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Supporting Information

S1 Experimental Section

Apparatus. Fluorescence measurements were carried out with a F-4500 fluorescence spectrophotometer (Hitachi, Japan). Morphology and size of GQDs were obtained by a FEI Tecnai G^2 20 transmission electron microscopy (USA) and an atomic force microscope from Molecular Imaging with PicoScan Controller. The UV-visible absorption spectra were recorded on a UV-2900 ultraviolet spectrophotometer (Hitachi, Japan). Zeta potential of GQDs were measured by a Zetasizer Nano ZS90 analyzer (Marvin, England). Fluorescence images of Hela cells were obtained by Olympus FV1000 confocal microscope (Japan).

Reagents. Graphene oxide was purchased from Nanjing XFNANO Materials Tech. Co. Ltd (Nanjing, China). DNA was purchased from Takara Biotechnology Co., Ltd (Dalian, China) with sequencesof 5'-(NH_2C6)TTCTTTCTTCCCTTgTTTgTT-3'. 1-ethyl-3-(3-dimethyllaminopropyl) carbodiimide hydrochloride (EDC) and N-hydroxysuccinimide (NHS) were purchased from Aladdin Industrial Inc. (Shanghai, China). Mercury sulfate, sodium hydroxide, sodium chloride and all the other reagents were of analytical grade without any further purification. Phosphate buffer solution was prepared by mixing stock solutions of Na_2HPO_4 and NaH_2PO_4. Milli-Q® ultrapure (18.2 MΩ) was used in the whole experiments.

Preparation of GQDs. GQDs were fabricated by a hydrothermal approach as follows: 40 mg GO was placed in a Teflon jar for 12 h at 200 ^oC to obtain reduced GO. 20 mg

reduced GO was oxidized with the concentrated HNO_3 (30mL) and H_2SO_4 (10mL) by stirring for 24 hours and ultrasonicating for 24 hours at room temperature. The mixture was diluted with 250 mL ultrapure water and filtered through a 0.22 μm microporous membrane to remove the residual acids. The filter was redissolved in 40mL ultrapure water and pH was tuned to 8.0 with NaOH. Finally, the solution was put into a Teflon jar and kept at 200 °C in the Muffle furnace for 24 hours. The resulting solution was filtered through a 0.22 μm microporous membrane to remove large particles and then collected the brown filtrate. The filtrate was further dialyzed 3 times with dialysis bag (retained molecular weight: 3500Da).The final product GQDs showed blue photoluminescence.[1,2]

Preparation of DNA-GQDs. To functionalize GQDs with DNAs, 0.58 mL GQDs were activated by a mixture of EDC/NHS (20/50 mM) for 20 minutes at pH 5.0, followed by addition of 0.1 mL DNAs (1 μM) in 10 mM phosphate buffer (pH=7.0) for the incubation of 2 hours at room temperature. The unreacted DNAs was removed by centrifugal ultrafiltration tubes (retained molecular weight: 3KDa). The obtained DNA-GQDs solution were stored at 4 °C.

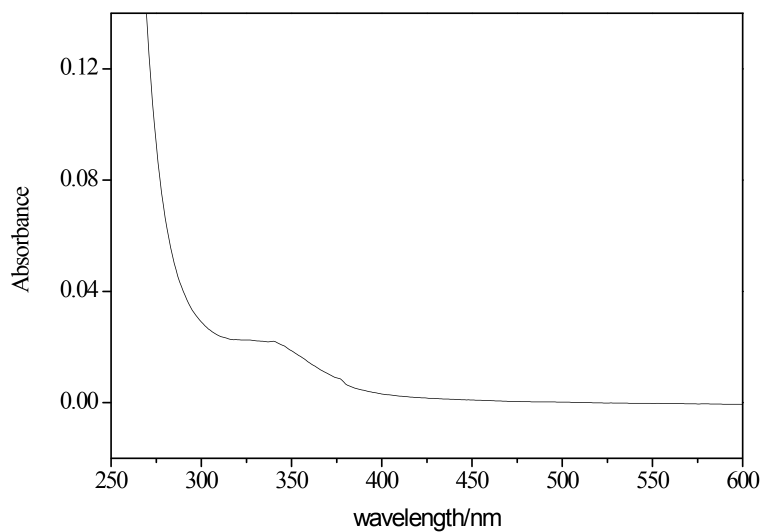
Detection of Hg^{2+} in aqueous solution. 150 μL DNA-GQDs was mixed with 390 μL PB buffer (20 mM, pH=7.4) and different concentrations of Hg^{2+}, followed by addition of ultrapure water to a total volume of 600 μL for incubating half an hour at room temperature. The mixture was analyzed by a fluorescence spectrophotometer. The fluorescent intensity was recorded from 350 to 600 nm with excitation wavelength at 320 nm. Similar procedures were performed for other metal ions.

