Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Multicolor Fluorescent Graphene Quantum Dots Colorimetrically Responsive to All-pH and Wide Temperature Range

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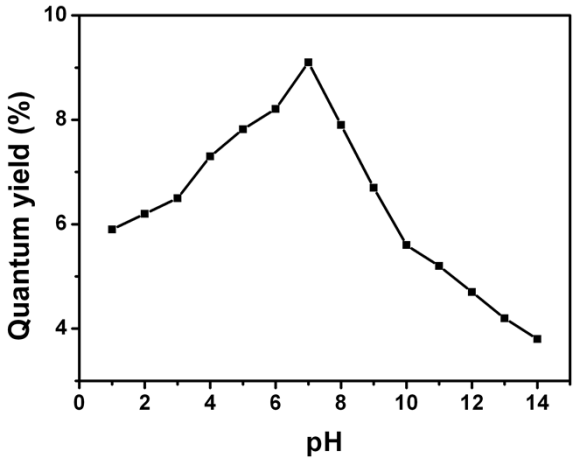

Figure S1. The influence of pH value on the fluorescence QY of MCF GQDs.

The QY of MCF GQDs gradually increased from 5.9% (pH 1) to the highest value about 9.1% (pH 7), and then gradually decreased to 3.8% (pH 14).

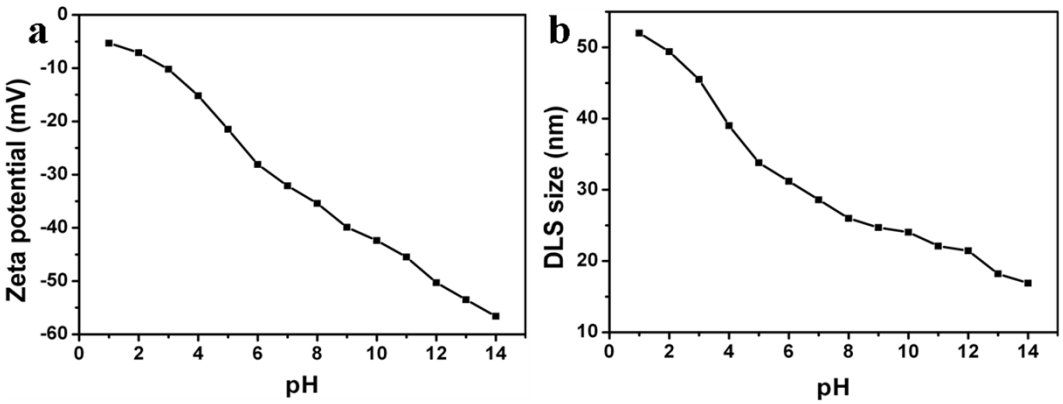

Figure S2. The influence of pH value on the Zeta potential and DLS size of MCF GQDs.

The Zeta potential of MCF GQDs gradually increased from -5.30 mV (pH 1) to -56.60 mV (pH 14) and DLS size gradually decreased from 52 nm (pH 1) to 16.9 nm (pH 14) due to the deprotonation of oxygen containing functional groups.

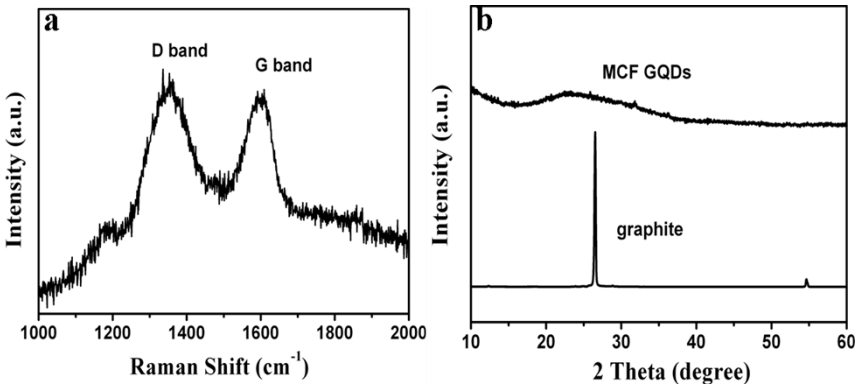

Figure S3. Raman spectrum (a) and XRD patterns (b) of MCF GQDs and graphite.

