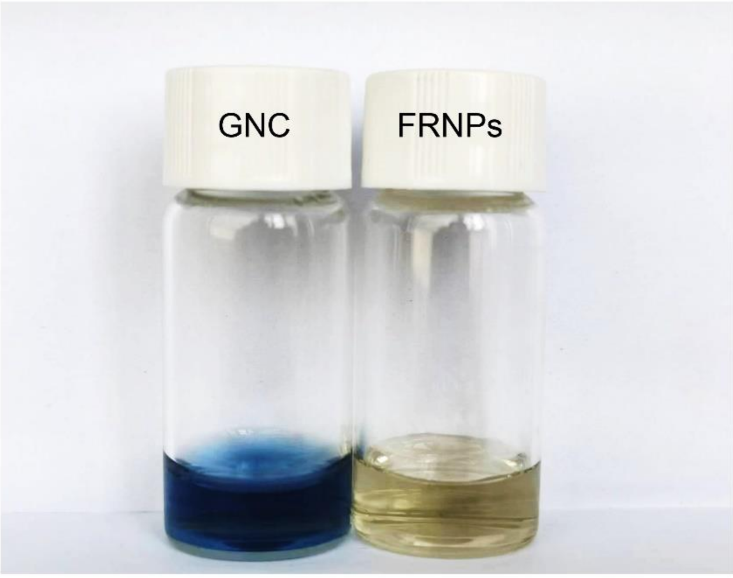

Figure S1. Photograph of the solutions of GNCs and resulting FRNPs, respectively.

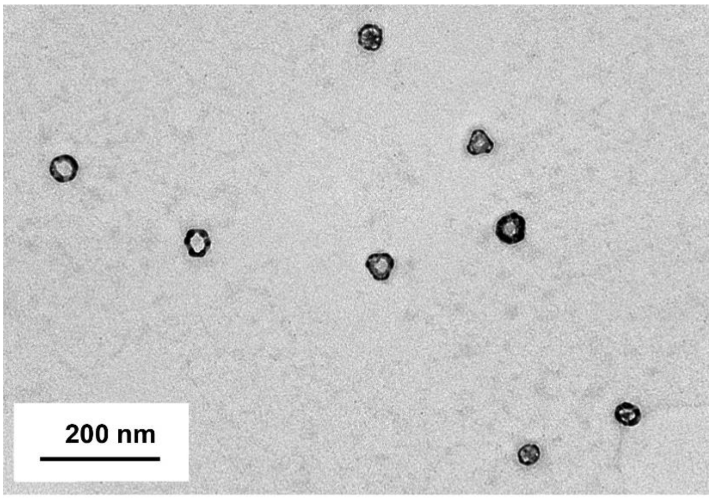

Figure S2. TEM image of FRNPs (scale bar = 200 nm).

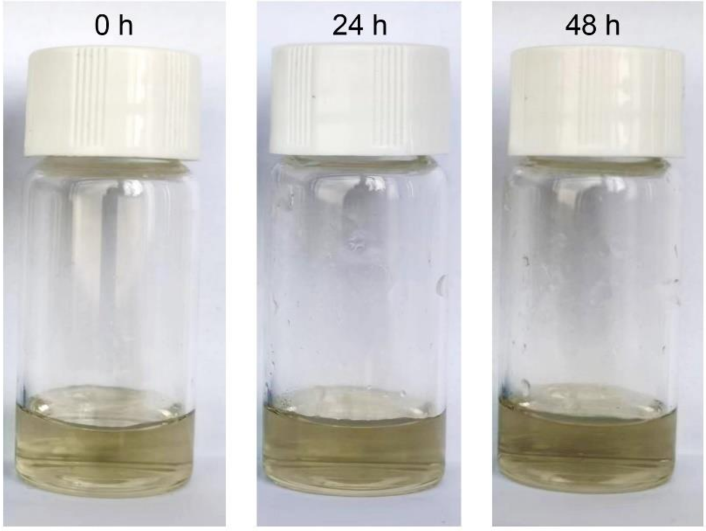

Figure S3. Photographs of aqueous solution of FRNPs at 0 h, 24 h, and 48 h.

