Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.
This journal is © The Royal Society of Chemistry 2017

Electronic Supplementary Information

Three-Dimensional Graphene/Polyimide Composite-Derived Flexible High-Performance Organic Cathode for Rechargeable Lithium and Sodium Batteries

Yanshan Huang,[a] Ke Li,[a] Jingjing Liu,[a] Xing Zhong,[b] Xiangfeng Duan,[b] Muhammad Imran Shakir,[c] and Yuxi Xu*[a]

[a]State Key Laboratory of Molecular Engineering of Polymers, Department of Macromolecular Science, Fudan University, Shanghai 200433, China.

[b]Department of Chemistry and Biochemistry University of California, Los Angeles California 90095, USA.

[c]Sustainable Energy Technologies Center, College of Engineering, King Saud University, Riyadh 11421, Kingdom of Saudi Arabia.

E-mail: xuyuxi@fudan.edu.cn

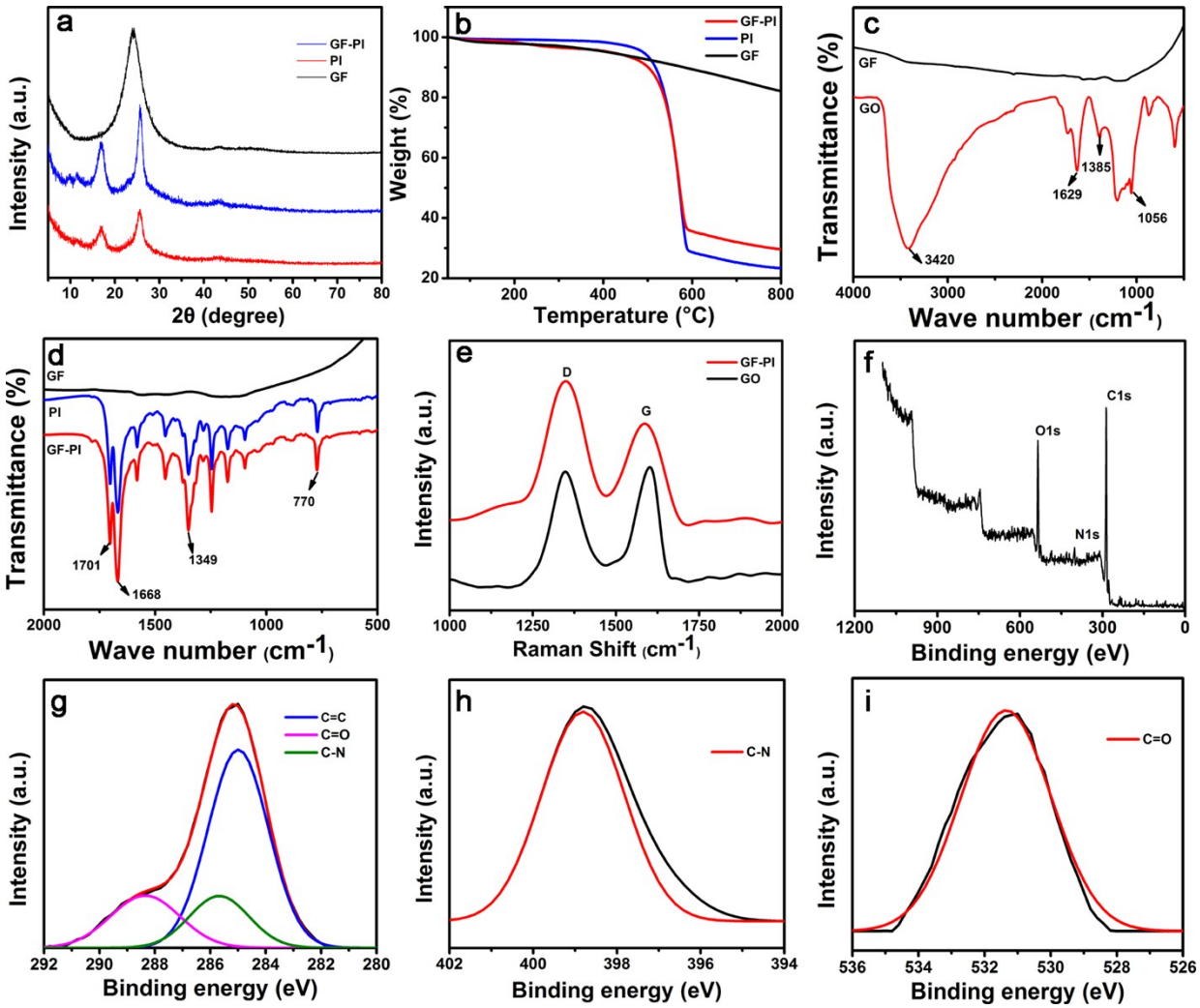


Fig. S1. (a) XRD patterns of GF-PI, PI and GF; (b) TGA of GF-PI, PI and GF, carried out in N_2 at a heating rate of 20 °C min^{-1}; FTIR of (c) GF, GO and (d) GF-PI, PI and GF; (e) Raman spectra of GF-PI and GO; (f) XPS spectrum of GF-PI and the corresponding high-resolution (g) C1s; (h) N1s and (i) O1s peaks.

The TGA curves indicates that the weight reduction in both GF-PI and GF occurred slightly quickly than pure PI between 200-400 °C, which is attributed to the decomposition of a small amount of residual oxygen functional groups on the GF surface. The content of PI is caculated as follow:

