

Supporting Information for:

## Layered polyaniline/graphene film from sandwich-structured polyaniline/graphene/polyaniline nanosheets for high-performance pseudosupercapacitors

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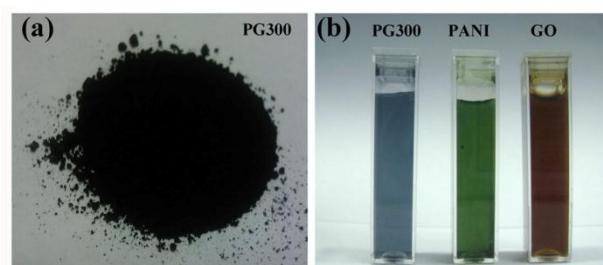
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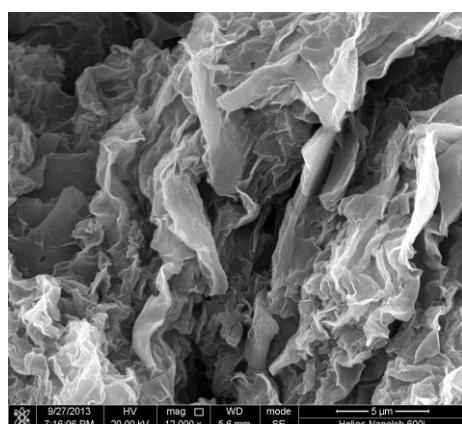
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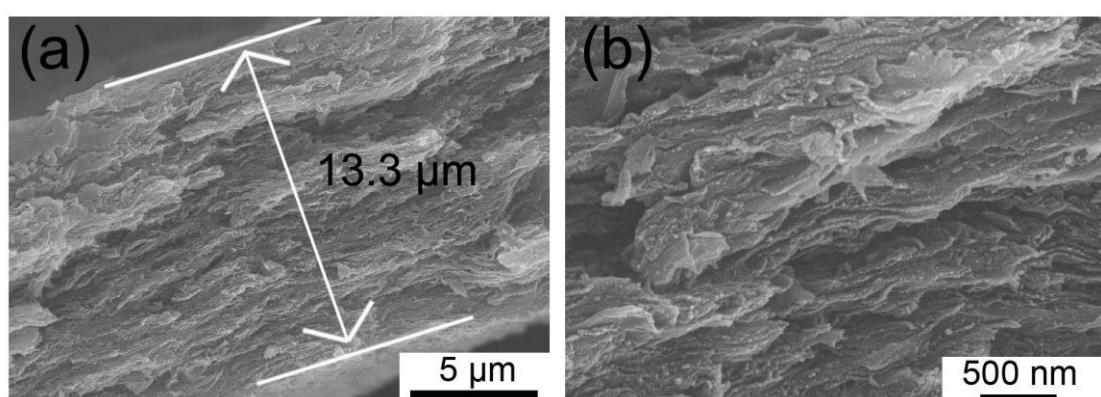
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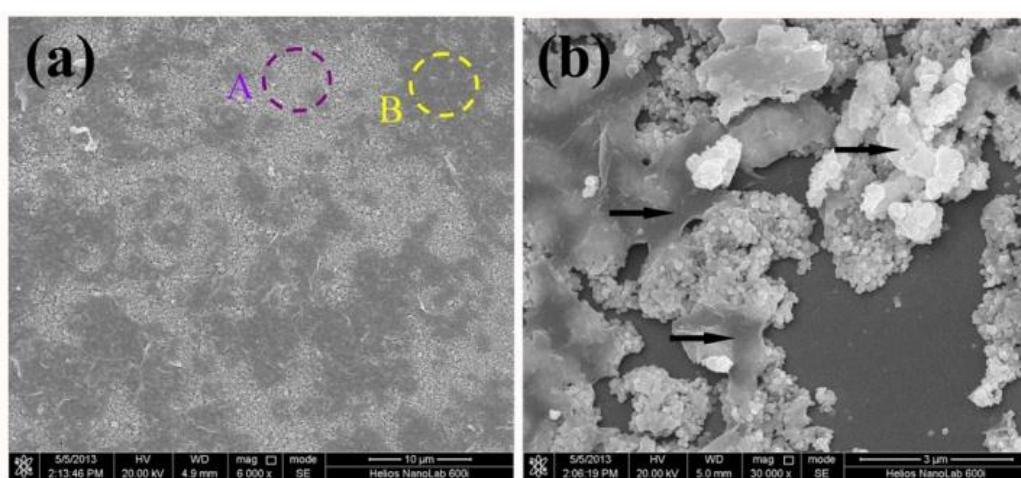
**Fig. S1.** (a) A digital picture of the PG300; (b) Dispersion stability of PG300, pure PANI and GO in water.