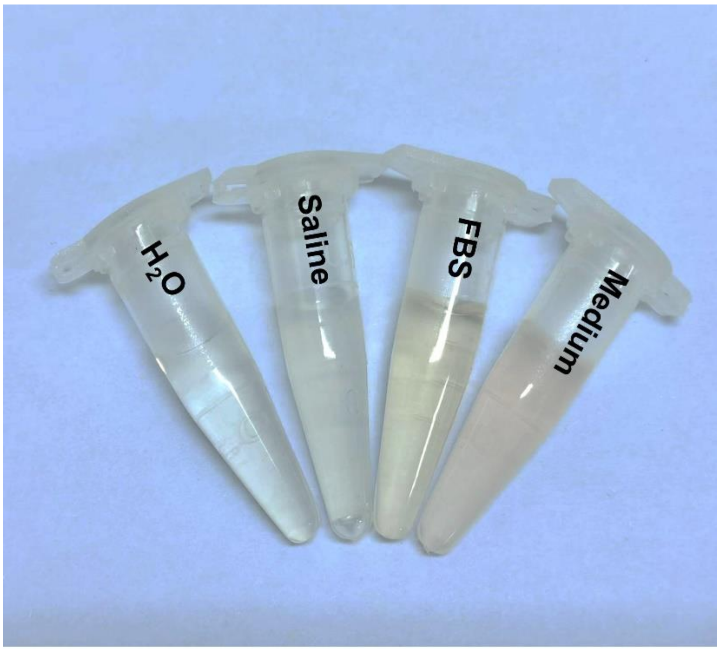

Figure S4. Photograph of FRNPs dispersed in water, saline, serum and cell culture medium.

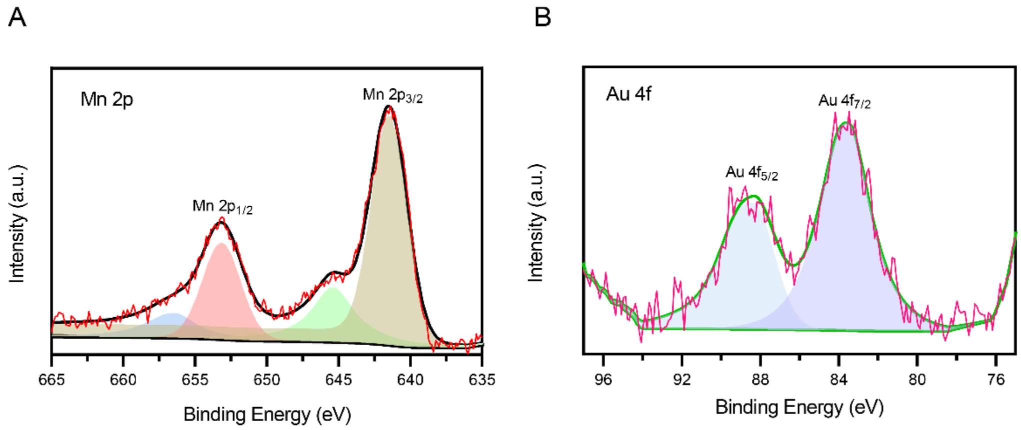

Figure S5. XPS spectrum of Mn 2p (A) and Au 4f (B) for FRNPs.