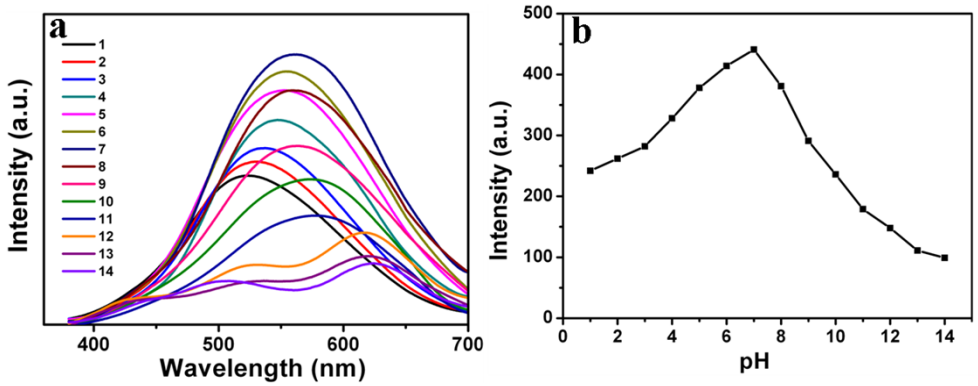

Figure S4. (a) The fluorescence spectra of MCF GQDs (excited at 365 nm) in different pH solutions. (b) The relationship of the fluorescence intensity of MCF GQDs to the solution pH values.

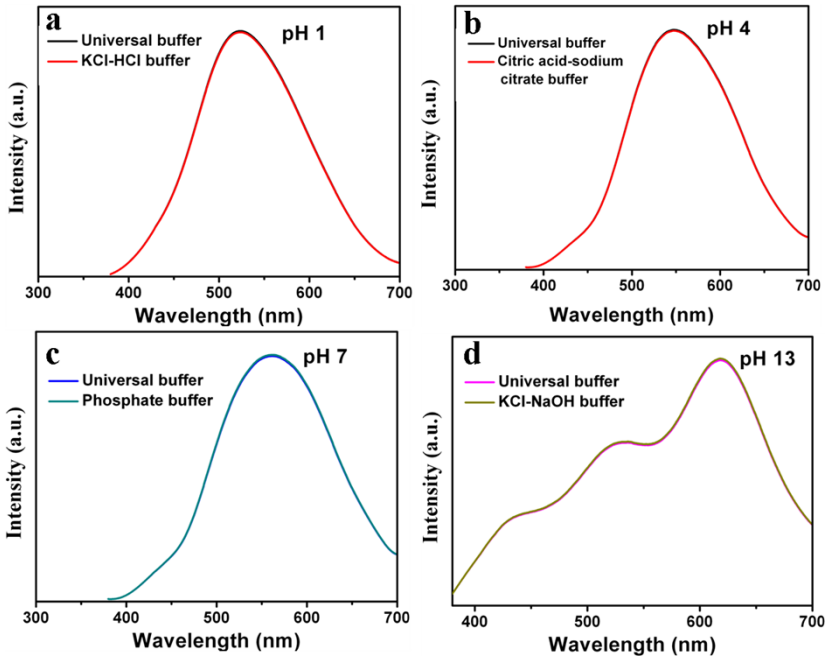

Figure S5. The fluorescence spectra of MCF GQDs in different buffer solutions of different pH values.

The fluorescence spectra of MCF GQDs in different buffer solutions of different pH values show almost the same fluorescence spectra as that in universal buffer solution of corresponding pH values. This indicates the obvious pH-responsive property is not resulting from the interaction between GQDs and buffer solution.

