**Supporting Information** 

## Cancer theranosis using mono-disperse, mesoporous gold nanoparticles

## obtained via a robust, high-yield synthetic methodology

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Figure S1. Representative TEM image of WNBs synthesized in a neutral aqueous solution.



**Figure S2.** a) Size variation of MPGNs over 30 days and b) TEM images of MPGNs 30 days after synthesis



**Figure S3.** Relative synthetic yield of MPGNs with varying MPGN size. All yields were above 90% regardless of MPGN diameter. (a)  $131.0 \pm 2.7$  nm; b)  $309.6 \pm 7.72$  nm; c)  $391.9 \pm 12.0$  nm).



**Figure S4.** a) Relative synthetic yield and b) hydrodynamic diameter of MPGNs before and after 2x scale up. Synthetic yield was measured using ICP-OES, and hydrodynamic diameter was analyzed by light scattering. c) Representative TEM images of MPGNs after 2x scale up. The inset is the magnified MPGN image.



**Figure S5.** a) Size distribution of sGNPs and DTPA- and Gd-modified sGNPs using light scattering at room temperature. After surface modification, sGNPs size increased to  $170.1 \pm 11.3$  nm. b) Representative TEM images of sGNPs and DTPA- and Gd-modified sGNPs.



Figure S6. Three-pulse electron spin echo (ESE) field-sweep spectra of (a)  $GdCl_3$  and (b) CG-MPGNs.



Figure S7. <sup>1</sup>H-Mims ENDOR spectra of (a)  $GdCl_3$  and (b) CG-MPGNs. All spectra were taken at 8 K.



**Figure S8.** Size of CG-MPGNs at a) different pH conditions and b) varying fetal bovine serum (FBS) concentrations, as determined by laser scattering.



**Figure S9.** Cell viability test of various concentrations of CG-MPGNs (5 x  $10^{-11} - 5 x 10^{-1} mg/mL$ ) against A-431 (blue bar) and MCF-7 (gray bar) cell lines (1x10<sup>4</sup> cells each) using the 3-(4,5-dimethylthiazolyl-2)-2,5-diphenyltetrazolium bromide (MTT) assay at 37 °C in a 5% CO<sub>2</sub> atmosphere.



**Figure S10.** TEM image of A-431 and MCF-7 cells incubated with CG-MPGNs (inset, magnification of the CG-MPGNs in the cytopolasm). CG-MPGNs were internalized via receptor meditated endocytosis (appeared as black dots)



**Figure S11.**  $\Delta R_1/R_{1\text{control}}$  graph of A-431 (EGFR+) and MCF-7 (EGFR-) cell lines (1x 10<sup>7</sup> cells each) after treatment with different amounts of CG-MPGNs (0.5 mg/mL and 0.1 mg/mL, respectively) ( $\Delta R_1 = R_1 - R_{1\text{ctrl}}$ ). Blue bar graphs represent CG-MPGN-treated A-431 cells. Gray bar graphs represent CG-MPGN-treated MCF-7 cells.



**Figure S12.** Fluorescence microscopy images of A-431 (EGFR+) and MCF-7 (EGFR-) cells stained with calcein AM and ethidium homodimer-1 (EthD-1) after NIR laser irradiation for 10 min (808 nm, 25 W cm<sup>-2</sup>). White-dotted curves represent the location of the laser beam. The scale bar represents 200  $\mu$ m.