

Supporting Information

Experimental section

Materials

Carbon nano-onions (CNOs) were prepared by 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. HNO₃ and KOH were purchased from Sigma-Aldrich. All chemicals were used as received without further purification. Dialysis bags were purchased from Spectrum Labs. Deionized water was used throughout the experiment.

Synthesis

GQDs were prepared by chemically oxidizing 100 mg of carbon nano-onions (CNOs) in 50 mL of HNO₃ (5 M) at 95 °C, After refluxing for 4h, the solution was cooled down to room temperature and a brownish supernatant was neutralized with KOH. After removing an excessive amount of KNO₃ precipitation formed during the neutralization, the solution was dialyzed to remove ions (dialysis with MWCO=500-1000) for 3 days with constant change of dialysis buffer water. Afterwards, the resultant solution was dialyzed again in another dialysis bag (MWCO=3500) for 1 hour. We found that this process efficiently separate graphene quantum dots (GQDs) into two parts. The solution outside of the dialysis bag (MWCO=3500) exclusively exhibited UV emission and was named GQD-UV. On the other hand, what's left inside the dialysis bag showed spectrally pure blue emission and was named GQD-B.

TEM, AFM, XPS, and optical characterizations:

TEM measurements were conducted on a JEM-2200FS analytical TEM with 200 kV electron acceleration voltage. TEM samples were prepared by placing several drops of GQD solution on a TEM substrate and drying it. Substrates used for the TEM characterization were copper grids coated with lacey carbon. AFM measurements were conducted on Park XE-70 atomic force microscopy with tapping mode. AFM samples were prepared by spin-casting GQDs onto mica substrate. X-ray photoelectron spectroscopic characterizations were conducted by K-Alpha XPS instrument (Thermo Scientific) with 72W Al K α radiation. XPS samples were prepared by drop casting GQD water solutions onto silicon substrate. UV-Vis spectra were obtained using Thermo Scientific Evolution 201 UV-Vis spectrophotometer. Photoluminescence (PL) and photoluminescence excitation (PLE) spectra were obtained using Horiba Scientific FluoroMax-4 spectrofluorometer. All optical measurements were obtained with GQDs dispersed in water which were contained in quartz cuvette with 1 cm path length.

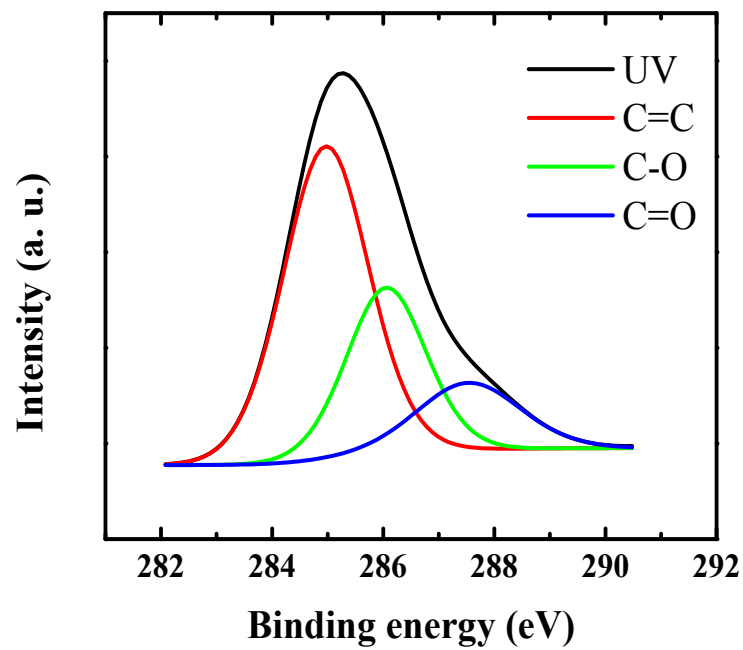
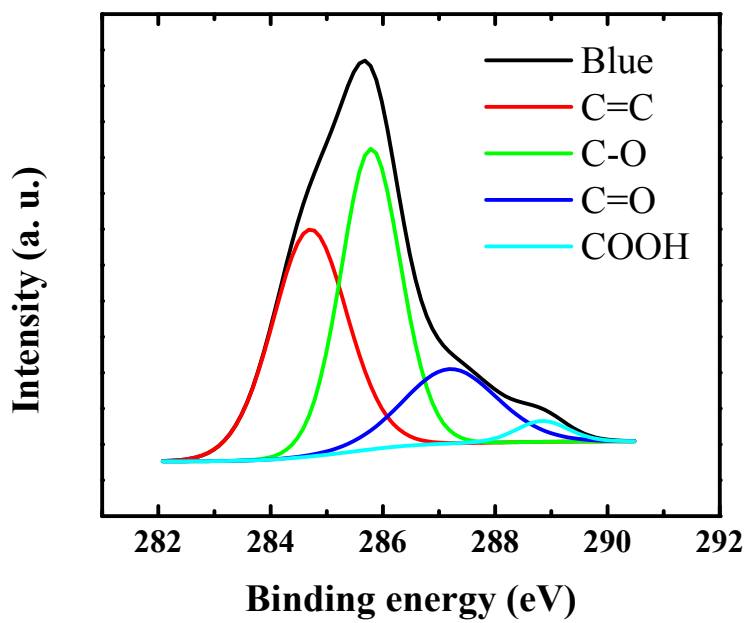


