Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A. This journal is © The Royal Society of Chemistry 2019

## Supporting Information

## Dense Organic Molecules/graphene Network Anodes with Superior Volumetric and Areal Performance for Asymmetric Supercapacitors

Lina Zhang,<sup>‡ab</sup> Daliang Han,<sup>‡ab</sup> Ying Tao,<sup>ab</sup> Changjun Cui,<sup>ab</sup> Yaqian Deng,<sup>c</sup> Ximan Dong,<sup>ab</sup> Wei Lv,<sup>c</sup> Zifeng Lin,<sup>d</sup> Shichao Wu,<sup>ab</sup> Zhe Weng,<sup>\*ab</sup> and Quan-Hong Yang<sup>\*ab</sup>

<sup>a</sup> Nanoyang Group, State Key Laboratory of Chemical Engineering School of Chemical Engineering and Technology, Tianjin University, Tianjin 300350, China, E-mail: zweng@tju.edu.cn; qhyangcn@tju.edu.cn

<sup>b</sup> Joint School of National Singapore and Tianjin University, International Campus of Tianjin

University, Binhai New City, Fuzhaou 350207, China.

<sup>c</sup> Shenzhen Key Laboratory for Graphene-based Materials, Graduate School at Shenzhen,

Tsinghua University, Shenzhen 518055, China.

<sup>d</sup> College of Materials Science and Engineering, Sichuan University, Chengdu 610065, China.

‡ These two authors are equal major contributors to this work.



Fig. S1 (a) Solubility of anthraquinone (AQ) and (b) anthraquinone-2-sulfonate (AQS) with different concentration.



Fig. S2 Cyclic voltammetry profiles of pure AQS.



Fig. S3 Electrical conductivity of pure AQS, HPGM and AQS/Gs with various AQS loading.



**Fig. S4** (a) Gravimetric specific capacitances of HPGM, AQS and AQS/Gs with different AQS mass loading. (b) Galvanostatic charge-discharge curves of AQS/G-1 at high current densities. (c) Cycling performance of AQS/G-1.



Fig. S5 UV-vis spectra of pure AQS aqueous solution, and the electrolyte before and after cycling.



Fig. S6 SEM image of (a) AQS/G-1 xerogel and (b) compact AQS/G-1.



Fig. S7 (a) CV curves, (b) gravimetric and (c) volumetric specific capacitances of AQS/G-1 xerogel after freezing drying.

![](_page_3_Figure_2.jpeg)

Fig. S8 SEM images of (a) AQS, (b) AQS/G-0.5, (c) AQS/G-1 and (d) AQS/G-2.

![](_page_3_Figure_4.jpeg)

Fig. S9 High magnification SEM images of (a) AQS/G-1 xerogel and (b) compact AQS/G-1.

![](_page_4_Figure_0.jpeg)

**Fig. S10** (a) TEM image and (b) HRTEM image of AQS/G-1. (c) Corresponding selected area electron diffraction (SEAD) image of AQS/G-1. (d) TEM image and (e) HRTEM image of AQS. (f) Corresponding SEAD image of AQS.

![](_page_4_Figure_2.jpeg)

Fig. S11 XRD patterns of pristine AQS, AQS/G-1 and pure AQS after hydrothermal treatment.

![](_page_5_Figure_0.jpeg)

Fig. S12 Raman spectra of HPGM and AQS/G-1.

![](_page_5_Figure_2.jpeg)

Fig. S13 (a) Survey XPS spectrum of AQS/G-1. (b) XPS high-resolution spectra of C1s and (c) O1s.

![](_page_5_Figure_4.jpeg)

Fig. S14 Capacitive contribution to the total charge at a scan rate of 50 mV s<sup>-1</sup>.

![](_page_6_Figure_0.jpeg)

Fig. S15 Schematical illustration of the asymmetric supercapacitor fabrication process with AQS/G-1 as the anode and  $RuO_2/G$  as the cathode.

![](_page_6_Figure_2.jpeg)

Fig. S16 (a) Cyclic voltammetry profiles and (b) galvanostatic charge-discharge curves of RuO<sub>2</sub>/G.

![](_page_7_Figure_0.jpeg)

**Fig. S17** (a) Cyclic voltammetry profiles of the AQS/G-1//RuO<sub>2</sub>/G ASC at different operating voltages at a constant scan rate of 5 mV s<sup>-1</sup>. (b) Galvanostatic charge-discharge curves of AQS/G-1 and RuO<sub>2</sub>/G in different potential windows at a current density of 0.5 A g<sup>-1</sup>.

![](_page_7_Figure_2.jpeg)

Fig. S18 (a) CV curves and (b) volumetric sepcific capacitance of the ASCs with various mass ratios (AQS/G-1 to  $RuO_2/G$ ).

![](_page_7_Figure_4.jpeg)

Fig. S19 Cycling performance of the AQS/G-1//RuO<sub>2</sub>/G asymmetric supercapacitor.