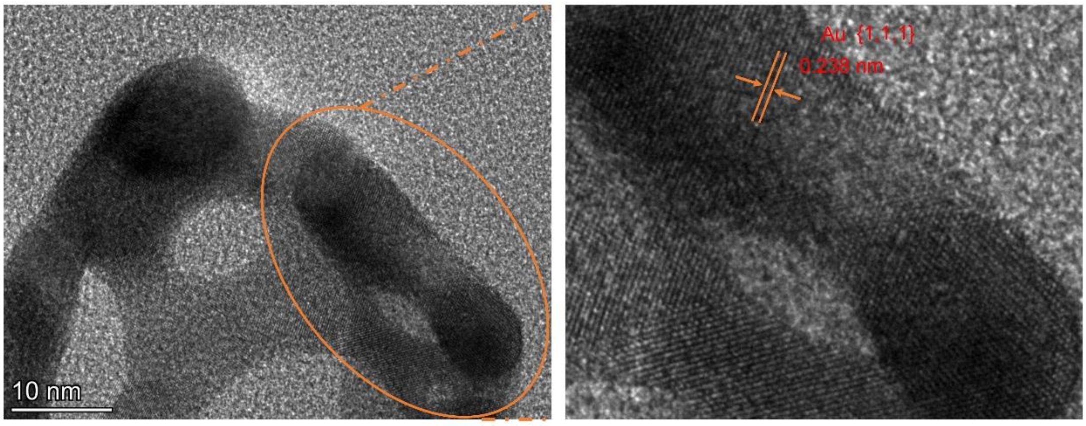

Figure S6. Lattice of Au in the cage structure.

Time (h)	Hydrodynamic size (nm)
0	145.37±4.47
24	146.23±3.62
48	148.14±5.76

Figure S7. Dynamic light scattering (DLS) analysis of FRNPs at different time point.

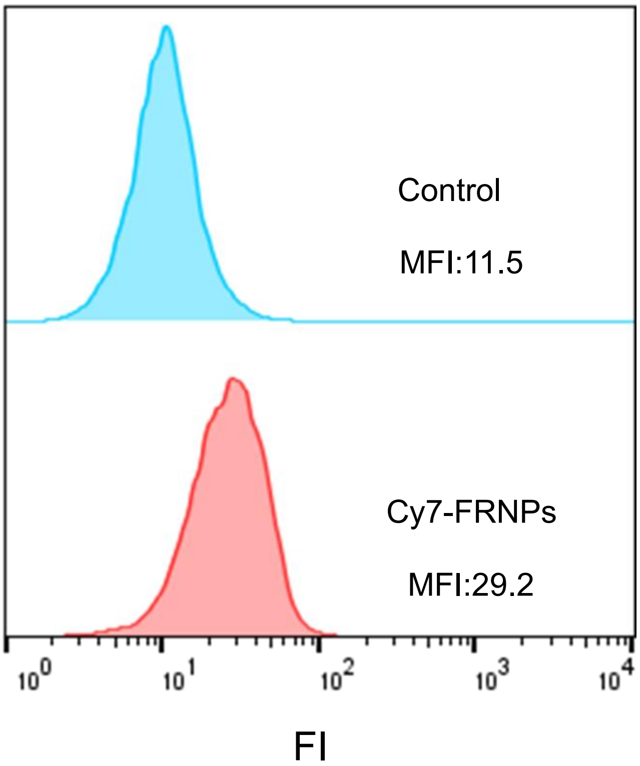

Figure S8. Flow cytometric analysis of the mean fluorescence intensity for ECA 109 cells incubated with PBS or Cy7-FRNPs (100 μg/ mL) for 24 h.