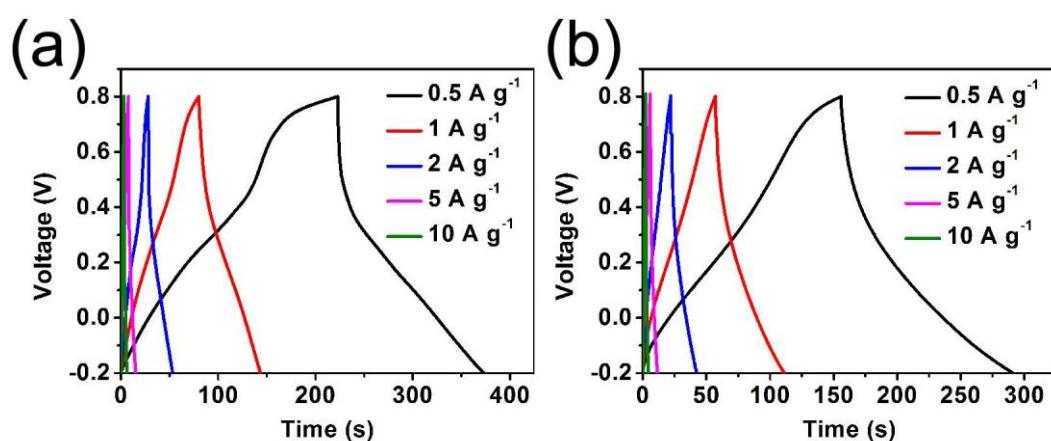
**Fig. S2.** SEM image of PG300.



**Fig. S3.** Cross-section SEM image of PG300 at low magnification (a) and high magnification (b).



**Fig. S4.** SEM images of the as-deposited film from PG30 with various magnifications. Dotted line circle “A” exhibits the PANI particles, while the dotted line circle “B” and the nanosheets arrows pointed show the PANI/graphene/PANI sheets.