$PI\% = [(\text{pure } GF_{\text{Weight retention ratio}} - \text{composite}_{\text{Weight retention ratio}})/(GF_{\text{Weight retention ratio}} - \text{pure } PI_{\text{Weight retention ratio}})]*100\%$

There are peaks at 3430, 1625 and 1385 cm^{-1} that attributed to –OH, O-C-O, O-C-O, C=O in the FT-IR of GO samples.[S1] For GF, such peaks are almost reduced or disappeared, indicating that GO is almost reduced by solvothermal and annealing treatment (Fig. S1c). Both pure PI and GF-PI have a peak at 1340 cm^{-1}, which is ascribed to the stretching vibration of the C-N bond. The absorption peaks at 1703 and 1670 cm^{-1} are ascribed to the asymmetry and symmetry stretching vibration of C=O bond, respectively (Fig. S1d).[S2] This finding indicates that the carbonyl group is still maintained in the polymer structure. The GF-PI increased intensity ratio of D peak to G peak in comparison with GO in the Raman spectra, which is ascribed to the solvothermal and thermal reduction (Fig. S1e).

The chemical compositions of the GF-PI are examined by X-ray photoelectron spectroscopy (XPS, Fig. S1e-h). The component peak C_I at 284.6 eV, corresponding to C-C coordination, is attributed to the carbon atoms of graphitic carbon. The peak C_{II} at 285.3 eV is related to the C-N bond. Peak C_{III} at 288.1 eV corresponds to C=O double bond. The high resolution N 1s spectra shows a clear peak at 399.3 eV, corresponding to the N-C bond of pyridinic N from PI unit.[S3] The component peak of O 1s locates at 531.3 eV and corresponds to C=O bond.