Detection of Hg^{2+} in living cells. Hela cells (human cervical cancer cells) were grown in H-DMEM (Dulbecco's Modified Eagle's Medium, high glucose) supplemented with 10% fetal bovine serum at 37 °C, 5% CO_2 and humid atmosphere.[3,4] Cells were plated on glass coverslips and allowed to adhere for 24h. The living cells were first incubated in new medium which has contained DNA-GQDs for 30 min, then the cells were washed three times with PBS solution (pH=7.4). 0 μM, 1 μM, 10 μM, and 20 μM Hg^{2+} was added in the medium to culture cells for another half an hour, respectively, followed by washing three times with PBS solution. After that, fluorescence images of the cells were observed with laser confocal microscopeat irradiation of 405 nm with a band path from 425 nm to 520nm under identical exposure condition.

Viability assay by MTT. Hela cells in logarithmic phase were collected and suspended in complete media with a concentration of 50000 cells/mL. 100 μL cells suspensions were added into each well and incubated for 24 h in a incubator. 250 μg/mL and 50 μg/mL of DNA-GQDs or GQDs (final concentration) were added into each well and incubated for another 24 h, followed by addition of 20 μL of MTT (5 mg/L) into the wells for 4 h. After that, the media in wells were carefully removed, followed by addition of 150 μL DMSO into each well and incubated at 37 ℃ for 10 minutes. Absorbance value (A) of each well were determined by a microplate reader under 490 nm. Zero wells (media, MTT, DMSO) and control wells (cells, PBS for DNA-GQDs or GQDs, media, MTT, DMSO) were carried out at the same conditions. The cell viability was calculated by the formula as follows:

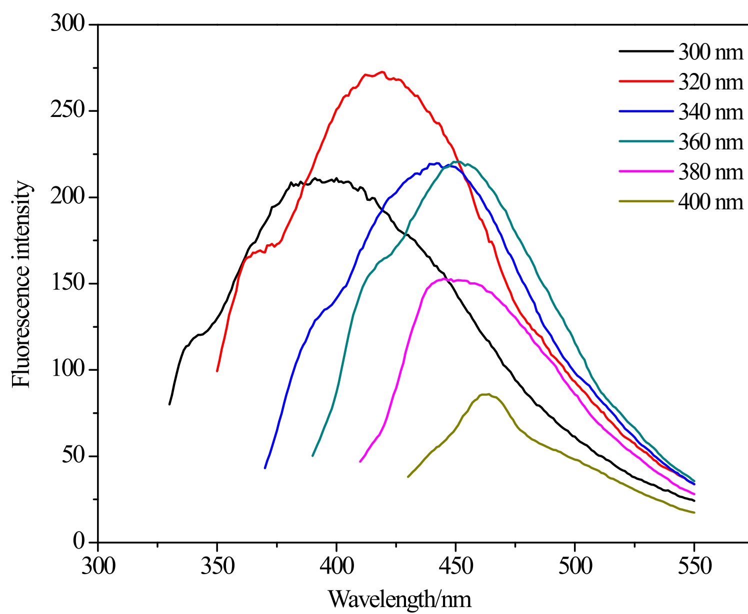
Cell viability = [experimental wells (A) – zero wells (A)] / [control wells (A) – zero wells (A)]×100%.