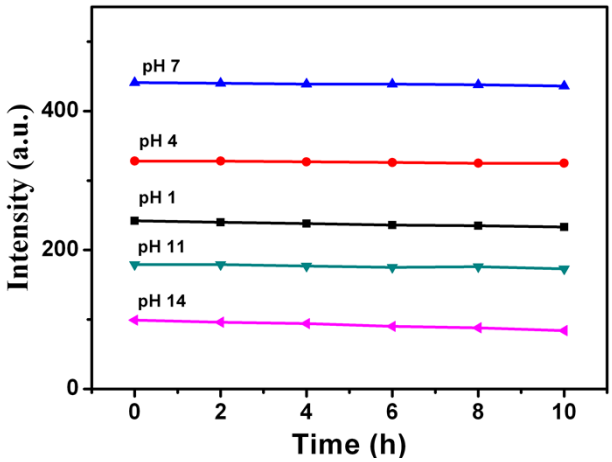

Figure S6. Photostability of MCF GQDs in different pH values irradiated with a 150 W Xe lamp with different time.

Table S1. The influence of pH value on the fluorescence lifetimes of MCF GQDs.

pH	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Fluorescence lifitimes (ns)	6.06	6.53	7.02	7.92	8.16	8.36	8.41	8.27	7.73	6.25	5.34	5.01	4.78	4.23

The average fluorescence lifetimes of MCF GQDs gradually increased from 6.06 ns (pH 1) to the highest value about 8.41 ns (pH 7), and then gradually decreased to 4.23 ns (pH 14).

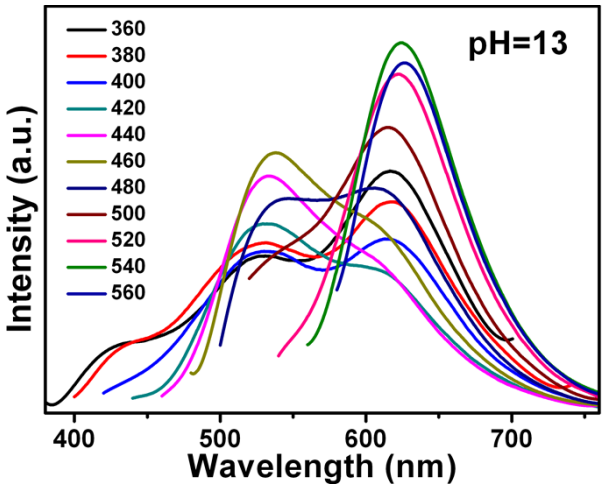

Figure S7. The fluorescence spectra of MCF GQDs in pH 13 solution.

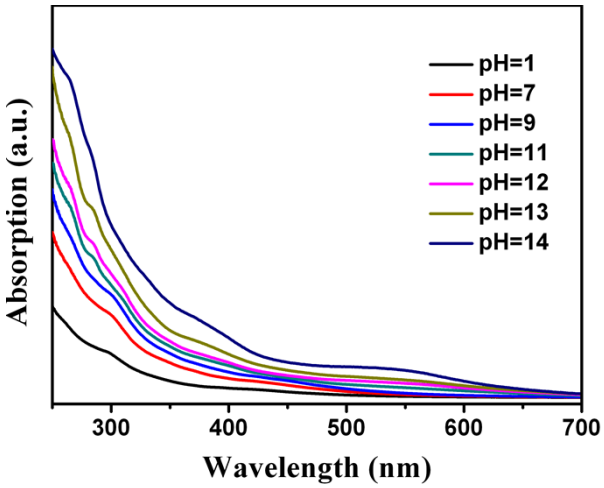

Figure S8. The UV-vis absorption spectra of MCF GQDs in different pH solutions.

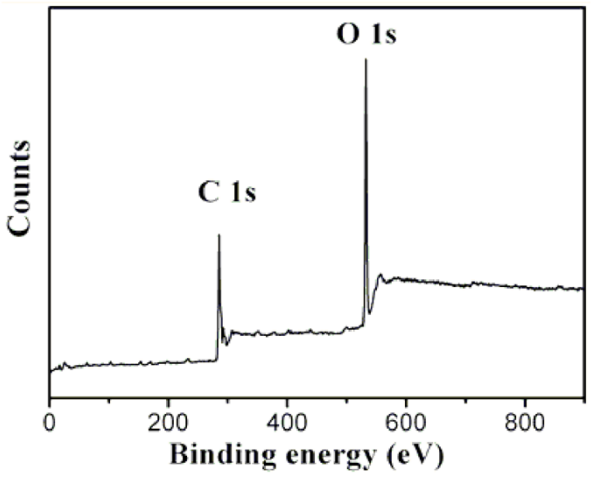

Figure S9. XPS full spectrum of MCF GQDs in neutral condition.

