Supplementary Information

Impact of Graphene Quantum Dot Edge Shapes on High-Performance Energy Storage Devices

Grainne Gilleece*[a], Natasha Shirshova*[a], Ensieh Hosseini[a], Karl Coleman[b], Marcos Perez-Pucheta[b], and Dagou Zeze[a]
[a] Department of Engineering, University of Durham, United Kingdom, [b] Department of Chemistry, University of Liverpool, United Kingdom

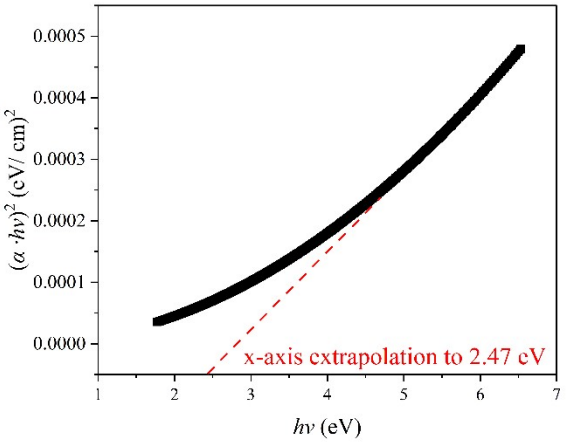

***Figure S1**: Tauc plot of HT1. The linear part of the plot is extrapolated to 2.48 eV, which is the bandgap.*

Optical bandgap calculations were conducted by graphical Tauc plots based on measured UV-Vis data. The Tauc method is based on equation (S1):

$$(\alpha \cdot hv)^{\frac{1}{\gamma}} = B(hv - E_g) \quad \text{(S1)}$$

where α is the absorption coefficient for the sample, h is Planck's constant, v is the photon frequency, B is a constant of proportionality, E_g is the bandgap energy, and γ is equal to ½ for a direct bandgap transition, or 2 for an indirect bandgap transition.[1]

From these plots, the bandgap for all GQDs was determined to be direct (as plots for indirect bandgap transitions resulted in negative bandgap values). This agrees with previously simulated data demonstrated through density functional theory (DFT) calculations that all GQDs displayed a direct bandgap transition.[2]