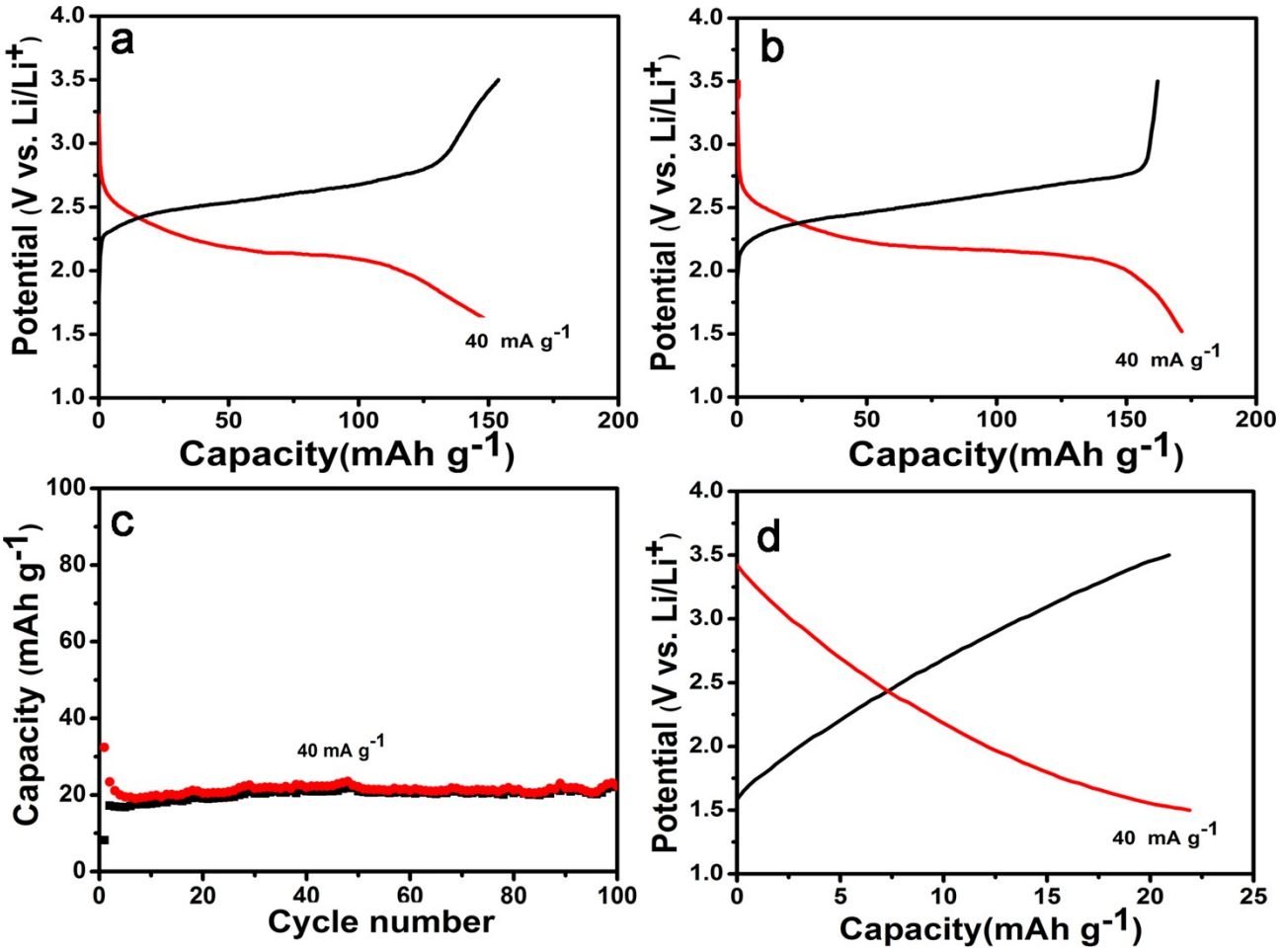


Fig. S2. (a, b) Charge/discharge profile of C-PI and G-PI at a current density of 40 mA g^{-1} , (c) Cycling performance and (d) charge/discharge profile of pure GF at a current density of 40 mA g^{-1} for LIBs,

Calculation of the utilization ratio of PI in the GF-PI composites is as follows:

$$utilization\ ratio = \frac{C_{composites} - C_{GF} \times P_{GF}}{(1 - P_{GF}) \times C_{polymer,\ theoretical}}$$

Where C is discharge capacity of GF-PI and P is weight percentage in the composite.

