Supporting Information

Nitrogen-Doped Graphene Quantum Dots@SiO_2 Nanoparticles as Electrochemiluminescence and Fluorescence Signal Indicators for Magnetically-Controlled Aptasensor with Dual Detection Channels

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Synthesis of the NGQDs. In brief, 50 mL aqueous solution containing 40 mg mL^{-1} of citric acid and 7.2 mg mL^{-1} of urea were transferred into an autoclave and were heated at 180 ^{o}C for 8 h. After cooling to room temperature, the pH of the NGQDs solution was tuned to 7.0 by using 5 mg mL^{-1} of NaOH. Then the NGQDs solution was mixed with excess ethanol, allowed it to stay there for 15 min, and then centrifuged at 13000 rpm for 5 min to remove impurities and excess urea. After purification, the resultant NGQDs was diluted to 50 mL with water for further use. Pure GQDs were also obtained under the same conditions without the addition of urea.

Preparation of the aptamer coupled Fe_3O_4@Au MBs (MB-aptamer). 200 μL of 100 μM freshly activated aptamer using TCEP was added to 40 mL of 5 mg mL^{-1} Fe_3O_4@Au MBs and sonicated for 1 min, and then incubated for 1 h with gentle shaking at 37 ^oC. After the salt aging, the MB-aptamer resultants were separated readily by a magnet to remove excess unbound aptamer. Finally, the MB-aptamer resultants were treated with 40 mL of 2 μM MCH and kept at 37 ^oC for 1 h, again under stirring to block the nonspecific binding sites. After magnetic separation and washing, the MB-aptamer resultants were then redispersed in 40 mL of Tris-HCl buffer.

Table S1 Comparison of the Present Aptasensor with Those Reported in the Literatures.

Detection method	Liner range (ng mL^{-1})	LOD (pg mL^{-1})	References
FL	0.02–10	10	(S1)
	10–80	9640	(S2)
	1–100	800	(S3)
	0.01–0.3	2	(S4)
	0.01–1	10	(S5)
	8–160	8000	(S6)
	0.01–100	3	This work
ECL	0.001–20	0.64	(S7)
	0.00002–40.3	0.004	(S8)
	0.05–500	0.02	(S9)
	0.0001–4.76	0.027	(S10)
	0.001–10	0.5	This work

Table S2 Results of ECL Detection of OTA in Peanuts (n=3).

Samples	OTA (ng mL^{-1})		RSD (%)	Recovery (%)
	Spiked	Found		
1	0.05	0.048	6.8	96.0
2	0.5	0.474	8.3	94.8
3	5	4.97	7.5	99.4

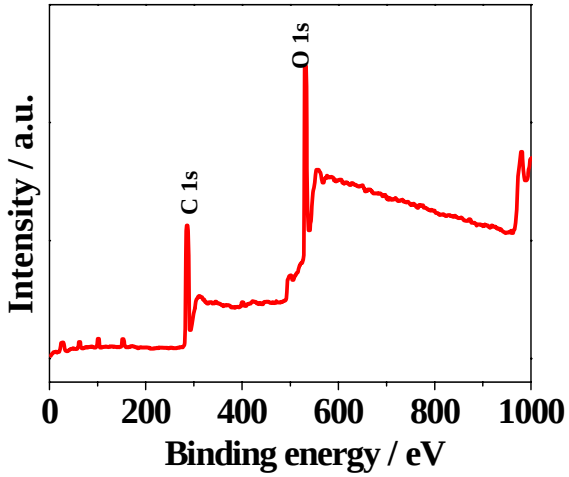

Figure S1. Wide scan XPS spectrum of the pure GQDs.

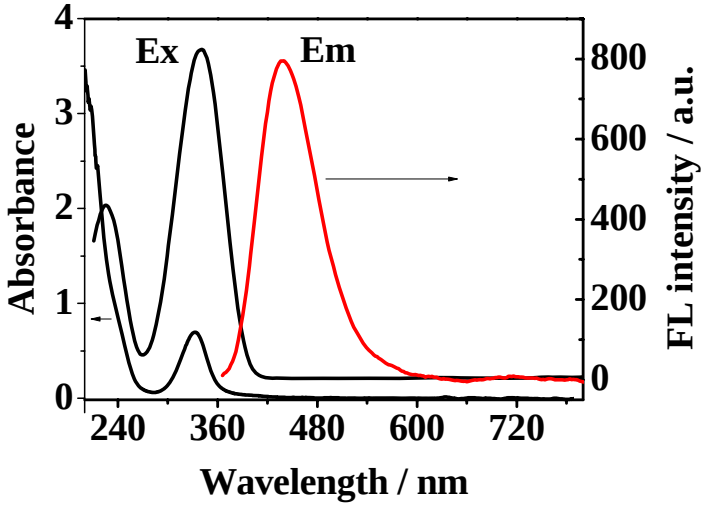

Figure S2. UV-vis absorption spectrum and the FL excitation (Ex) and emission (Em) spectra of the as-prepared NGQDs in aqueous solution.