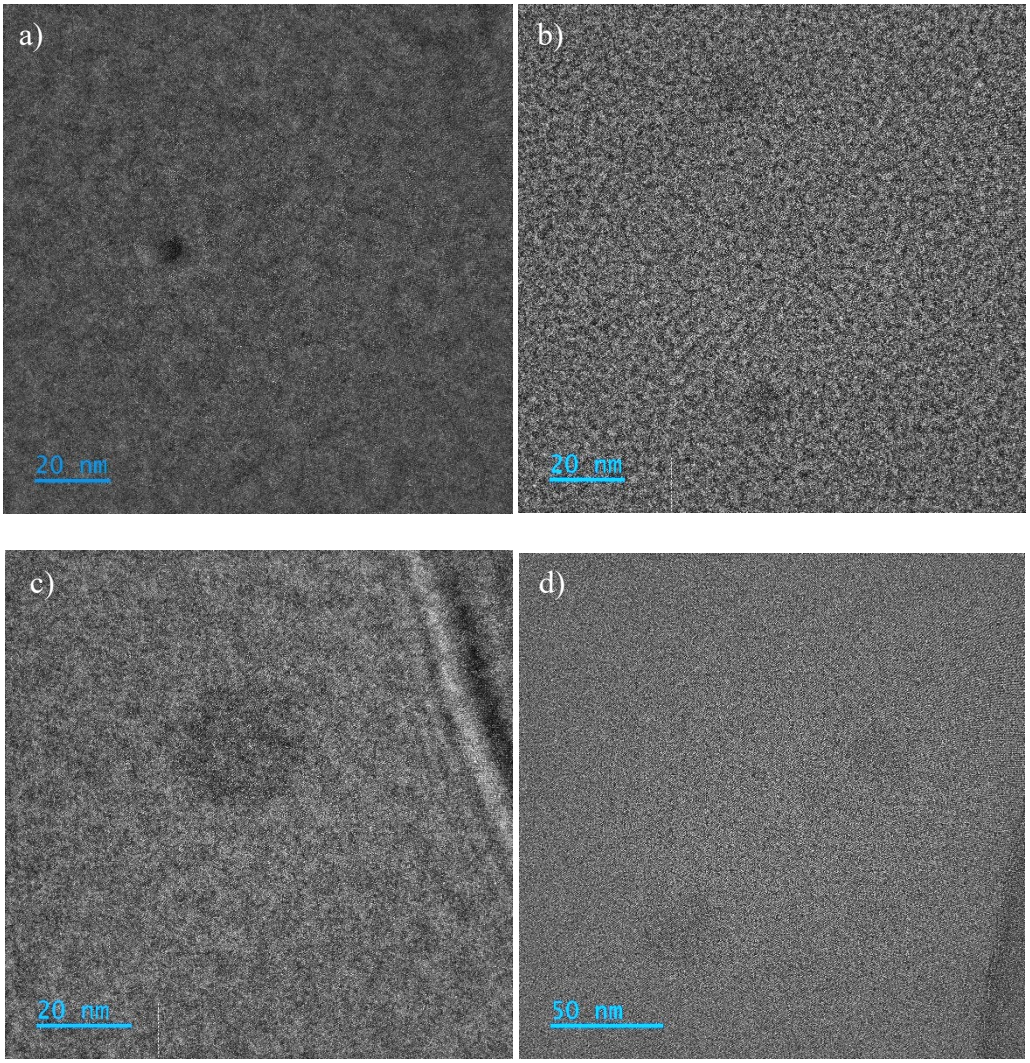

Figure S2: *TEM images of a) HT1, b) HT2, c) HT3, d) HT4., corresponding to 150, 165, 180 and 195 s heating times, respectively. The TEM images display the increase in the lateral dimension of the N-GQD size upon increased microwave reaction time (~ 5 nm for HT1, ~ 18 nm for HT2, ~27 nm for HT3, ~40 nm for HT4.*

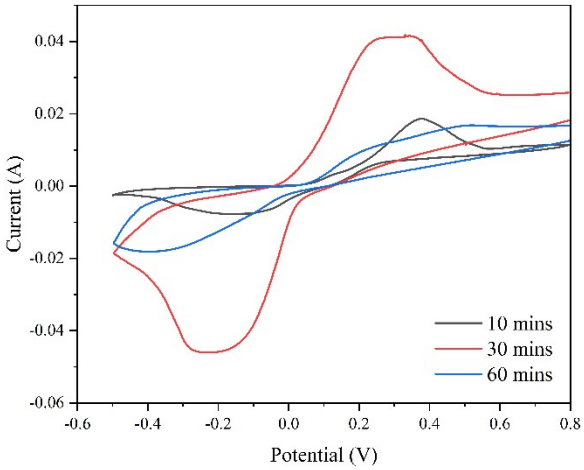

Figure S3*: Cyclic voltammograms at 5 mV/s of carbon fibres with electrodeposited HT1 in 1M H_2SO_4 electrolyte for various electrodeposition times.*

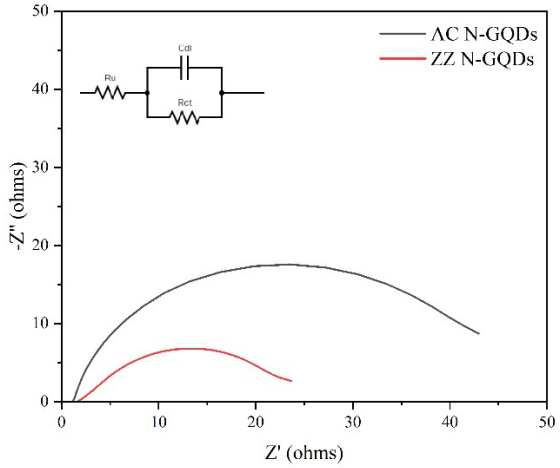

Figure S4: Nyquist plots of carbon fibers with deposited AC and ZZ N-GQDs between 50 mHz and 10 kHz. ZZ N-GQDs display a lower charge transfer resistance. The equivalent circuit is displayed.

