Supporting Information

Experimental Methods

Materials

Carbon nano-onions were prepared by the thermal annealing of commercially available nanodiamond powders (Nanostructured and Amorphous Materials, Inc., Houston, TX) at 1650 °C for 1h under the flow of helium in a graphitization furnace. H_2SO_4, HNO_3, K_2CO_3 and NH_4OH were purchased from Sigma-Aldrich. The 200 mL stainless steel hydrothermal reactor houses a PTFE sample chamber in which the ox-GQDs were reacted with aqueous ammonia. All samples were dialyzed against deionized water in 1 kDa MWCO dialysis bags purchased from Spectrum Labs.

Instrumentation and Sample Preparation

The size of GQDs and N-GQDs were characterized by a JEOL JEM-2200FS, 200 kV electron acceleration voltage TEM and the thickness was measured with a Park Systems XE-70 AFM operating in non-contact mode. TEM samples were prepared by dipping a TEM copper grid (lacey carbon support film) into a dilute solution of GQD sample. AFM samples were prepared by dropping 5 µL GQD sample onto a mica substrate disc and spin casting to dry. FTIR spectra were taken with a Thermo Scientific Model Nicolet 6700 FTIR Spectrometer operating with a diamond ATR crystal. FTIR samples were prepared by dropping 5 µL GQD samples onto the ATR crystal and drying in an oven until a thick film was observed. The XPS characterization was conducted with a Thermo Scientific Model K-Alpha XPS instrument. The XPS samples were prepared by dropping 5 µL of GQD sample onto a silicon wafer and drying between depositions until a thick film could be observed. Absorption spectra were collected with a Thermo Scientific Evolution 201 Spectrophotometer and emission spectra were recorded with a Jobin-Yvon

Spectromax 4 Spectrofluorometer. Optical properties were measured using a quartz cuvette with 10 mm

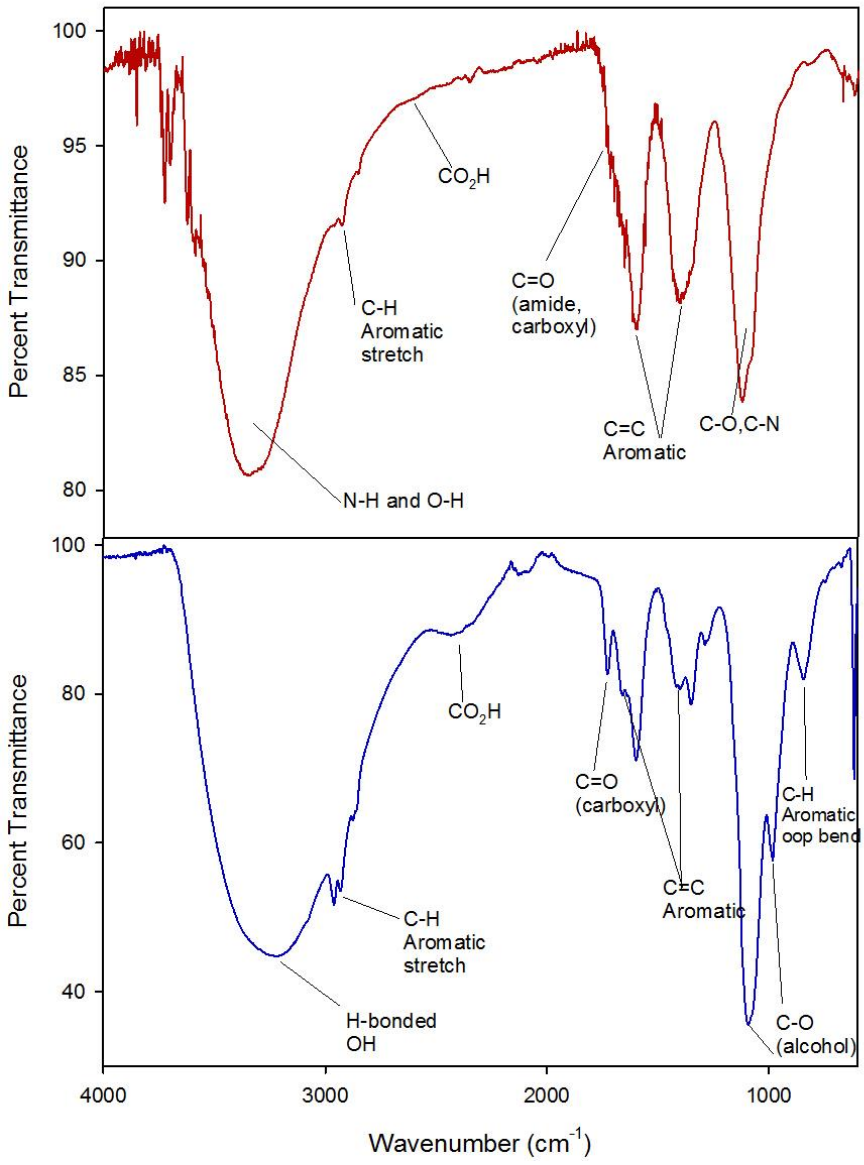

Figure S1: FTIR Spectra of N-GQD-150 (top) and ox-GQD (bottom).

path length.