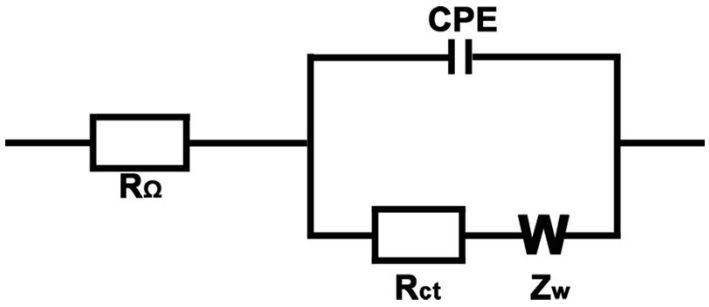


Fig. S3. Equivalent circuit for GF-PI, G-PI and C-PI electrodes

Table S1. The resistance of C-PI, G-PI and GF-PI electrodes

samples	R_Ω (Ω)	R_{ct} (Ω)
C-PI	3.5	279.2
G-PI	2.5	184.8
GF-PI	2.2	66.5
GF-PI after 600 cycles	2.2	48.3

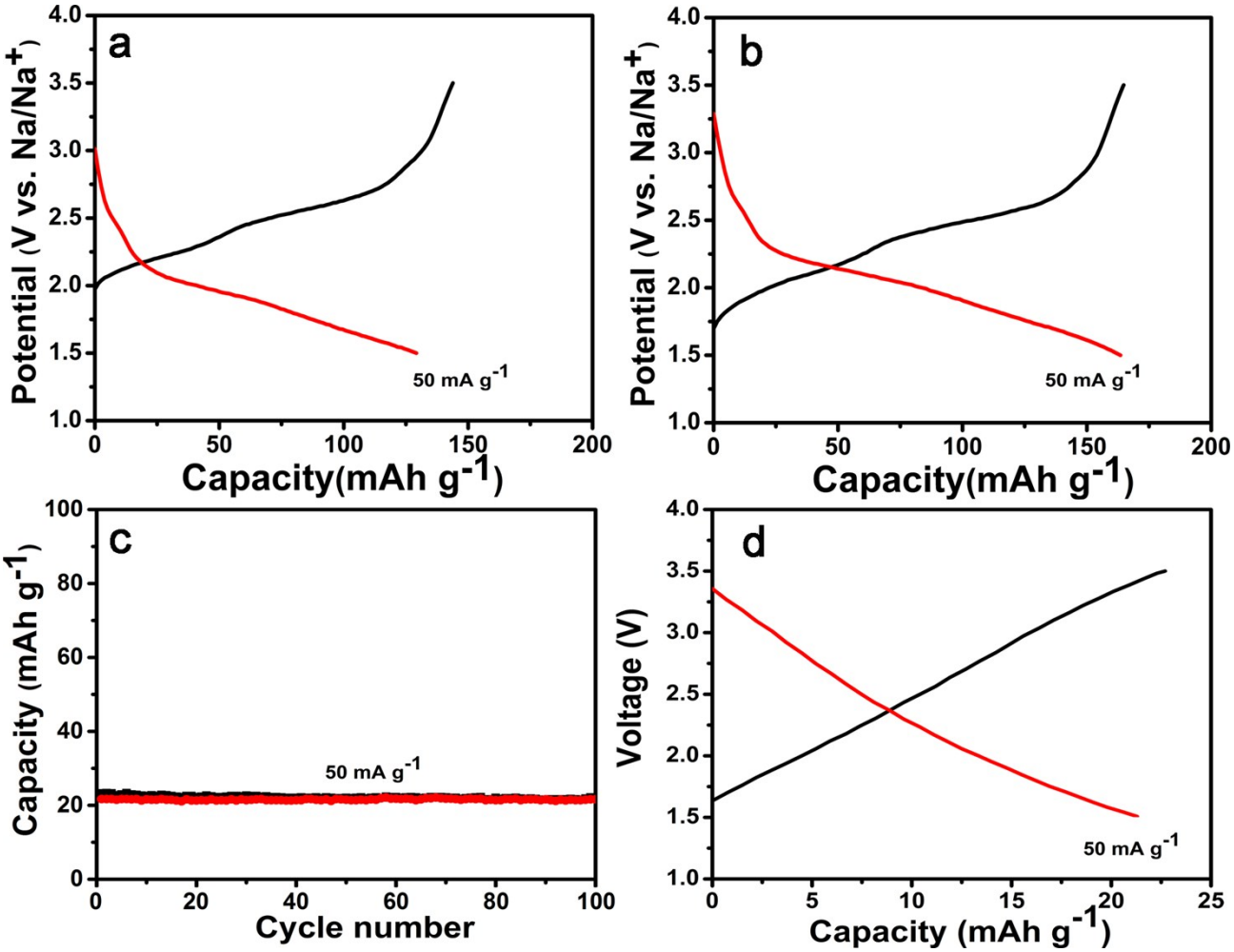
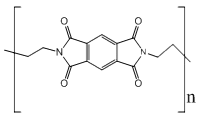
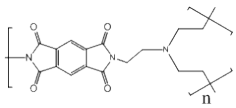
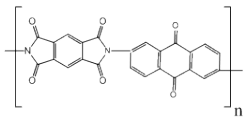
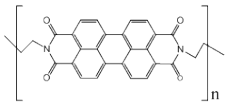
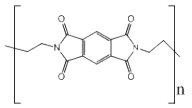


