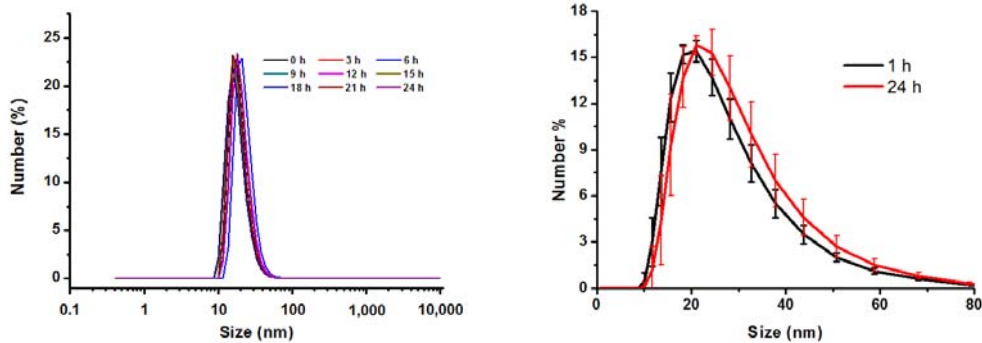


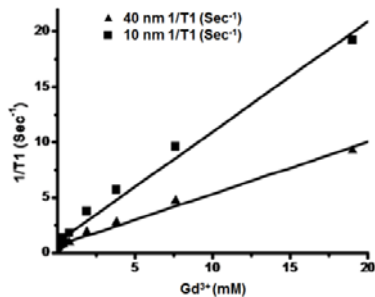
1. Stability test using DLS measurement



Supplemental Figure 1. Stability test using DLS measurement

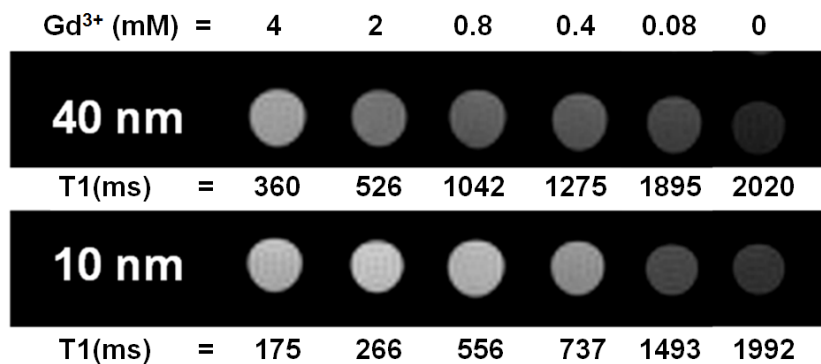
2. Magnetic properties of UCNPs

Measurement of T_1 relaxation time. Proton relaxation time was measured by the Carr-Purcell-Meiboom-Gill (CPMG) sequence at room temperature: TR 7 s, 100 echoes with 8.7 ms even echo space, number of acquisitions 1, point resolution of 417 - 469 μm and section thickness of 2 mm. The resulting T_1 (spin-lattice relaxation time) values were averaged and plotted as $1/T_1$ vs Gd^{3+} molar concentration. The slopes of the graph provided the molar relaxivity r_1 .



