

Electronic Supplementary Information

Table S1. T_1 relaxometry of $\text{NaGdF}_4\text{:Yb}^{3+}/\text{Er}^{3+}$ nanoparticles.

Concentration (mg/ml)	Relaxation time (ms)
5	63±1
2.5	170±5
1.25	302±5
0.625	503±1
0	2750±141

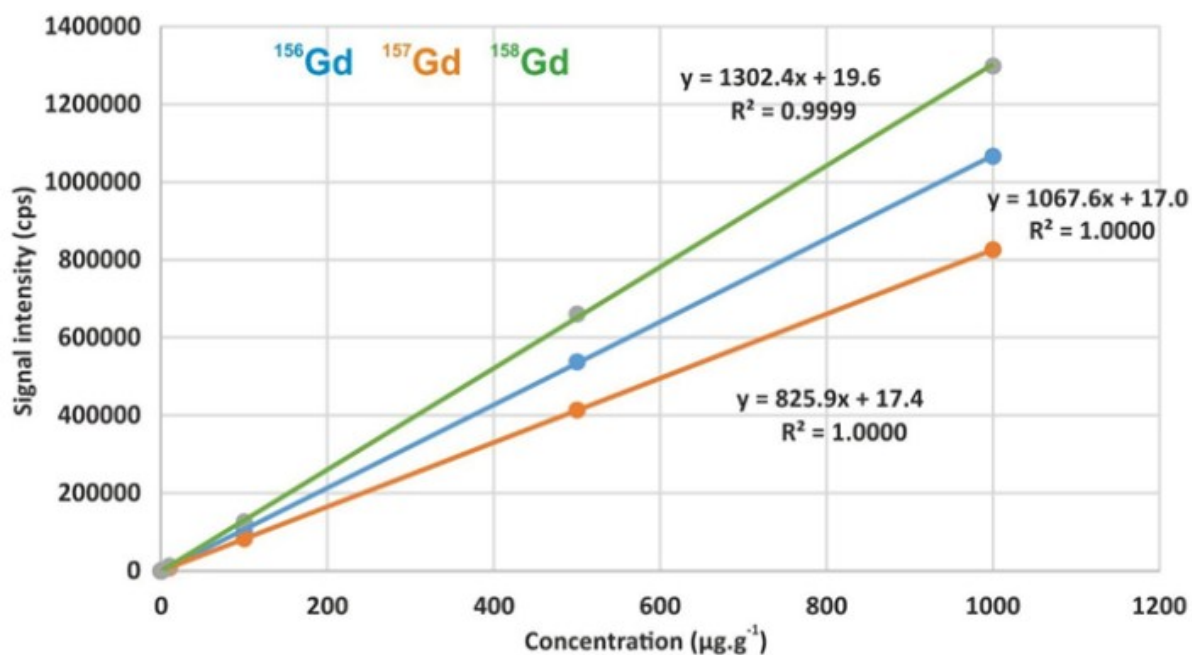


Fig. S1. Gadolinium calibration curves obtained using LA-ICP MS with a 110 μm laser focus.

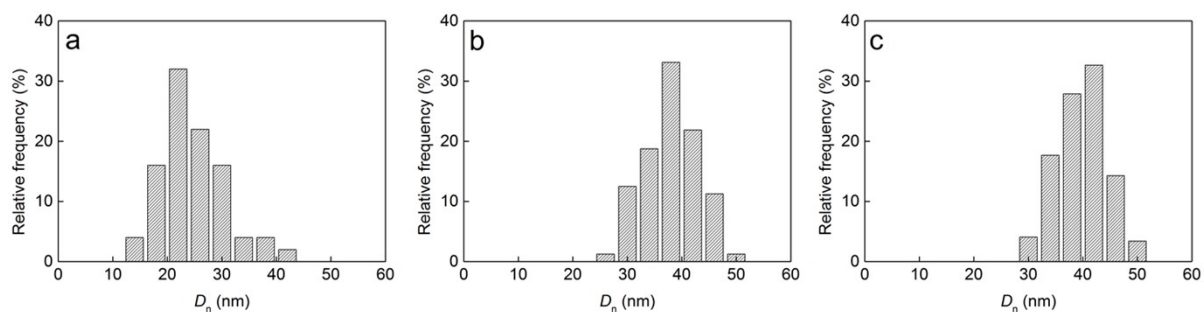


Fig. S2. Size distribution histograms of (a) starting $\text{NaGdF}_4\text{:Yb}^{3+}/\text{Er}^{3+}$ and (b, c) $\text{NaGdF}_4\text{:Yb}^{3+}/\text{Er}^{3+}@\text{SiO}_2$ nanoparticles; (b) 20 μl and (c) 40 μl TMOS in the feed; number-average diameter (D_n) of the individual particles was determined by image analysis of TEM micrographs.