Fig. S4. (a, b) Charge/discharge profile of C-PI and G-PI at a current density of 50 mA g^{-1} , (c) Cycling performance and (d) charge/discharge profile of pure GF at a current density of 50 mA g^{-1} for SIB in the voltage range of 1.5–3.5 V.

Table S2. Electrochemical performance of typical PI-based cathode materials for LIB

Materials	Ratio of active materials	Current density (mA g^{-1})	Initial capacity（mAh g^{-1}）	High current density (mA g^{-1})	Capacity (mAh g^{-1})	Capacity Retention (Cycles)	Current density (mA g^{-1})	Ref.
PI/SWNT (PMDA/EDA) 	<100%	44	226 N. A. **(by electrode)**	8860	120 N. A. **(by electrode)**	85% (200)	221	*Adv. Mater.*, 2014, 26, 3338
PMTA/SWCNT (PMDA/TDA) 	65%	38	160 104 **(by electrode)**	3830	74 48 **(by electrode)**	87% (200)	191	*Adv. Mater.* 2015, 27, 6504
PMAQ–SWNT (PMDA/AQ) 	100%	44	190 **(by electrode)**	4180	120 **(by electrode)**	91% (300)	221	*JMC.A* 2016, 4, 2115
PI/CNT (PTCDA/EDA) 	85%	27	125 106 **(by electrode)**	546	115 98 **(by electrode)**	74% (300)	100	*JMC.A* 2013, 1, 6366
PI-FLEG (PMDA/EDA) 	65%	44	177 115 **(by electrode)**	2200	38 25 **(by electrode)**	80% (221)	200	*RSC Adv.* 2016, 6, 33287
3D-RGO/PI (PMDA/EDA)	80%	44	175 140	886	101 81	82% (150)	221	*JMC.A* 2014, 2, 10842

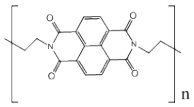
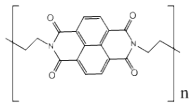
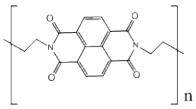
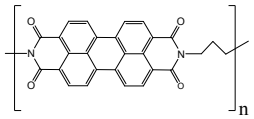
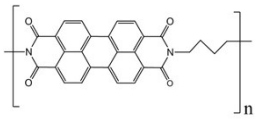
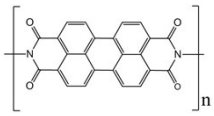
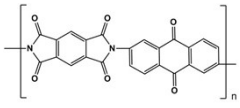
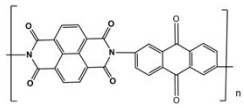
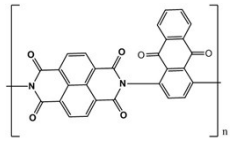
			(by electrode)		**(by electrode)**			
PI-FGS-b (NTCDA/EDA) 	60%	37	205 123 **(by electrode)**	3670	135 81 **(by electrode)**	N. A.		*Nano Lett.* 2012, 12, 2205
PI-4 (NTCDA/EDA) 	60%	18	188 112 **(by electrode)**	74	173 104 **(by electrode)**	95% (100)	74	*Angew. Chem. Int. Ed.* 2010, 49, 8444
GF-PI (NTCDA/EDA) 	100%	40	240 **(by electrode)**	4000	102 **(by electrode)**	91% （100） 81% (600)	50 100	This work