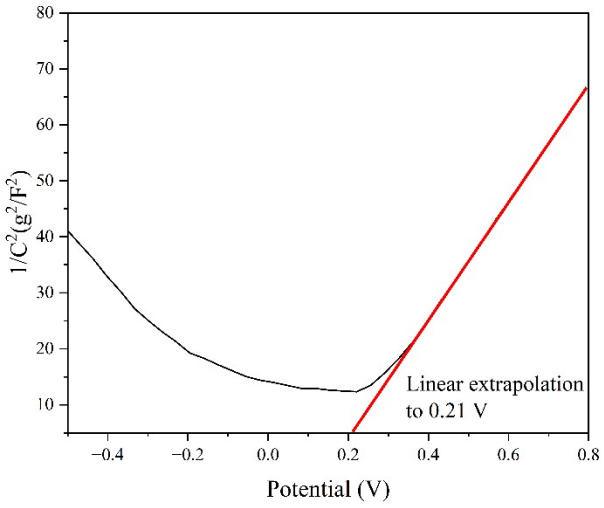

***Figure S5**: Mott Schottky plot for AC GQDs (EC1) showing n-type conductivity with a flatband potential of 0.21V.*

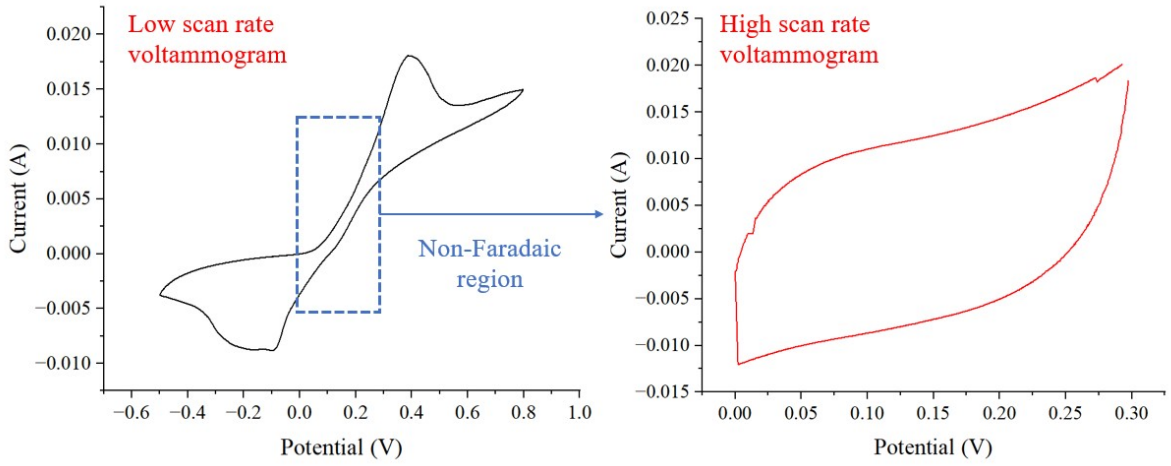

***Figure S6**: Cyclic voltammograms of electrochemically synthesised N-GQDs: a) between -0.5 V and 1.2 V at a scan rate of 0.005 V/s. The non-Faradaic region is highlighted, and b) between 0 V and 0.3 V at a scan speed of 1 V/s. The rectangular shape of the voltammogram is indicative of purely double layer capacitance.*

References

1 P. Makuła, M. Pacia and W. Macyk, *Journal of Physical Chemistry Letters*, 2018, **9**, 6814–6817.

2 M. Ghandchi, Gh. Darvish, M.K. Moravvej-Farshi, *TJEE*, 2021, **2**, 213-220