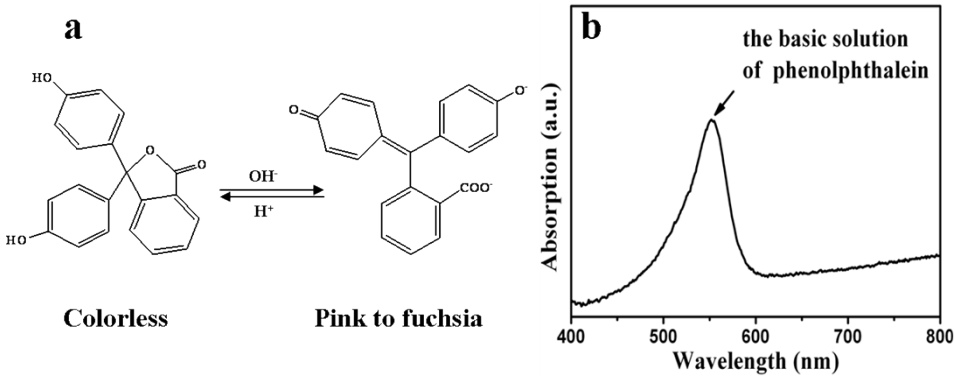

Figure S10. (a) Structure evolution of phenolphthalein from acidic or near-neutral to alkaline condition. (b) UV-vis absorption spectrum of phenolphthalein in alkaline condition.

Table S2. The optimized Cartesian Coordinates (in Å) of species studied.

Species	Cartesian coordinates			Species	Cartesian coordinates		
Ground state	C	0.72518	0.91915	0.30628	Excited state	C	0.18496	0.86591	0.46147
	C	-0.70470	0.75578	0.29653		C	-1.21405	0.62136	0.54540
	C	-1.20008	-0.51527	0.55154		C	-1.57347	-0.72794	0.90218
	C	-1.52772	1.90878	-0.07585		C	-2.13404	1.66685	0.15651
	C	-2.74097	1.77554	-0.81138		C	-3.41995	1.41969	-0.42144
	C	-3.49006	2.92492	-1.07779		C	-4.26054	2.49942	-0.69033
	C	-1.01346	3.19943	0.19808		C	-1.69030	3.01422	0.28526
	C	-1.76944	4.32590	-0.12063		C	-2.54127	4.06493	-0.03834
	C	-3.02036	4.18851	-0.72401		C	-3.83859	3.81562	-0.49528
	C	0.35964	3.34532	0.82631		C	-0.29533	3.28945	0.81560
	C	1.30901	2.22185	0.46820		C	0.70017	2.19744	0.49812
	H	-1.37472	5.31523	0.09837		H	-2.19067	5.08902	0.07185
	C	1.53382	-0.19895	0.18764		C	1.07523	-0.21278	0.28916
	C	-0.34775	-1.64228	0.25180		C	-0.69102	-1.75762	0.50867
	O	0.99779	-1.44273	0.07209		O	0.60390	-1.48480	0.17339
	C	2.66643	2.35874	0.42273		C	2.04975	2.41160	0.35361
	H	3.12346	3.33638	0.55740		H	2.44406	3.42476	0.38903
	C	-0.86269	-2.88063	-0.02486		C	-1.10990	-3.07751	0.37715
	C	-2.29743	-3.05129	-0.00257		C	-2.50928	-3.39363	0.63579
	H	-0.23595	-3.66525	-0.43433		H	-0.45806	-3.83321	-0.04726
	H	-4.43233	2.80891	-1.60331		H	-5.24452	2.28810	-1.09713
	H	0.79207	4.31424	0.55386		H	0.06595	4.25781	0.45191
	C	-3.20760	0.48528	-1.49760		C	-3.87152	0.05263	-0.92755
	O	-4.43420	0.19479	-1.41698		O	-5.06101	-0.30381	-0.68705
	O	-2.33362	-0.18878	-2.11315		O	-3.02323	-0.63572	-1.56911
	O	-2.91125	-3.91138	-0.68754		O	-3.06487	-4.38225	0.07695
	H	-3.60882	5.07199	-0.95732		H	-4.50187	4.64341	-0.73018
	H	0.25567	3.36181	1.92428		H	-0.34037	3.37924	1.91498
	Na	-3.74829	-1.96638	-1.96840		Na	-4.30794	-2.44944	-1.10624
	C	-2.55127	-0.81027	0.93392		C	-2.80504	-1.12924	1.48256
	C	-3.05425	-2.05948	0.76808		C	-3.24394	-2.43229	1.42656
	C	4.91849	1.35851	0.17014		C	4.34071	1.55650	0.01324
	H	5.35845	2.35025	0.25001		H	4.71397	2.57805	0.03693
	C	3.79119	-1.20769	-0.02836		C	3.37761	-1.08114	-0.04788
	C	3.52968	1.22716	0.23324		C	2.97267	1.33316	0.16597
	H	3.34885	-2.19565	-0.10745		H	3.00347	-2.09979	-0.07161
	C	2.95932	-0.09654	0.12914		C	2.47827	-0.02638	0.13696
	C	7.98469	-0.72463	-0.21113		C	7.51919	-0.33299	-0.51358
	C	7.41239	-2.02533	-0.31141		C	7.03032	-1.66215	-0.54527
	C	7.18509	0.37858	-0.05497		C	6.65022	0.72249	-0.33160
	H	8.06257	-2.88685	-0.43501		H	7.72438	-2.48538	-0.68927

