Supporting Information

Multifunctional PLA-PEG-grafted Graphene Quantum Dots for Intracellular MicroRNA Imaging and Combined Specific-Gene-Targeting Agents Delivery for Improved Therapeutics

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Materials and Reagents: Graphite was purchased from XF NANO (Nanjing, China), terminal amino polythylyene glycol (NH_2-PEG-NH_2) was synthesized by Sinopeg Biotech Co., Ltd (Xiamen, China), and carboxyl-terminated polylactic acid (COOH-PLA) was obtained from Daigang Biomaterial Co., Ltd (Jinan, China). 1-ethyl-3-(3-dimethylami-nopropyl) carbodiimide (EDC), N-hydroxy-succinimide (NHS) and MTT were purchased from Sigma-Aldrich (China). Sulfuric acid (H_2SO_4, 98%) and nitric acid (HNO_3, 65%) were obtained from Sinopharm Chemical Reagent Co., Ltd (Beijing, China).All other reagents were of analytical grade. Annexin V FITC/propidium iodide (PI) cell apoptosis kit was obtained from KeyGen Biotech. Co., Ltd. (Nanjing, China).

The oligonucleotides were purchased from Sangon Biological Engineering Technology & Co., Ltd (Shanghai, China) and purified using high-performance liquid chromatography. Their sequences were as follows:

Molecular beacon (MB) detection probe for miRNAs-21:5' FAM-TC TAG CAT CAG TCT GAT TCT GAT AAG CTA GCT AGA-Dabcyl 3'

Survivin antisense oligodeoxynucleotide (ASODN): 5'CCC AGC CTT CCA GCT CCT TG 3'

The RNA sequences were purchased from Shanghai Gene Pharma Co., Ltd. (Shanghai, PRC) and purified using high-performance liquid chromatography, which were listed as follows:

Inhibitor probe (IP)-21: 5' UCA ACA UCA GUC UGA UAA GCUA 3'

Characterization: The morphologies of GQDs and f-GQDs were examined with atomic force microscopy (NanoscopeIIIa, USA) and a JEM 2100 transmission electron microscope. AFM measurement was carried out on NanoscopeIIIa (Digital

Instrument, USA) under tapping mode. The UV−visible (UV-vis) absorption and X-ray-photoelectron spectroscopy analyses were recorded with an UV-1800 spectrophotometer (Shimadzu, Japan) and an ESCALAB 250 spectrometer (Thermo-VGScientific, USA), respectively. All fluorescence measurements were performed on a Hitachi F-4500 fluorescence spectrofluorometer (Tokyo, Japan) at room temperature, with an excitation wavelength of 320 nm. Zeta potential analysis was performed on Nano ZS (Malvern, UK).Fourier transform infrared spectra were recorded on Nicolet 400 Fouriertransform infrared spectrometer (Madison, WI) and electrochemical impedance spectra was obtained with a PGSTAT30/FRA2 system (Metrohm Autolab, The Netherlands)

Figure S1.

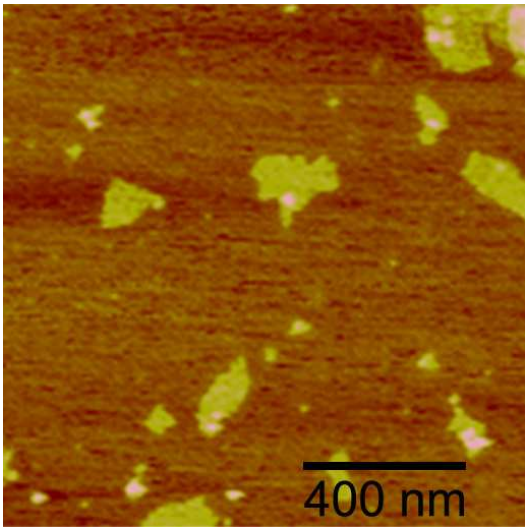

Figure S1. AFM of as-prepared GO

Figure S2.

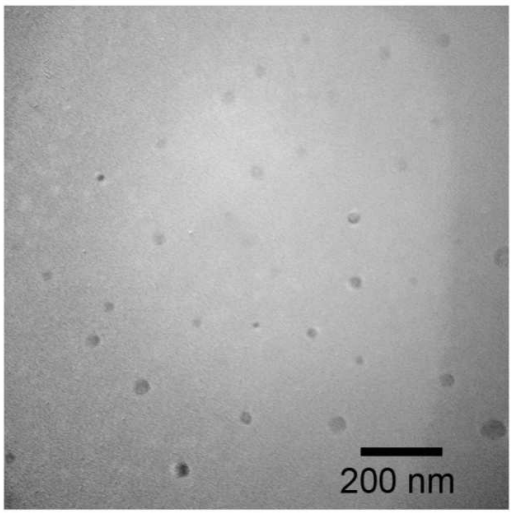

Figure S2. TEM images of as-prepared GQDs-PEG

Figure S3.

