## **Supporting Information**

### For

# DNA-Templated Nanoparticle Complex for Photothermal Imaging and Labeling of Cancer Cells

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#### 1.1 FRET efficiency determination



**Figure a.** Normalized absorption spectra of GNPs and PL emission spectra of QDs with emission maxima at 624 nm (top). Schematic illustration of the model used for calculation of FRET efficiency between GNP and QD (bottom).

The spectral overlap integral  $J(\lambda)$ , Förster distance  $R_0$ , and separation distance r are calculated according to the methods reported in *J. Am. Chem. Soc.* **2011**, 133, 10482-10489. The spectral overlap integral  $J(\lambda)$  is determined using the equation below:

$$J(\lambda) = \int_0^\infty \varepsilon_A(\lambda) \lambda^4 F_D(\lambda) d\lambda$$

where  $\varepsilon_A(\lambda)$  is the extinction coefficient of QDs in units of M<sup>-1</sup> cm<sup>-1</sup>,  $\lambda$  is the wavelength in cm and F<sub>D</sub>( $\lambda$ ) is the wavelength dependent donor emission spectrum normalized to an area of 1. For GQC,  $J(\lambda)=1.81\times10^{-10} cm^3 M^{-1}$ 

The Förster distance  $R_0$  is determined using the equation below:

$$R_0 = 9.78 \times 10^3 [\kappa^2 d^{-4} Q_D J(\lambda)]^{1/6}$$
  
$$R_0 = 13.5 \ nm$$

where  $\kappa^2$  is the dipole orientation factor, assumed to be 2/3, QD is the quantum yield of the donor, and *d* is the refractive index of the medium (*d*(H<sub>2</sub>O)=1.333).

The average number of acceptors per donor, n, as determined by absorbance spectroscopy, was taken into account. In the case where one donor species can interact with several acceptors the energy transfer efficiency can be expressed as:

$$E = nR_0^{6}(nR_0^{6} + r^6)$$

where r is the separation distance between the donor and the acceptor.

$$r = 10.8 \text{ nm}$$

In this plausible scenario the length of DNA molecule is estimated according to the parameters of Bform DNA duplex. The radius of QD is 1.9 nm For GQC, E = 0.79 (theroretical)

The FRET efficiency can be measured experimentally and can be calculated using the equation below.  $E = 1 - \frac{F_{DA}}{F_D}$ 

where  $F_D$  and  $F_{DA}$  designate the fluorescence intensities of the donor alone and donor in the presence of acceptor, respectively.

In QGC,  $\frac{F_{DA}}{F_D} = 0.08$ ; E = 0.92 (experimental)

The experimental FRET efficiency is slightly higher than the theoretical value, which could be attributed to a small portion of QDs associated with two or more GNPs.

#### **1.2 Melting temperature calculation**



The GC content (%) of the double-stranded DNA linker is 46.2%. The theoretically calculated Tm (137 mM NaCl) is 46.4 °C according IDT Oligo Analyzer 3.1 software, which is consistent with the experimental data.



**Figure S1.** Melting temperature measurement of free DNA linker and DNA linker in GQC. (a) The melting curve and (b) its first derivative of free DNA linker; (c) the first derivative of the melting curve of DNA linker in GQC.



**Figure S2.** Characterization of GNPs and DNA conjugation. (a) TEM image of GNPs; (b) Size statistics of GNPs; (c) Absorption spectra of GNPs before and after DNA1 conjugation; (d) TEM image of DNA1-GNPs; (e) Dynamic light scattering measurements of GNPs before and after DNA1 conjugation; (f) Standard curve for determination of average number of DNA1 on each GNP using MCH displacement method. DNA1 is labeled with TAMRA.



**Figure S3.** Characterization of DNA2-QDs. (a) Absorption and photoluminescence spectra of DNA2-capped CdTe QDs; (b) Low magnification and high-resolution TEM images of DNA2-QDs; (c) Size statistics of DNA2-GNPs; (d) Native PAGE of DNA2-QDs and DNA2-QDs with capture DNA.



