Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A. This journal is © The Royal Society of Chemistry 2016

**Supplementary Information** 

## **Battery-Like Supercapacitors from Diamond Networks**

## and Water-Soluble Redox Electrolytes

Siyu Yu,<sup>a</sup> Nianjun Yang,<sup>\*a</sup> Hao Zhuang,<sup>a</sup> Soumen Mandal,<sup>b</sup> Oliver A. Williams,<sup>b</sup> Bing Yang,<sup>c</sup>

Nan Huang,<sup>c</sup> Xin Jiang\*a

<sup>a</sup> Institute of Materials Engineering, University of Siegen, 57076 Siegen, Germany

<sup>b</sup> School of Physics and Astronomy, Cardiff University, Cardiff CF24 3AA, UK

<sup>c</sup> Shenyang National Laboratory for Materials Science, Institute of Metal Research (IMR),

Chinese Academy of Sciences (CAS), No.72 Wenhua Road, Shenyang 110016 China

E-mail: nianjun.yang@uni-siegen.de; xin.jiang@uni-siegen.de

## **Table of Content**

Table S1. Capacitance comparison of diamond based electrochemical capacitors.

Figure S1. Performance of a diamond PC using Fe(CN)<sub>6</sub><sup>3-/4-</sup> redox electrolytes.

Figure S2. Performance of a diamond PC using other water-soluble redox electrolytes.

Figure S3. Performance of a diamond network PC using water-soluble redox electrolytes.

Figure S4. Performance of diamond symmetric EDLC and PC devices.

References

Туре	Capacitor Electrode	Capacitance	Electrolyte	Ref.
		[mF cm <sup>-2</sup> ]		
EDLC	Diamond	$(4-7) \times 10^{-3}$	Na <sub>2</sub> SO <sub>4</sub>	1
	Diamond network	3.53	$Na_2SO_4$	This work
	Diamond/Silicon nanowires Diamond foam	0.1	PMPyrrTFSI + propylene	2
			carbonate	
		1.5	Et <sub>3</sub> NH TFSI	3
		0.60	NaClO <sub>4</sub>	4
		0.44	PMPyrrTFSI + propylene	4
			carbonate	
	Honeycomb diamond	1.97	$H_2SO_4$	5
		3.91	$H_2SO_4$	6
		0.67	TEABF <sub>4</sub> + propylene	7
			carbonate	
	Porous diamond	3	LiClO <sub>4</sub>	8
	BDD/TiO <sub>2</sub>	7.46	NaNO <sub>3</sub>	9-11
	TiO <sub>2</sub> /BDD/Ta	5.23	Na <sub>2</sub> SO <sub>4</sub>	12
	BDD/Nanotube	0.58	PBS	13
РС	Diamond	41.51	$Na_2SO_4 + Fe(CN)_6^{3-/4-}$	This work
	Diamond network	73.42	$Na_2SO_4 + Fe(CN)_6^{3-/4-}$	This work
	MnO <sub>2</sub> /Diamond	7.82	Na <sub>2</sub> SO <sub>4</sub>	1
	Ni(OH) <sub>2</sub> /Diamond	91	NaOH	14
	Nanowire			

Table S1. Capacitance comparison of diamond nanostructures based electric double layer capacitors (EDLCs) and pseudocapacitors (PCs).



Figure S1. Performance of a diamond PC using  $Fe(CN)_6^{3-/4-}$  redox electrolytes. (a) Cyclic voltammograms of 0.05 M  $Fe(CN)_6^{3-/4-}$  in 1.0 M Na<sub>2</sub>SO<sub>4</sub> at the scan rates of 10, 20, 50, and 100 mV s<sup>-1</sup>. (b) Charge/discharge curves of 0.05 M  $Fe(CN)_6^{3-/4-}$  in 1.0 M Na<sub>2</sub>SO<sub>4</sub> at the current densities of 1.0, 2.0, and 5.0 mA cm<sup>-2</sup>. (c) Cyclic voltammograms of  $Fe(CN)_6^{3-/4-}$  with the concentrations of 0.01, 0.05, 0.1, and 0.2 M in 1.0 M Na<sub>2</sub>SO<sub>4</sub> at the scan rate of 10 mV s<sup>-1</sup>. (d) Capacitance comparison calculated from cyclic voltammogramms at different scan rates and different sconcentrations of  $Fe(CN)_6^{3-/4-}$ . (e) Nyquist plots in the frequency range of 10<sup>6</sup> - 0.01 Hz with and without 0.05 M  $Fe(CN)_6^{3-/4-}$  in 1.0 M Na<sub>2</sub>SO<sub>4</sub> as the electrolyte. (f) Capacitance

retention at a charge/discharge current density of 5 mA cm^2 in 0.05 M  $Fe(CN)_6{}^{3\text{-/-}}$  + 1.0 M  $Na_2SO_4$  .