	H	7.62143	1.37153	0.02162		H	7.02221	1.74387	-0.30682
	C	6.05277	-2.19094	-0.25315		C	5.68363	-1.91544	-0.39522
	C	5.76474	0.24969	0.01016		C	5.25664	0.49953	-0.17344
	H	5.61128	-3.18149	-0.33003		H	5.30884	-2.93558	-0.42052
	C	5.18514	-1.06806	-0.09099		C	4.75825	-0.85193	-0.20528
	H	9.06427	-0.61192	-0.25966		H	8.58282	-0.14841	-0.63345
	H	-3.16349	-0.00795	1.33236		H	-3.44204	-0.36266	1.91420
	H	-4.07952	-2.30371	1.03333		H	-4.21540	-2.72127	1.81853

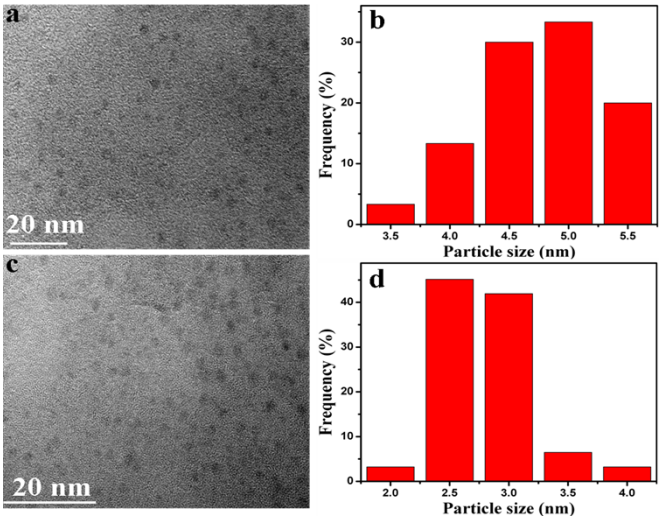

Figure S11. TEM images and size distribution of MCF GQDs-100 °C (a, b) and MCF GQDs-140 °C (c, d).