Table S3. Electrochemical performance of typical PI-based cathode materials for SIB

Materials Precursor, structure	Ratio of active materials	Current density (mA g^{-1})	Initial capacity（mAh g^{-1}）	High current density (mA g^{-1})	Capacity (mAh g^{-1})	Capacity Retention (Cycles)	Current density (mA g^{-1})	Ref.
PMDA-based PI2 (PMDA/EDA) 	60%	25	124 74.4 **(by electrode)**	N. A.		40% (62)	200	*Adv. Energy Mater.* 2014, 4, 1301651

NTCDA-based PI2 (NTCDA/EDA) 	60%	25	132 79.2 **(by electrode)**	N. A.		65% (100)	200	*Adv. Energy Mater.* 2014, 4, 1301651
PTCDA-based PI2 (PTCDA/EDA) 	30%	25	150 45 **(by electrode)**	10000	75 22.5	87.5% (5000)	200	*Adv. Energy Mater.* 2014, 4, 1301651
PTCDA-based PI3 (PTCDA/1,3-propanediamine) 	60%	25	116 69.6 **(by electrode)**	N. A.	N. A.	N. A.	N. A.	*Adv. Energy Mater.* 2014, 4, 1301651
PTCDA-based PI4 (PTCDA/1,4-diaminobutane) 	60%	25	100 60 **(by electrode)**	N. A.	N. A.	N. A.	N. A.	*Adv. Energy Mater.* 2014, 4, 1301651
PI (PTCDA/$N_2H_4{\cdot}H_2O$) 	60%	100	126 75.6 **(by electrode)**	800	90 54	90% (50)	100	*J. Mater. Chem. A* 2015, 3, 10453
PAQI PMDA/26DAAQ 	40%	50	162 64.8 **(by electrode)**	N. A.	N. A.	95% (150)	50	*Electro. Commun.* 2015, 60, 117

PAQI NTCDA/26DAAQ 	40%	50	179 71.6 **(by electrode)**	N. A.	N. A.	95% (150)	50	*Electro. Commun.* 2015, 60, 117
PAQI NTCDA/14DAAQ 	40%	50	220 88 **(by electrode)**	1000	75 30 **(by electrode)**	82% (150)	50	*J. Mater. Chem. A* 2016, 4, 11491
PNTCDA NTCDA/EDA 	30%	140	140 42 **(by electrode)**	2520	84 25.2 **(by electrode)**	68% (500)	140	*RSC Adv.* 2014, 4, 25369
GF-PI (NTCDA/EDA) 	100%	50	213 **(by electrode)**	1000	116 **(by electrode)**	~100% (150) 80.4%（1000）	50 1000	This work

Reference

(S1) Huang, Y.; Wu, D.; Wang, J.; Han, S.; Lv, L.; Zhang, F.; Feng, X. *Small* 2014, **10**, 2226-2232.

(S2) Chen, L.; Li, W.; Wang, Y.; Wang, C.; Xia, Y. *RSC Adv.* 2014, **4**, 25369-25373.

(S3) Ma, L.; Niu, H.; Cai, J.; Zhao, P.; Wang, C.; Bai, X.; Lian, Y.; Wang, W. *Carbon* 2014, **67**, 488-499.