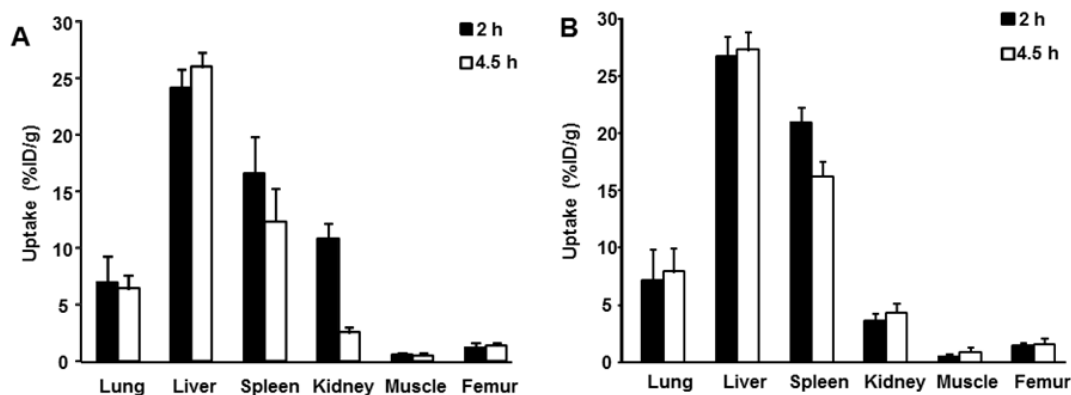
Supplemental Figure 2. Linearity of the concentration dependent contrast effect is obtained by plotting $1/T_1$ as a function of $[\text{Gd}^{3+}]$. The specific relaxivities (r_1) for 10 and 40 nm UCNPs were determined to be 0.99 and 0.47 $\text{mM}^{-1}\text{s}^{-1}$, respectively.

To further evaluate the magnetic properties of $\text{NaGdF}_4: \text{Yb}^{3+}, \text{Er}^{3+}$, the aqueous solution of UCNPs at various concentrations was measured for their T_1 relaxation time. MR images in Supplemental Figure 2 show the Gd^{3+} concentration dependent contrast effect of the UCNPs. UCNPs with smaller size of 10 nm exhibited higher T_1 contrast enhancing effect. Linearity of the concentration dependent contrast effect is obtained by plotting $1/T_1$ as a function of $[\text{Gd}^{3+}]$. The specific relaxivities (r_1) for 10 and 40 nm UCNPs were determined to be 0.99 and 0.47 $\text{mM}^{-1}\text{s}^{-1}$, respectively. The result showing the size dependent relaxivity was also reported in the literature (Park, Y. I. et al. Adv. Mater. 2009, 21, 4467).

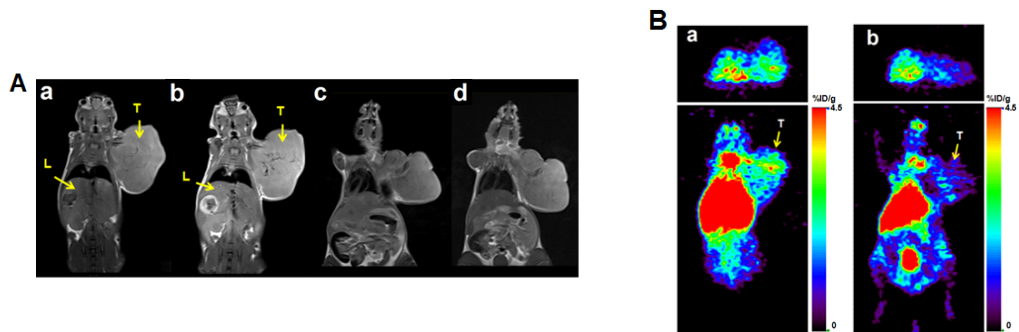


Supplemental Figure 3. T₁ weighted magnetic resonance images of UCNPs at various [Gd³⁺] concentrations in water.

3. PET image-based biodistribution



Supplemental Figure 4. PET image-based *in vivo* biodistribution of ¹²⁴I-(cRGDyk)₂-UCNPs (A) and ¹²⁴I-(cRGDyk)₂-UCNPs (B) with blocking dose of (cRGDyk)₂ peptides in U87MG tumor bearing mice. The PET images were analyzed with Inveon Research workplace (IRW) software for organ radioactivity quantification. The radioactivity (%ID/g) in various tissues at 2 h and 4.5 h post-injection of radiotracer was measured by image region-of-interest (ROI) analysis of microPET datasets.



Supplemental Figure 5. (A) T1-weighted MR coronal images in U87MG tumor bearing mice; (a) before injection of ^{124}I -(cRGDyk) $_2$ -UCNPs and (B) Small animal PET transverse (upper) and coronal (lower) images of U87MG tumor bearing nude mice.