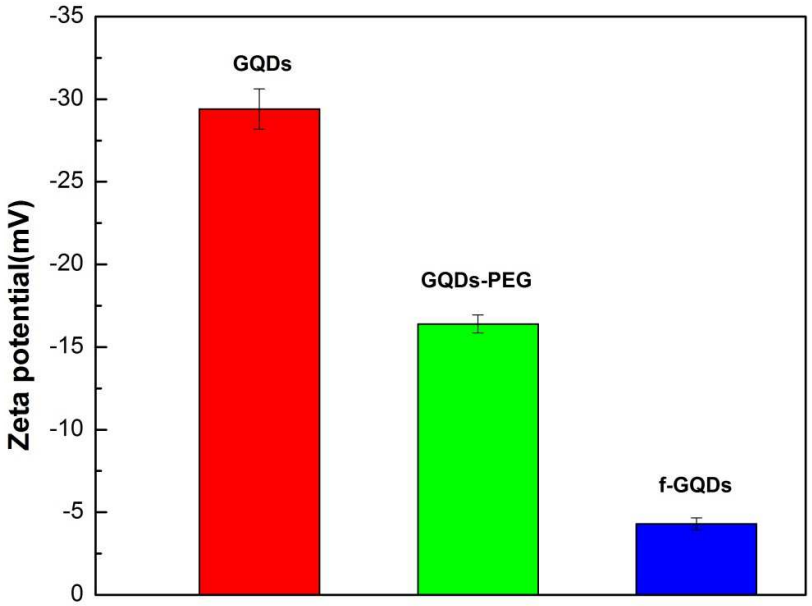

Figure S3. Zeta potential of GQDs, GQDs-PEG and f-GQDs.

Figure S4.

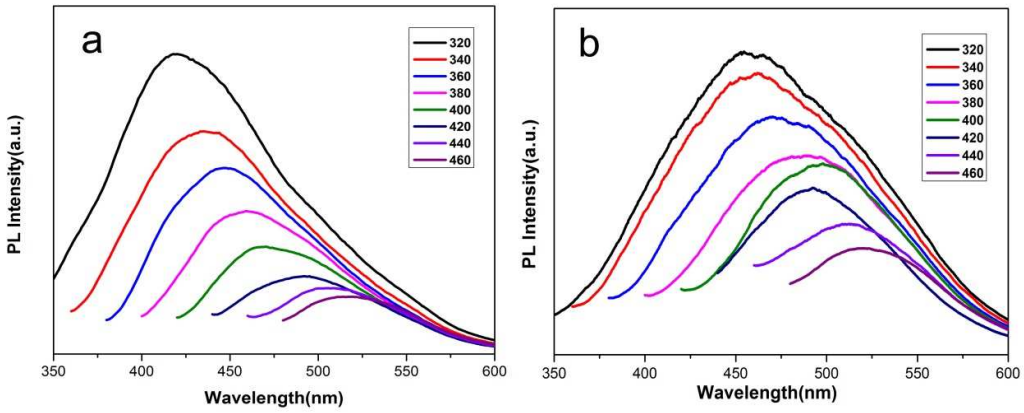

Figure S4. (a) PL spectra of the GQDs at different excitation wavelengths.(b) PL spectra of the as prepared f-GQDs at different excitation wavelengths.

Figure S5.

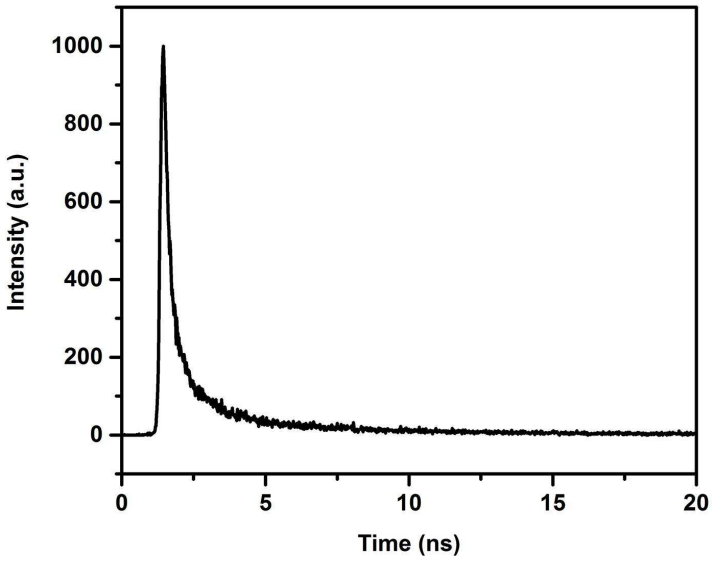

Figure S5. The time resolved fluorescence spectrum of f-GQDs.