**Fig. S5.** Galvanostatic charge–discharge curves of PANI (a) and PG30 (b) at different current densities.

**Table S1** List of specific capacitance values of various graphene/PANI hybrid electrodes from recent literatures.

Binder-free	Preparation method	Specific capacitance	Measurement condition	Reference (year)
Yes (our work)	Electrophoretic deposition	384 F g <sup>-1</sup>	0.5 A g <sup>-1</sup>	(2013)
		312 F g <sup>-1</sup>	1 A g <sup>-1</sup>	
		324 F g <sup>-1</sup>	At 20 mV s <sup>-1</sup> by CV	
Yes	Layer-by-layer (LbL) assembly	375.2 F g <sup>-1</sup>	At 0.5 A g <sup>-1</sup>	[1] (2012)
		ca. 225 F g <sup>-1</sup> (*)	At 1 A g <sup>-1</sup>	
	Electropolymerization	233 F g <sup>-1</sup>	At 20 mV s <sup>-1</sup> by CV	[2] (2009)
	Vacuum filtration	210 F g <sup>-1</sup>	At 0.3 A g <sup>-1</sup>	[3] (2010)
	Electropolymerization	763 F g <sup>-1</sup>	At 1 A g <sup>-1</sup>	[4] (2013)
	Chemical polymerization	250 F g <sup>-1</sup>	At 100 mV s <sup>-1</sup> by CV	[5] (2012)
	Vacuum filtration	301 F g <sup>-1</sup>	At 0.5 A g <sup>-1</sup>	[6] (2011)
	Electropolymerization	202 F g <sup>-1</sup> (**)	At 0.5 A g <sup>-1</sup>	[7] (2013)
		181 F g <sup>-1</sup> (**)	At 1 A g <sup>-1</sup>	
No	Chemical polymerization	385 F g <sup>-1</sup>	At 0.5 A g <sup>-1</sup>	[8] (2013)
		362 F g <sup>-1</sup>	At 1 A g <sup>-1</sup>	
	Electrospun	267 F g <sup>-1</sup> (***)	At 0.35 A g <sup>-1</sup>	[9] (2013)
	Chemical polymerization	261 F g <sup>-1</sup>	At 0.5 A g <sup>-1</sup>	[10] (2010)
		ca. 243 F g <sup>-1</sup> (*)	At 1 A g <sup>-1</sup>	
	Chemical polymerization	361 F g <sup>-1</sup>	At 0.3 A g <sup>-1</sup>	[11] (2012)
		349 F g <sup>-1</sup>	At 0.5 A g <sup>-1</sup>	
		323 F g <sup>-1</sup>	At 1 A g <sup>-1</sup>	
	Chemical polymerization	890 F g <sup>-1</sup> (**)	At 0.5 A g <sup>-1</sup>	[12] (2013)
	Chemical polymerization	497 F g <sup>-1</sup>	At 0.5 A g <sup>-1</sup>	[13] (2012)

**Note:**

(\*): Calculated from the figure of “Variation of the specific capacitance with current density” in the literature, because the authors did not show the exact number in their publication.

(\*\*): The composites were graphene oxide/carbon nanotube/polyaniline composites.

(\*\*\*): The active materials were pure PANI.

**Reference:**

- [1] T. Lee, T. Yun, B. Park, B. Sharma, H.-K. Song and B.-S. Kim, *J. Mater. Chem.*, 2012, **22**, 21092-21099.
- [2] D. W. Wang, F. Li, J. Zhao, W. Ren, Z. G. Chen, J. Tan, Z. S. Wu, I. Gentle, G. Q. Lu and H. M. Cheng, *ACS Nano*, 2009, **3**, 1745-1752.
- [3] Q. Wu, Y. X. Xu, Z. Y. Yao, A. R. Liu and G. Q. Shi, *ACS Nano*, 2010, **4**, 1963-1970.
- [4] H. P. Cong, X. C. Ren, P. Wang and S. H. Yu, *Energy Environ. Sci.*, 2013, **6**, 1185–1191.

- [5] N. A. Kumar, H. Choi, Y. R. Shin, D. W. Chang, L. Dai and J. Baek, *ACS Nano*, 2012, **6**, 1715-1723.
- [6] S. Liu, X. H. Liu, Z. P. Li, S. R. Yang and J. Q. Wang, *New J. Chem.*, 2011, **35**, 369-374.
- [7] Z.-D. Huang, R. Liang, B. Zhang, Y.-B. He and J.-K. Kim, *Compos. Sci. Technol.*, 2013, **88**, 126-133.
- [8] Y. N. Meng, K. Wang, Y. J. Zhang and Z. X. Wei, *Adv. Mater.*, 2013, **25**, 6985-6990.
- [9] S. Chaudhari, Y. Sharma, P. S. Archana, R. Jose, S. Ramakrishna, S. Mhaisalkar and M. Srinivasan, *J. Appl. Polym. Sci.*, 2013, **129**, 1660-1668.
- [10] K. Zhang, L. L. Zhang, X. S. Zhao and J. Wu, *Chem. Mater.*, 2010, **22**, 1392-1401.
- [11] J. Zhang and X. S. Zhao, *J. Phys. Chem. C*, 2012, **116**, 5420-5426.
- [12] M. K. Liu, Y.-E. Miao, C. Zhang, W. W. Tjiu, Z. B. Yang, H. S. Peng and T. X. Liu, *Nanoscale*, 2013, **5**, 7312-7320.
- [13] B. Ma, X. Zhou, H. Bao, X. W. Li and G. C. Wang, *J. Power Sources*, 2012, **215**, 36-42.