Figure S4. Energy dispersive spectrum of GNP-QD complex.



**Figure S5.** Evaluation of the stability of GQC under different conditions. (a) Colloidal stability of GQC under different pH measured by absorption spectra (left) and emission spectra (right); (b) Colloidal stability of GQC under different ionic strength measured by absorption spectra (left) and emission spectra (right); (c) Nuclease stability of GQC with DNase I measured by absorption spectra (left) and emission spectra (right).



Figure S6. Characterization of as-prepared PT agents. (a) TEM images of PEG-GNRs, PEG-GNSs, and PVP-PBNPs. (b) Absorption spectra of PEG-GNRs, PEG-GNSs, and PVP-PBNPs.



**Figure S7.** PEG modification of GNRs. (a) UV-Vis spectra of as-synthesized GNRs and PEGylated GNRs; (b) Zeta potential of as-synthesized GNRs and PEGylated GNRs.



**Figure S8.** PEG modification of GNSs. (a) UV-Vis spectra of as-synthesized GNSs and PEGylated GNSs; (b) Zeta potential of as-synthesized GNSs and PEGylated GNSs.



**Figure S9.** Characterization of PVP-PBNPs. (a) Fourier transform infrared spectroscopy (FTIR) spectra of PBNPs (blue line) and PVP (red line). The PBNPs exhibited a FTIR absorption peak at 2085 cm<sup>-1</sup>, which is characteristic of CN stretching in the  $Fe^{2+}$ –CN– $Fe^{3+}$  bond of PB, and a peak at 1673 cm<sup>-1</sup>, indicative of C=O stretching in the PVP amide unit, which is consistent with that of pure PVP; (b) UV–vis spectra of PVP-PBNPs with known concentrations; (c) Calibration curve of PVP-PBNPs for concentration.



**Figure S10.** Analysis of non-specific adsorption of serum protein on nanoparticles. (a) Schematic illustration of isolation and characterization of protein adsorbed on nanoparticles. (b) SDS PAGE analysis of proteins adsorbed on PEG-GNRs, PEG-GNSs, and PVP-PBNPs. A band around 70 KDa was observed for all the three types of nanomaterials.



**Figure S11.** Cell viabilities of HeLa cells incubated with (a) GQC; (b) PEG-GNRs; (c) PEG-GNSs; (d) PVP-PBNPs measured by MTT assay.



**Figure S12.** A comparison of the photothermal performance of PEG-GNRs, PEG-GNSs, PBNPs in  $1 \times$  PBS and 10% FBS. (laser power = 1.0 W/cm<sup>2</sup>).



Figure S13. Cellular uptake of GQC with or without Lipofectamine-2000 for different incubation time.

Bright field



## TAMRA fluorescence



**Figure S14.** Fluorescence images of HeLa cells transfected with TAMRA-labeled capture DNA using Lipofectamine-2000.





Time (h)

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Time (h)



**Figure S16.** Preparation and characterization of PEI-FA-GQC-GNR complex. (a) Absorption spectra of FA, PEI, and PEI-FA conjugates; (b) FTIR spectra of FA, PEI, and PEI-FA. The formation of amide bond was depicted; (c) Absorption spectrum of PEI-FA-GQC-GNR complex; (d) zeta potential of GNRs/GQC, PEI-FA, and PEI-FA-GQC-GNR complex; (e) TEM image of PEI-FA-GQC-GNR complex; (f) hydrodynamic size of PEI-FA-GQC-GNR complex.



**Figure S17.** Quantitative analysis of cellular uptake of PEI-FA-GQC-GNR complex. (a) Schematic illustration of sample preparation for ICP-AES measurements. (b) Cellular uptake of complex measured by Au element.



**Figure S18.** Long-term imaging of HeLa cells treated with PEI-FA-GQC-GNR complex and NIR irradiation. (a) Fluorescence microscopy images of HeLa cells incubated for 0, 1, 3, 5, 7 days after treatment with PEI-FA-GQC-GNR complex and NIR irradiation; (b) Mean PL intensities of HeLa cells under the same treatments.