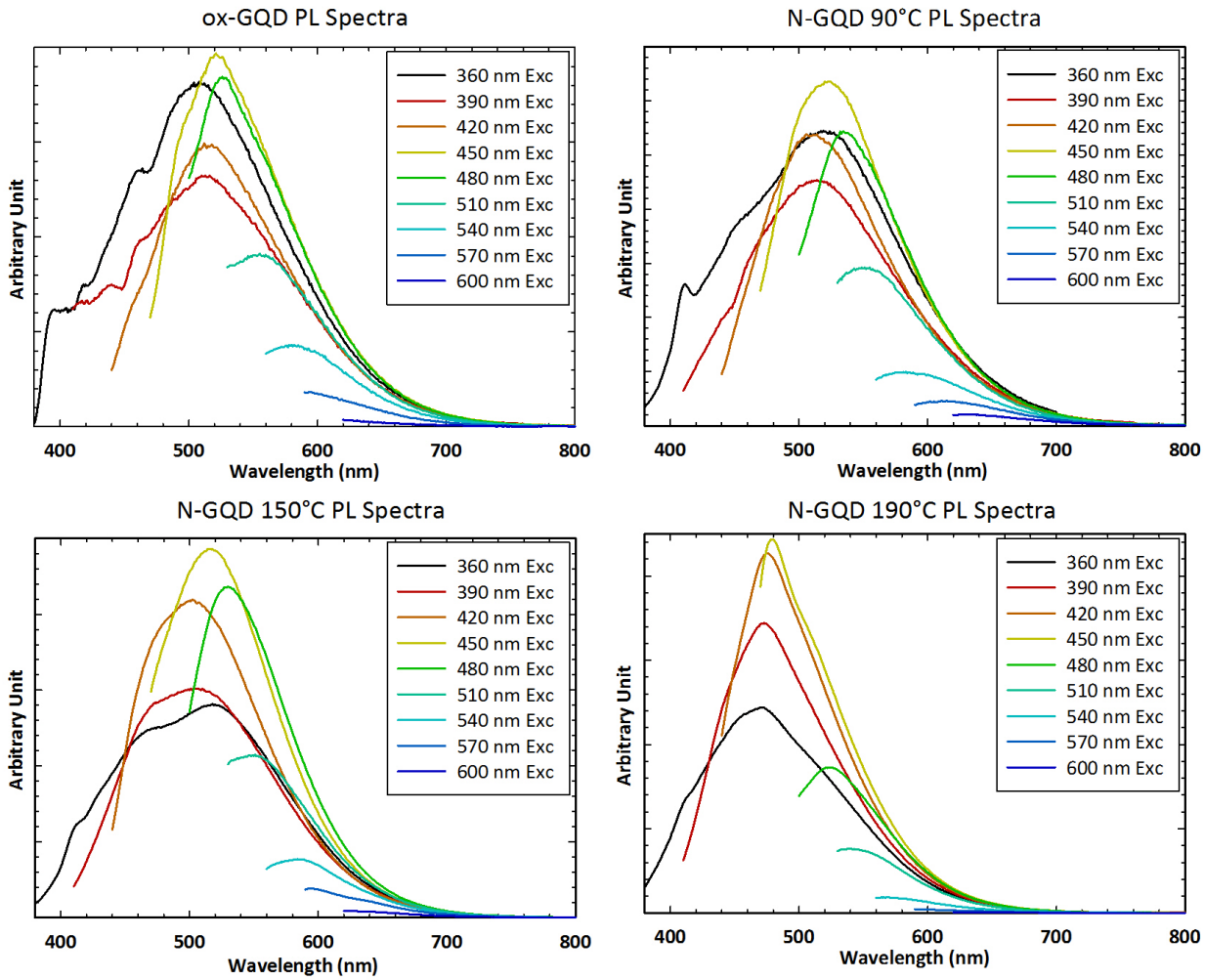

Figure S2: Emission spectra of ox-GQD (top left), N-GQD-90 (top right), N-GQD-150 (bottom left), and N-GQD-190 (bottom right).

Experimental Work	Year Published	~398.5 eV	~399.5 eV	~400 eV	>401 eV
Tetsuka et al.[7]	2012	---	Amine N (399.7 eV)	---	---
Dai et al.[9]	2014	---	Pyrrolic N (399.7 eV)	---	Graphitic N (401.6 eV)
Li et al.[10]	2012	Pyridinic N (398.5 eV)	---	---	Pyrrolic N (401 eV)
NIST Database[25]	2012	Pyridinic N (398.6 eV)	Amine N (399.4 eV)	Pyrrolic N (399.9 eV)	Graphitic N (401.3 eV)
This work	2015	Pyridinic N (398.5 eV)	Amine N (399.3 eV)	Pyrrolic N (400.2 eV)	Graphitic N (401.3 eV)

Table S1: XPS N1s peak assignments in recent literature and this work.