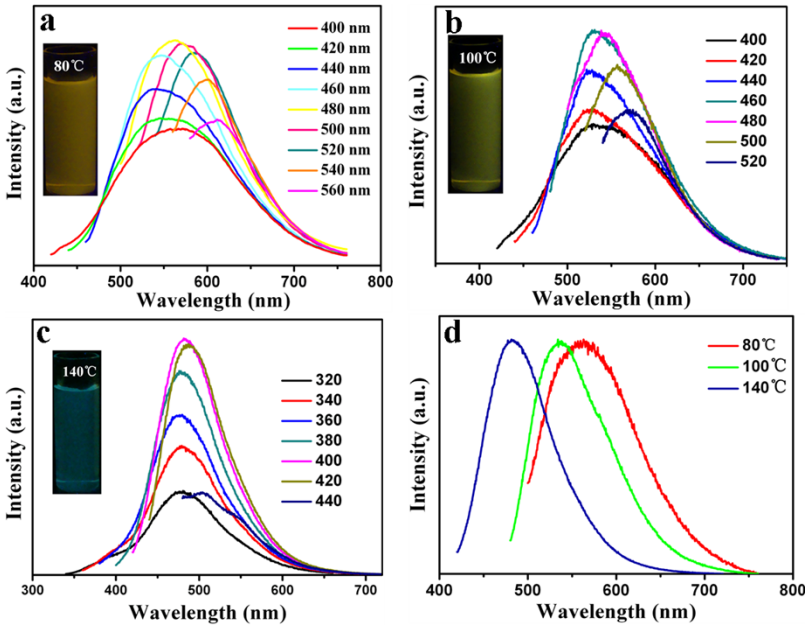

Figure S12. (a, b, c) Fluorescence spectra of MCF GQDs prepared at different reaction temperature in aqueous solution excited under different excitation wavelengths. Insets show photographs of relative MCF GQDs under UV light (excited at 365 nm) at room temperature. (d) The normalized maximum fluorescence emission spectra of MCF GQDs prepared at different reaction temperature in aqueous solution.

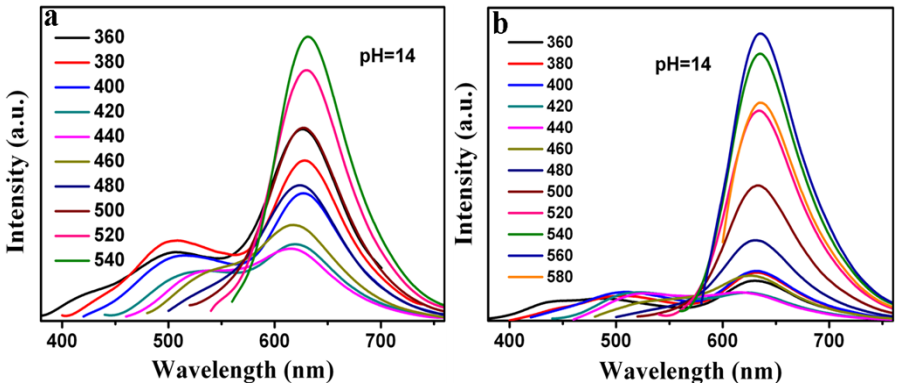

Figure S13. Fluorescence spectra of MCF GQDs prepared at 100 °C (a) and 140 °C (b) in strong alkaline solution excited under different excitation wavelengths.