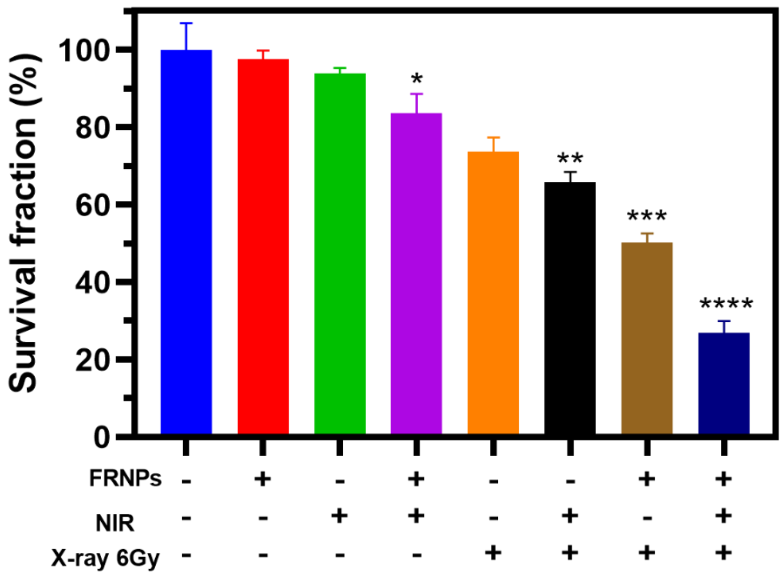

Figure S9. FRNPs in combined NIR photothermal and X-ray radiation exposure decreased ECA 109 cell survival rate detected by colony formation assay. Data represented were mean ± SD of three identical experiments made in three replicates. Error bars were based on standard deviation of three parallel samples. P-values were calculated by the student's test: $*p < 0.05$, $**p < 0.01$, $***p < 0.001$, $****p < 0.0001$ compared to the control group.

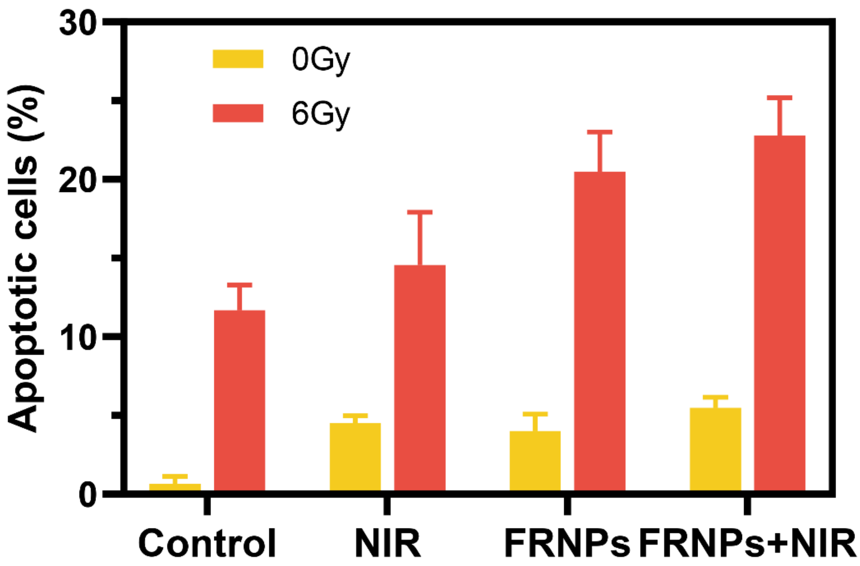

Figure S10. Quantitative analysis of cell apoptosis assay. Data are shown as mean ± SD.