Fig. S1



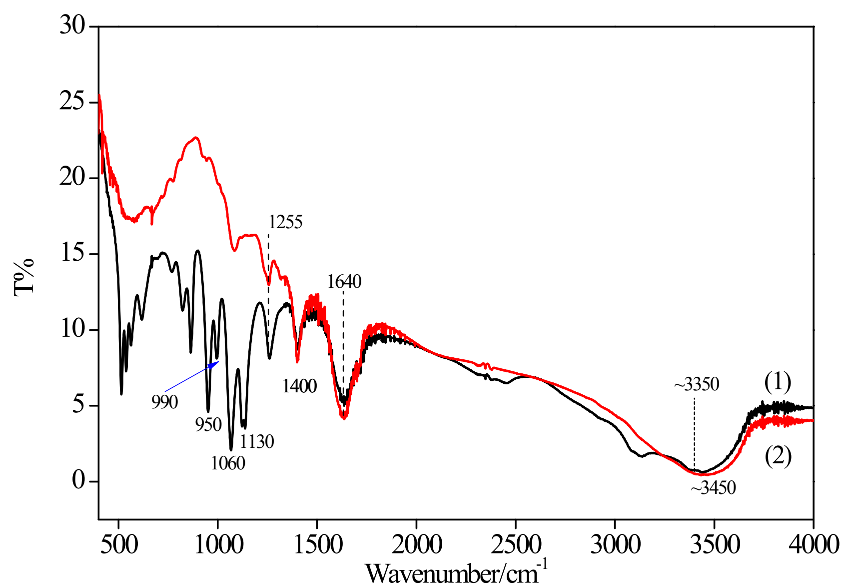
UV-absorbance of GQDs solution

Fig.S2



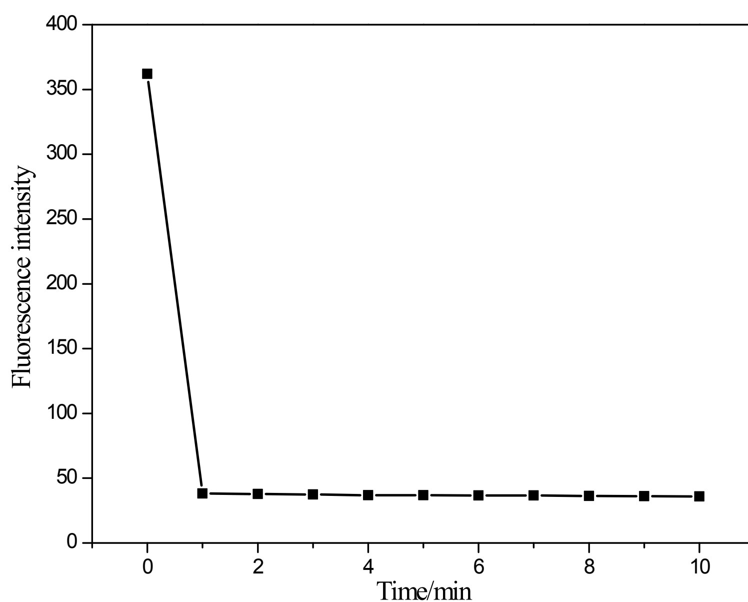
Fluorescent emission profiles of GQDs at different excitation wavelengths in 10 mM PBS buffer (pH 7.4)

Fig.S3



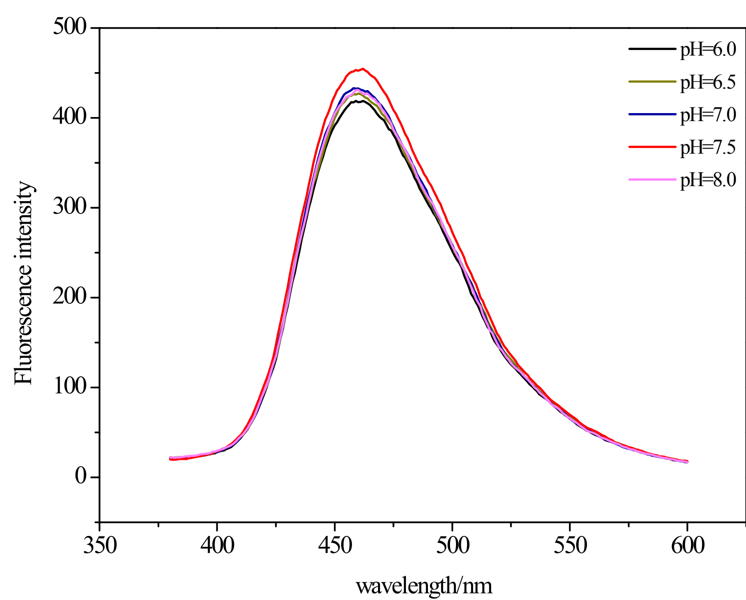
FTIR spectra of ~~GODs~~ GQDs (red) and ~~GODs~~GQDs-DNA (black) the DNA modification. The FTIR samples were prepared with KBr tablet after freeze-drying of ~~GODs~~ GQDs and ~~GODs~~GQDs-DNA solution.

Fig.S4



Fluorescence intensity of DNA-GQDs after addition of Hg^{2+} *versus* time

Fig. S5



The quenching dynamics of DNA-GQDs by Hg^{2+} in 10 mM PBS buffer

Table S1 A comparison between the present method and similar methods for the detection of Hg2+ based on GQDs or carbon nanoparticles (CNPs).

Probes	Analytes	Linear range	LOD/nM	Ref.
GQDs	Pb^{2+}	9.9~535 nM	0.6	5
CNPs	Hg^{2+}	5~20 nM	2.6	6
GQDs	Hg^{2+}	0.8~9 μM	100	7
N-doped CNPs	Hg^{2+}	0~25 μM	230	8
CNPs	Hg^{2+}	0~3 μM	4.2	9
CNPs	Hg^{2+}	0~3 μM	42	10
CNPs	Hg^{2+}	0~3 μM	0.25	11
O/N-doped CNPs	Hg^{2+}	0.001~20 μM	0.37	12
CNPs	Hg^{2+}	1~12 μM	226	13
CNPs	Hg^{2+}	0~5 μM	10	14
GQDSs	Hg^{2+}	0~60 μM	3360	15
CNPs	Hg^{2+}	0.5~10 nM	0.5	16
CNPs	Hg^{2+}	10~100 nM	3.4	17
O-doped and N-riched carbon nanoribbons	Hg^{2+}, Ag^{+}	0.002~60 μM 0.005~80 μM	0.68 1.73	18
N-doped CNPs	Hg^{2+}	0.01~50 μM	3	19
CNPs	Hg^{2+}	10~500 μM	8.2	20
S/N-doped CNPs	Hg^{2+}	1nM~50 μM	0.05	21
GQDs	Hg^{2+}	1 nM~10 μM	0.25	Present method

Supporting References

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