**Table S2** List of cyclic stability values of various graphene/PANI hybrid electrodes from recent literatures.

Binder-free	Preparation method	Capacitance retention	Measurement condition	Reference (year)
Yes(our work)	Electrophoretic deposition	84%	At 2 A g <sup>-1</sup> ; 1000 cycles	(2013)
Yes	Layer-by-layer (LbL) assembly	90.7%	At 3 A g <sup>-1</sup> ; 500 cycles	[1] (2012)
	Electropolymerization	82%	At 5 A g <sup>-1</sup> ; 1000 cycles	[2] (2013)
	Vacuum filtration	66.8%	At 0.5 A g <sup>-1</sup> ; 400 cycles	[3] (2011)
	Chemical polymerization	90%	At 5 A g <sup>-1</sup> ; 5000 cycles	[4] (2013)
	Vacuum filtration	79%	At 3 A g <sup>-1</sup> ; 800 cycles	[5] (2010)
No	Chemical polymerization	89%	At 1 A g <sup>-1</sup> ; 1000 cycles	[6] (2013)
	Chemical polymerization	87.4%	At 3 A g <sup>-1</sup> ; 10000 cycles	[7] (2013)
	Chemical polymerization	82%	At 0.3 A g <sup>-1</sup> ; 10000 cycles	[8] (2012)
	Chemical polymerization	94.3%	At 1 A g <sup>-1</sup> ; 2000 cycles	[9] (2012)
	Chemical polymerization	92%	At 1 A g <sup>-1</sup> ; 2000 cycles	[10] (2010)

**Reference:**

- [1] T. Lee, T. Yun, B. Park, B. Sharma, H.-K. Song and B.-S. Kim, *J. Mater. Chem.*, 2012, **22**, 21092-21099.
- [2] H. P. Cong, X. C. Ren, P. Wang and S. H. Yu, *Energy Environ. Sci.*, 2013, **6**, 1185–1191.

- [3] S. Liu, X. H. Liu, Z. P. Li, S. R. Yang and J. Q. Wang, *New J. Chem.*, 2011, **35**, 369-374.
- [4] Y. N. Meng, K. Wang, Y. J. Zhang and Z. X. Wei, *Adv. Mater.*, 2013, **25**, 6985-6990.
- [5] Q. Wu, Y. X. Xu, Z. Y. Yao, A. R. Liu and G. Q. Shi, *ACS Nano*, 2010, **4**, 1963-1970.
- [6] M. K. Liu, Y.-E. Miao, C. Zhang, W. W. Tjiu, Z. B. Yang, H. S. Peng and T. X. Liu, *Nanoscale*, 2013, **5**, 7312-7320.
- [7] H. L. Cao, X. F. Zhou, Y. M. Zhang, L. Chen and Z. P. Liu, *J. Power Sources*, 2013, **243**, 715-720.
- [8] J. Zhang and X. S. Zhao, *J. Phys. Chem. C*, 2012, **116**, 5420-5426.
- [9] B. Ma, X. Zhou, H. Bao, X. W. Li and G. C. Wang, *J. Power Sources*, 2012, **215**, 36-42.
- [10] J. J. Xu, K. Wang, S.-Z. Zu, B.-H. Han and Z. X. Wei, *ACS Nano*, 2010, **4**, 5019-5026.