Fig. 1 XPS high resolution C1s spectra of (left) GQD-B and (right) GQD-UV.

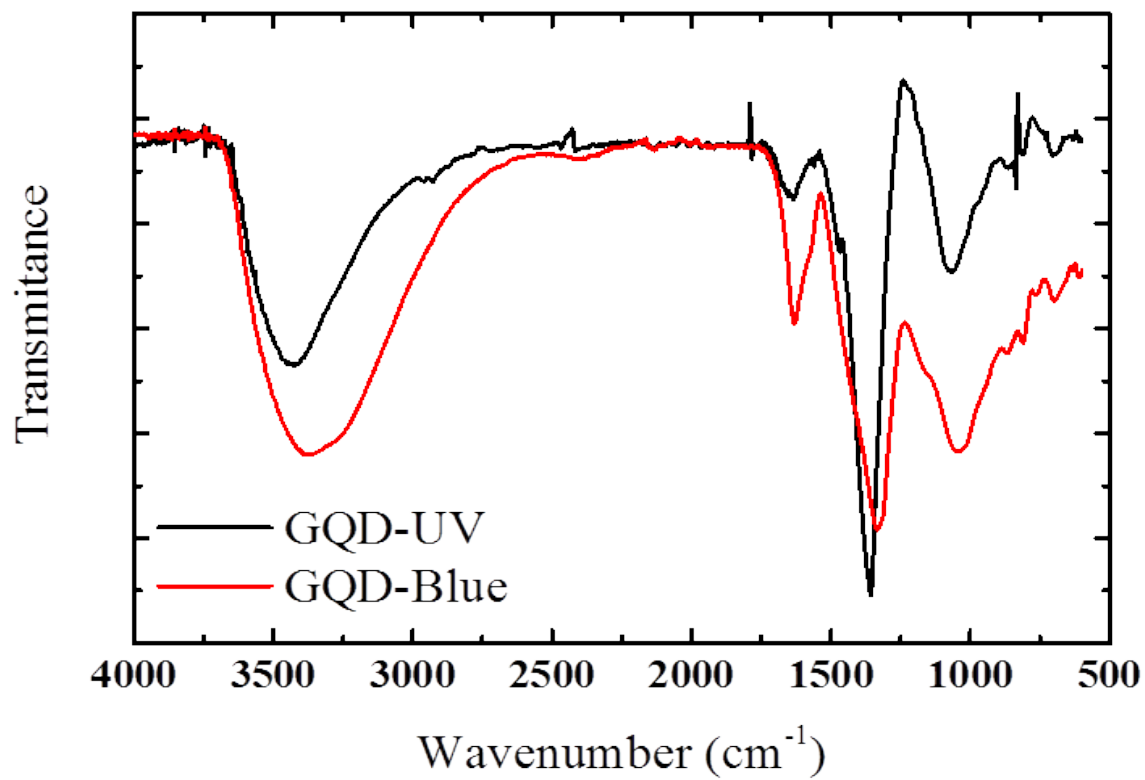


Fig. 2 FT-IR spectra of GQD-B (red) and GQD-UV (black).

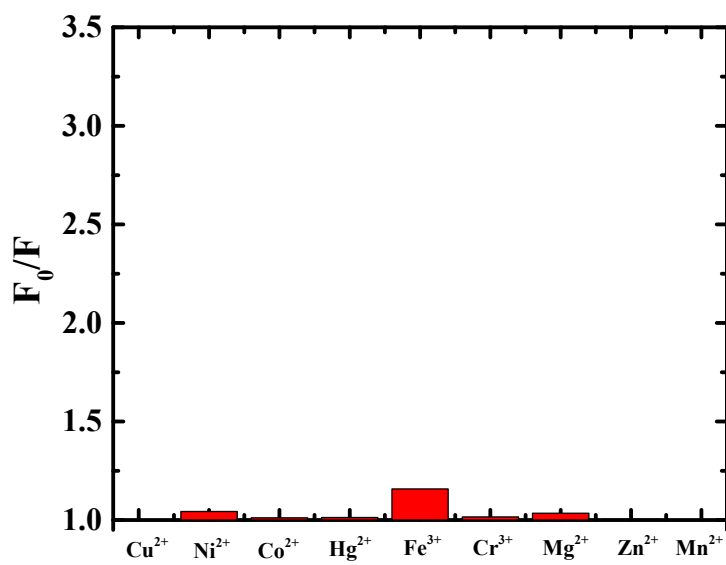


Fig. 3 Selectivity of GQD-UV for the detection of metal ions. The concentration of metal ions was 50 μM .

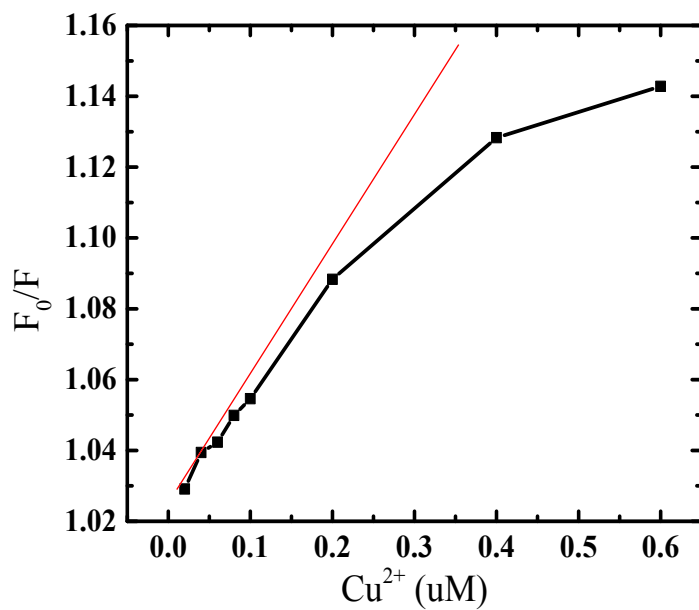


Fig. 4 The plot of F_0/F vs. Cu^{2+} concentration. F_0 is the fluorescence intensity of GQD-B in the absence of Cu^{2+} and F is the fluorescence intensity of GQD-B in the presence of Cu^{2+} . The non-linear relationship of F_0/F vs. Cu^{2+} concentration suggests that collision-induced quenching is not a main mechanism for the fluorescence suppression of GQDs.