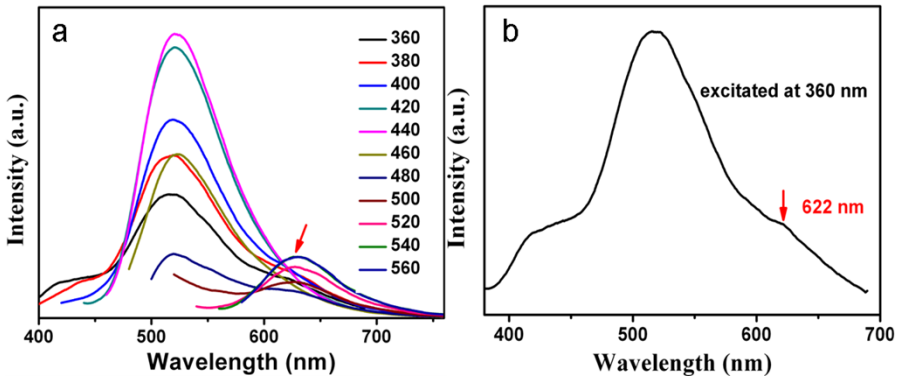

Figure S14. GQDs prepared through oxidation of graphite powder in strong acid media at 120 °C for 24 h. The fluorescence spectra of GQDs in strong alkaline condition excited under different excitation wavelengths (a), excited at 360 nm (b).

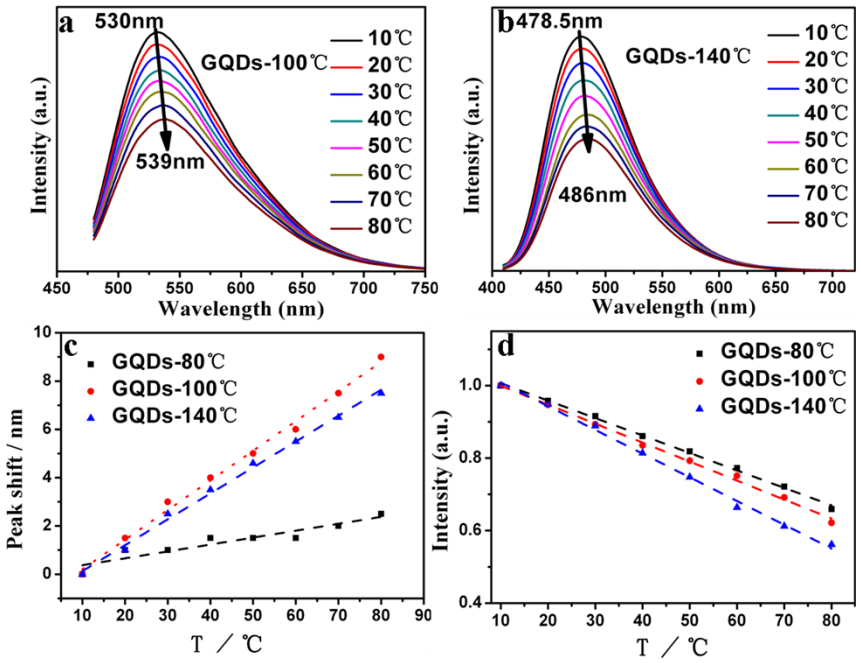

Figure S15. The fluorescence spectra of MCF GQDs-100 °C (a) and MCF GQDs-140 °C in neutral condition in the temperature range 10-80 °C. Temperature-induced peak shift (c) and fluorescence intensity decrease (d) for MCF GQDs obtained from different temperatures.

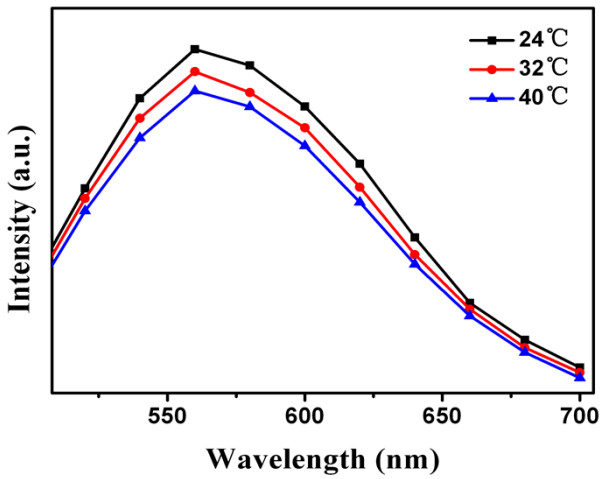

Figure S16. The fluorescence signal of MCF GQDs decreased as increasing the imaging temperature from 24 °C to 40 °C in Hela cells.