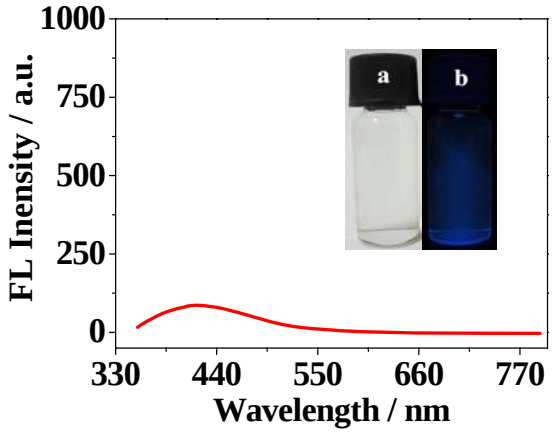

Figure S3. FL spectrum of the pure GQDs. Inset: photographs of the aqueous dispersed GQDs solution under daylight (a) and 365 nm UV irradiation (b).

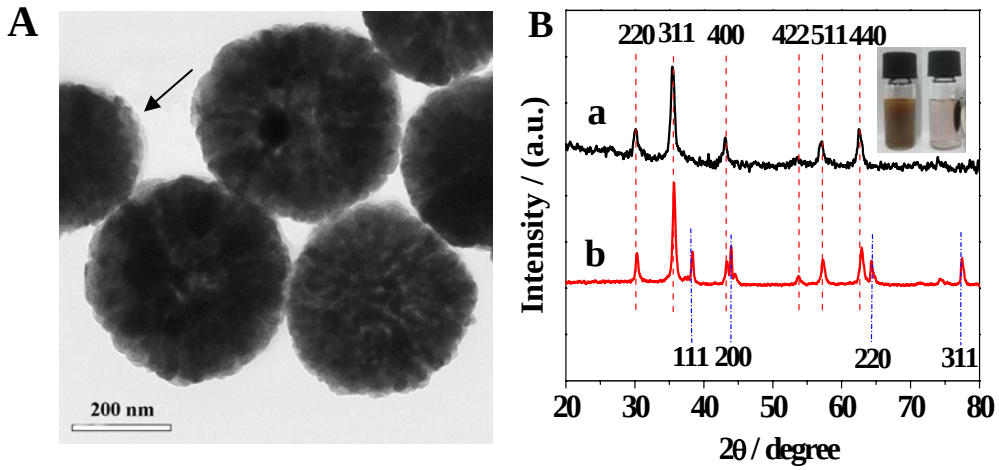

Figure S4. (A) TEM image of the as-prepared Fe_3O_4@Au MBs. (B) XRD patterns of Fe_3O_4 microspheres (a) and Fe_3O_4@Au MBs (b). Inset: photographs of the aqueous dispersed Fe_3O_4@Au MBs in absence (left) and presence (right) of an external magnetic field.

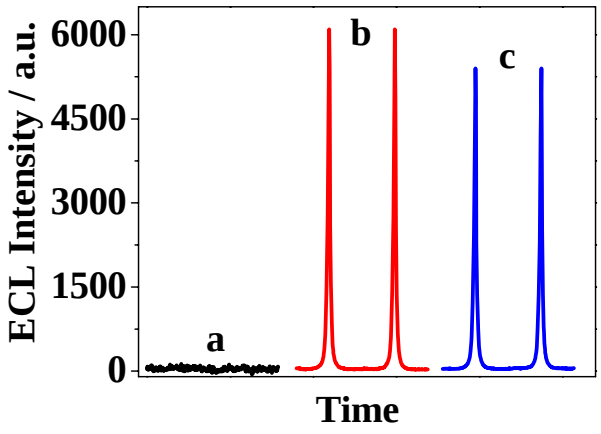

Figure S5. ECL response of the MB-aptamer/cDNA-NGQDs@SiO_2 modified mGCE in the air-saturated 0.1 M pH 7.4 Tris-HCl buffer containing 0.1 M KCl in the absence (a) and presence (b) of 0.1 M $K_2S_2O_8$. ECL response of the MB-aptamer/cDNA-NGQDs@SiO_2 modified mGCE in the nitrogen-saturated 0.1 M pH 7.4 Tris-HCl buffer containing 0.1 M KCl and 0.1 M $K_2S_2O_8$ (c).

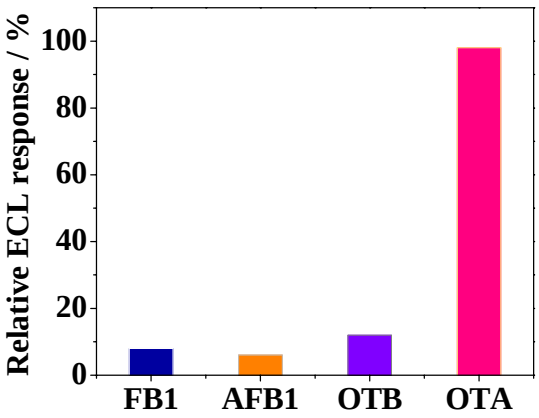

Figure S6. The selectivity of the aptasensor. The concentration of OTA is 5 ng mL^{-1} while that of FB1, AFB1 and OTB is 10 ng mL^{-1}.

References

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