

## **Graphene structures prepared *via* pulse alternating current technique**

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### **Synthesis and characterization of graphene nanostructures**

For the preparation of graphene nanostructures two thermally expanded graphite (TEG) GF-100 electrodes with the same area were immersed in the water-based electrolytes (2M LiOH, 2M NaOH or 2M KOH), and the electrodes were connected to an AC source operating at 50 Hz. Current density was  $1 \text{ A cm}^{-2}$ . In turn, the TEG electrodes were exfoliated by the applied alternating current with the symmetrical pulses. The suspension of graphene was then filtrated and the prepared material was rinsed with H<sub>2</sub>O to achieve neutral pH and was dried at 80°C to until their constant weight was attained. The Raman spectra of samples were excited by an argon laser ( $\lambda = 514.5 \text{ nm}$ ) and recorded in the backscattering geometry over a range of 50-3200  $\text{cm}^{-1}$  on a Renishaw spectrometer equipped with a CCD detector. Transmission electron microscopy (TEM) studies were performed on a Hitachi HT-7700 TEM (High-Technologies Europe GmbH) system at the field emission of 100 kV. Scanning electron microscopy (SEM) studies were performed on Hitachi S-4800 II at the field emission of 20 kV.

### **Supercapacitors measurements**

15 mg of a synthesized graphene structures was suspended in a mixture of 2-propanol (0.5 mL) and 10 wt% aqueous Nafion solution (0.02 mL). The mixture was stirred with a magnetic stirrer for 30 min and the prepared mixture was then dropped onto a glassy carbon foil electrode and was air-dried. The electrochemical measurements were carried out using standard three-electrode cell with Pt wire as a counter electrode and Ag/AgCl reference electrode.

### **Li-ion batteries measurements**

The graphene structures were incorporated in a slurry containing 95 wt% active material and 5 wt% binder (carboxymethyl cellulose, CMC). After mixing, the slurry was coated on a copper current collector using a coating machine from Zehntner Testing Instruments. The electrodes were dried for 12 h at 120 °C under vacuum. The mass of the active material in the coated electrodes was around  $2.5 \text{ mg cm}^{-2}$ . For testing the anode material, a standard electrolyte solution 1 M LiPF<sub>6</sub> in EC:DMC (1:1) was used. The electrochemical measurements were carried out using CR2016 coin cells, closed

by a hydraulic crimping machine (MSK 110 from MTI Corporation). The cell assembly, coin cells crimping and the electrochemical measurements were performed at room temperature in a glove-box.

**Table S1** Comparison between characteristics of graphene and Pt/graphene materials towards the SCs, LIBs and EOR applications.

Sample	Preparation	Morphology	Electrochemical measurements	Specific capacity, F g <sup>-1</sup>	Capacity, mAh g <sup>-1</sup>	E <sub>onset</sub> , V (EOR)	Ref.
Supercapacitor application							
Graphene	Vacuum filtration deposition	graphene platelets with the average diameters of approximately 5–10 μm and average thickness of 6–8 nm	CV, 6 M KOH, 10 mV s <sup>-1</sup>	152			S1
Graphene oxide	Standard Hummers method	1 layer, 1.2 μm thick	CV, 100 mV s <sup>-1</sup> , 30 wt % KOH	205	-		S2
Graphene	Electrochemical exfoliation in sodium dodecylbenzene sulfonate	2 layers	2.0 M KOH Galvanostatic charge-discharge at 0.5 A g <sup>-1</sup>	26.10	-		S3
Graphene structures	Electrochemical exfoliation of thermal expansion graphite under PAC	multi-layered graphene flakes with lateral size of 1–3 μm	CV, 2-200 mV s <sup>-1</sup> , 2M NaOH	44-53			This work
Lithium-ion batteries application							
Few-layered graphene	microwave-assisted exfoliation	Graphene sheets with lateral dimensions of at least 1 μm <sup>2</sup>	0.1C, voltage range of 0.005–3.0 V		~400		S4
Porous graphene framework	Modified Hummers method	3D interconnected graphene electrode with large-scale continuity	0.1C, voltage range of 0.01–3.0 V		341		S5
Graphene structures	Electrochemical exfoliation of thermal expansion graphite under PAC	multi-layered graphene flakes with lateral size of 1–3 μm	0.1C, voltage range of 0.0–1.0 V		457		This work
Fuel cell application							
Pt/FLG	The FLG support was prepared by CVD; The catalyst was synthesized using the polyol-assisted reduction method	1.8 ± 0.6 nm Pt particles supported onto few-layered graphene	1M H <sub>2</sub> SO <sub>4</sub> +1M C <sub>2</sub> H <sub>5</sub> OH with a scan rate of 50 mV s <sup>-1</sup>			0.62	S6
Pt/GNS	Graphene nanosheets was prepared through chemical vapor deposition; the catalysts were synthesized by using the one-pot polyol-assisted reduction method	1.8 nm Pt particles supported onto GNS	1M H <sub>2</sub> SO <sub>4</sub> +1M C <sub>2</sub> H <sub>5</sub> OH with a scan rate of 50 mV s <sup>-1</sup>			0.62	S7
Pt/Graphene	Both graphene structures and Pt/Graphene catalyst were prepared through PAC technique	Pt nanoparticles supported onto multi-layered graphene flakes	0.5M H <sub>2</sub> SO <sub>4</sub> +0.5M C <sub>2</sub> H <sub>5</sub> OH with a scan rate of 20 mV s <sup>-1</sup>			0.53	This work

## References

- S1 S. Zhang, Y. Li and N. Pan, *J. Power Sources*, 2012, **206**, 476.
- S2 Y. Wang, Z. Shi, Y. Huang, Y. Ma, C. Wang, M. Chen and Y. Chen, *J. Phys. Chem. C*, 2009, **113**, 13103.
- S3 A. Hamra, H. Lim, W. Chee and N.J.A.S.S. Huang, *Appl. Surf. Sci.*, 2016, **360**, 213.
- S4 S. Petnikota, N.K. Rotte, V.V. Srikanth, B.S. Kota, M. Reddy, K.P. Loh and B. Chowdari, *J. Solid State Electrochem.*, 2014, **18**, 941.
- S5 Z. Zuo, T.Y. Kim, I. Kholmanov, H. Li, H. Chou and Y. Li, *Small*, 2015, **11**, 4922.
- S6 Y. Shen, K. Xiao, J. Xi and X. Qiu, *J. Power Sources*, 2015, **278**, 235.
- S7 Y. Shen, Z. Zhang, K. Xiao and J. Xi, *ChemCatChem*, 2014, **6**, 3254.