Figure S2. Performance of a diamond PC using other water-soluble redox electrolytes. (a) Cyclic voltammograms of 0.1 M hydroquinone in 1.0 M  $H_2SO_4$ . (b) Cyclic voltammograms of 2 mM FcTMAPF<sub>6</sub> in 0.1 KCl. The scan rates were 10, 20, 50, and 100 mV s<sup>-1</sup>.



Figure S3. Performance of a diamond network PC using water-soluble redox electrolytes. (a) Cyclic voltammograms of 0.05 M Fe(CN)<sub>6</sub><sup>3-/4-</sup> in 1.0 M Na<sub>2</sub>SO<sub>4</sub> at the scan rates of 10, 20, 50, and 100 mV s<sup>-1</sup>. (b) Charge/discharge curves of 0.05 M Fe(CN)<sub>6</sub><sup>3-/4-</sup> in 1.0 M Na<sub>2</sub>SO<sub>4</sub> at current densities of 1.0, 2.0, and 5.0 mA cm<sup>-2</sup>. (c) Nyquist plots in the frequency range of 10<sup>6</sup> - 0.01 Hz with and without 0.05 M Fe(CN)<sub>6</sub><sup>3-/4-</sup> in 1.0 M Na<sub>2</sub>SO<sub>4</sub> as the electrolyte.



Figure S4. Performance of diamond symmetric EDLC and PC devices. (a) Charge/discharge curves of a diamond EDLC device in 1.0 M Na<sub>2</sub>SO<sub>4</sub> at the current density of 1.0, 2.0, 5.0 and 10.0  $\mu$ A cm<sup>-2</sup>. (b) Ragone plots of a diamond EDLC device in 1.0 M Na<sub>2</sub>SO<sub>4</sub> with a cell voltage of 1.0, 2.0 and 2.4 V. (c) Charge/discharge curves of a diamond PC device in 0.05 M Fe(CN)<sub>6</sub><sup>3-/4-</sup> + 1.0 M Na<sub>2</sub>SO<sub>4</sub> at the current density of 1.0, 2.0, 5.0 and 10.0 mA cm<sup>-2</sup>. (d) Ragone plots of a diamond PC device in 0.05 M Fe(CN)<sub>6</sub><sup>3-/4-</sup> + 1.0 M Na<sub>2</sub>SO<sub>4</sub> with a cell voltage of 1.0, 2.0 and 2.4 V.

- 1. S. Yu, N. Yang, H. Zhuang, J. Meyer, S. Mandal, O. A. Williams, I. Lilge, H. Schönherr and X. Jiang, *The Journal of Physical Chemistry C*, 2015, **119**, 18918-18926.
- F. Gao, G. Lewes-Malandrakis, M. T. Wolfer, W. Müller-Sebert, P. Gentile, D. Aradilla, T. Schubert and C. E. Nebel, *Diamond Relat. Mater.*, 2015, 51, 1-6.
- D. Aradilla, F. Gao, G. Lewes-Malandrakis, W. Müller-Sebert, D. Gaboriau, P. Gentile, B. Iliev, T. Schubert, S. Sadki, G. Bidan and C. E. Nebel, *Electrochemistry Communications*, 2016, 63, 34-38.
- 4. F. Gao, M. T. Wolfer and C. E. Nebel, *Carbon*, 2014, **80**, 833-840.
- 5. K. Honda, T. N. Rao, D. A. Tryk, A. Fujishima, M. Watanabe, K. Yasui and H. Masuda, J. *Electrochem. Soc.*, 2000, **147**, 659-664.
- K. Honda, T. N. Rao, D. A. Tryk, A. Fujishima, M. Watanabe, K. Yasui and H. Masuda, J. Electrochem. Soc., 2001, 148, A668-A679.
- M. Yoshimura, K. Honda, R. Uchikado, T. Kondo, T. N. Rao, D. A. Tryk, A. Fujishima,
  Y. Sakamoto, K. Yasui and H. Masuda, *Diamond Relat. Mater.*, 2001, 10, 620-626.
- 8. C. Hébert, E. Scorsone, M. Mermoux and P. Bergonzo, *Carbon*, 2015, **90**, 102-109.
- 9. K. Siuzdak, R. Bogdanowicz, M. Sawczak and M. Sobaszek, *Nanoscale*, 2015, 7, 551-558.
- 10. M. Sobaszek, K. Siuzdak, M. Sawczak, J. Ryl and R. Bogdanowicz, *Thin Solid Films*, 2016, **601**, 35-40.
- 11. M. Sawczak, M. Sobaszek, K. Siuzdak, J. Ryl, R. Bogdanowicz, K. Darowicki, M. Gazda and A. Cenian, *Journal of The Electrochemical Society*, 2015, **162**, A2085-A2092.
- C. Shi, H. Li, C. Li, M. Li, C. Qu and B. Yang, *Applied Surface Science*, 2015, 357, Part B, 1380-1387.
- C. Hébert, J. P. Mazellier, E. Scorsone, M. Mermoux and P. Bergonzo, *Carbon*, 2014, 71, 27-33.
- 14. F. Gao and C. E. Nebel, *physica status solidi* (a), 2015, **212**, 2533-2538.