Communication

Nitrogen-Doped Graphene Quantum Dots with Oxygen-rich Functional Groups

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The electrochemical process for the formation of N GQDs is shown in Figures S1 and S ι Just like the oxidation of other car bon materials by an oxidant

ods. Comparing with GQDs however N GQDs exhibit a broader D band suggesting that the intercalation of N atoms into the con jugated carbon backbone has led to somewhat disordered struc tures.

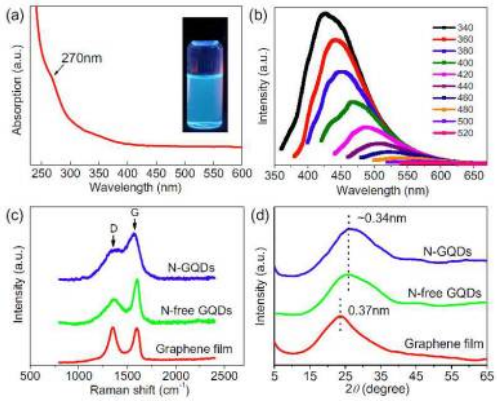

Figure 3. a and b UV vis absorption and photoluminescence PL spectra of N GQDs in water respectively. c Raman spectra and d XRD patterns of the original graphene film the N free and N GQDs. Inset in a is a photo of the N GQD solution in water under nm UV irradiation.

Figure d shows typical XRD profiles for the original graphene film and the as prepared N free and N GQDs. Just like their N free counterparts the N GQDs show a broader diffraction peak at around 2^{o} which is substantially higher than that of the gra phene film *ca.* 2 °. The more compact interlayer spacing *ca.* 0 nm probed by XRD for N GQDs than the original graphene film *ca.* 0 nm is consistent with the TEM observation Figure S . The reduced interlayer spacing in N GQDs could be attrib uted to the effective π π stacking of tiny graphenes with few struc ture defects Figure a . On the other hand the possible formation of hydrogen bonding between the O containing functional groups surrounding the edges of the graphene layers in N GQDs Figures a S and S may further facilitate the compact stacking of gra phene layers Figure S . It is also worth to note that N GQDs thus prepared do not show any diffractions in the region of *ca.* 10^{o} characteristic of graphene oxides evidently indicating that the N GQDs are different from graphene oxide though both contain oxygen enriched functional groups Figure .

N doped carbon nanomaterials such as N CNTs and N graphene have been demonstrated to hold promise as metal free electrocatalysts in replacing the commercially available Pt based catalyst for ORR. Apart from their unique luminescent properties N GQDs are also expected to possess the electrocatalytic activi ties for ORR. To avoid any possible effect of the glassy carbon GC base electrode Figure S we used a large area and electri cally conductive graphene assembly to support the N GQD as ORR catalysts. The graphene supported N GQDs N GQDs graphene were prepared by hydrothermal treatment of the suspension of well dispersed graphene oxides with N GQDs Figure S10 . This mild process ensured the formation of N GQD graphene assemblies without acutely changing the intrinsi cally chemical nature of N GQDs Figures S11 S1 . The N GQD graphene film Figure S1 thus formed was demonstrated to exhibit a good conductivity of *ca.* 0 S cm and superior elec trocatalytic ability for ORR see below .

Figures a and b depict CVs for O reduction on the N GQD graphene in comparison with a commercial Pt C catalyst 0wt% platinum on carbon black . Sim f l e t 2 1 o aandoporeo c T 11 Tf 10 s 2 fc
el h s 2 h n 0 1 G 1 Q Tf 1 0 Td 10 2 Td G 1 Q 2Td o 10 2Td w 1 TJ

In summary we have developed a simple yet effective electrochemical strategy to generate N doped GQDs with O rich functional groups which show unique optoelectronic features distinctive from their N free counterparts Supported by graphene sheets N GQDs were demonstrated to possess superior electrocatalytic ability Apart from the use of N GQDs as metal free catalyst for ORR their unique luminescent properties indicate potentials for bioimaging and light emitting diodes among many other potential applications

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Supporting Information The experimental details for preparation of N GQDs N GQD graphenes electrode fabrication related characterization and supplementary results and discussion This material is available free of charge via the Internet at http pubs acs org

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