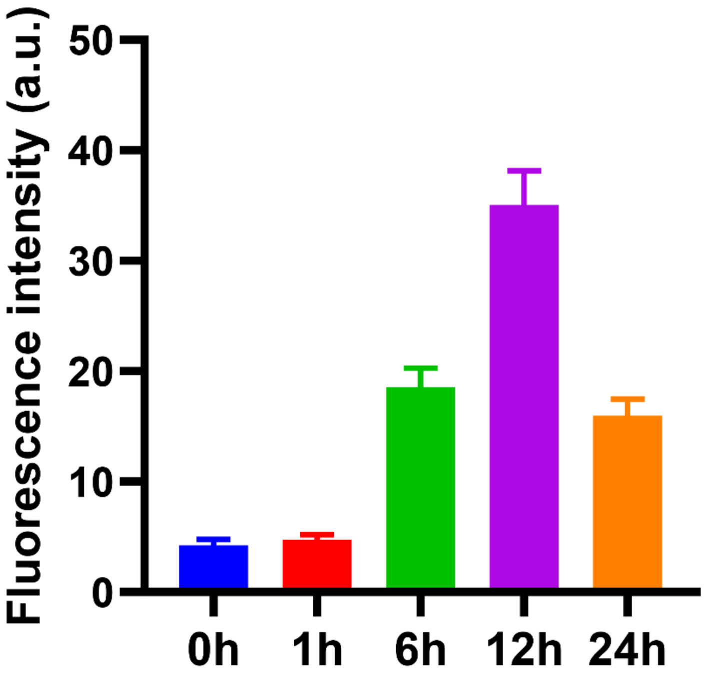

Figure S11. The fluorescence intensity of tumor area at different time intervals after injection of Cy7 -FRNPs.

Tabel S1. Summary of longitudinal relaxation rate of T1 contrast agent used in clinic or biomedicine.

T1 contrast agent	Commercial name	r_1 (mM^{-1} s^{-1})	Ref
Gd-DTPA	Magnevist	4.32	[1]
Gd-DTPA-BMEA	OptiMARK	4.10	[2]
Gd-DTPA-BMA	Omniscan	3.80	[3]
Gd-DOTA	Dotarem	3.50	[4]
Gd-DO3A-Butriol	Gadavist	3.70	[5]
Gd-DO3A-HP	ProHance	3.60	[6]
Gd-BOPTA	Multihance	5.20	[7]
Gd-diphenyl-cyclohexy-lphosphodiester-DTPA	Vasovist	4.56	[8]
Fe3O4@SiO2(Gd-DTPA)-RGD NPs		4.20	[9]
ES-MIONs		2.40	[10]
FRNPs		4.76	

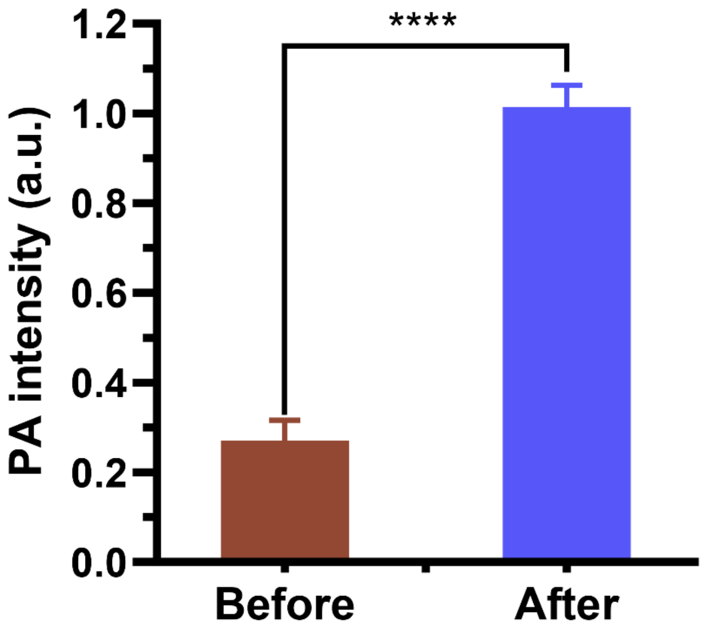

Figure S12. PA intensity at the tumor site before and after i.v. injection with FRNPs. P-values were calculated by the student's test: ****p < 0.0001.

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X-ray Structure of the 10-[2-[[2-Hydroxy-l-(hydroxymethyl)ethyl]amino]-l-[(phenylmethoxy)methyl]-2-oxoethyl]-l,4,7,10-tetraazacyclododecane-l,4,7-triacetic Acid-Gadolinium(III) Complex. Inorganic Chemistry. 1992;31(12):2422-2428.

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