocomposite.



**Figure S5.** Relative content of  $\beta$ -phase in BNNSs/P(VDF-TrFE-CFE) nanocomposite determined by FTIR spectroscopy.



Figure S6. Dynamic thermomechanical analysis of composite (a) storage modulus, (b) loss modulus, and (c) loss factor.



Figure S7. Two-parameter Weibull distribution plots of monolayer composite.



**Figure S8.** The unipolar P-E loops of monolayer films: (a) pure PEI, (b) pure P(VDF-TrFE-CFE), (c) 0.1% composite, (d) 0.3% composite, (e) 0.5% composite, and (f) 0.8% composite.



Figure S9. The P-E loops of all-polymer trilayer films: (a) L-6%N-L, (b) L-8%N-L,
(c) L-10%N-L, (d) L-12%N-L. The P-E loops of BNNSs/LNL films with different BNNSs mass fractions: (e) 0.1%, (f) 0.3%, (g) 0.5%, and (h) 0.8%.



Figure S10. Maximum electrical displacements of pure PEI, pure P(VDF-TrFE-CFE), monolayer and trilayer composites.



Figure S11. Energy density and charge-discharge efficiency of BNNSs/P(VDF-TrFE-CFE) composite.