SUPPLEMENTARY MATERIAL

Phase and Size Controllable Synthesis of $NaYbF_4$ Nanocrystals in Oleic Acid/ Ionic Liquid Two-Phase System for Targeted Fluorescent Imaging of Gastric Cancer

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1. Materials

Rare earth oxides Yb_2O_3 (99.99%), Gd_2O_3 (99.99%), Tm_2O_3 (99.99%) were purchased from Aladdin Chemistry Co.Ltd. Sodium oleate, oleic acid (OA), ethanol, ammonia and tetraethylorthosilicate (TEOS) were purchased from Sinopharm Chemical Reagent Co.1–butyl-3-methylimidazolium tetrafluoroborate ($BmimBF_4$) and Tetrabutylammonium hexafluorophosphate ($BmimBF_6$) was purchased from Shanghai Cheng Jie Chemical Co. Ltd. APS (98%), Ethylcarbodiimide (EDC) and N-hydroxysuccinimide (NHS) were purchased from Aladdin Chemistry Co. Ltd. Cell culture products and reagents were purchased from GIBCO. All the chemical regents are of analytical grade and were used directly without further purification. Deionized water was used throughout.

2. Methods

X-ray powder diffraction (XRD) analysis

X-ray powder diffraction (XRD) measurements were performed on a Dmax-r C X-ray diffractometer (Rigaku, Japan) at a scanning rate of 6ºmin-1 in the 2θrange of 10-70°, with Cu Kα radiation at 1.540Aº. The sizes and morphologies of the nanocrystals were characterized at 200 kV with a JEM 2010 microscopy (JEOL, Japan) and High-resolution TEM (HR-TEM) on JEM 2100F microscopy (JEOL, Japan).Energy-dispersive X-ray analysis (EDXA) of products was also performed during high-resolution TEM measurements. Fourier transform infrared (FT-IR) spectra were measured by using an EQUINOX 55 spectroscope (Bruker, Germany) with KBr pellets. Upconversion fluorescence spectra were recorded on Fluorolog-3 spectrofluorometer (Jobin Yvon, France) at room temperature. UV-vis spectra were recorded by Varian Cary50 and UV-vis spectrophoto metre equiped with a 10 nm quartz cell. Zeta potentials were measured by using NICOMP 380ZLS zeta potential/particle size.

3. Characterization

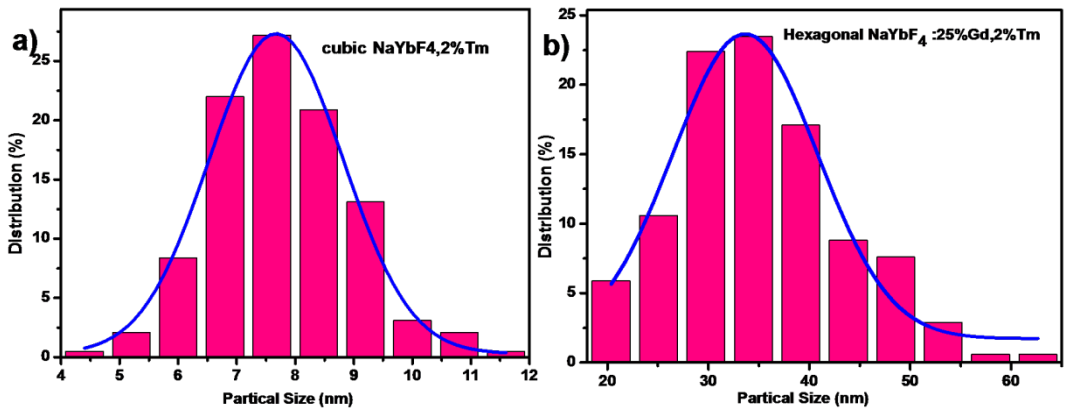

Figure S1 a) Size distribution of cubic phase $NaYbF_4$, 2%Tm nanocrystals b) Size distribution of water-soluble hexagonal phase $NaYbF_4$:25%Gd, 2%Tm nanocrystals

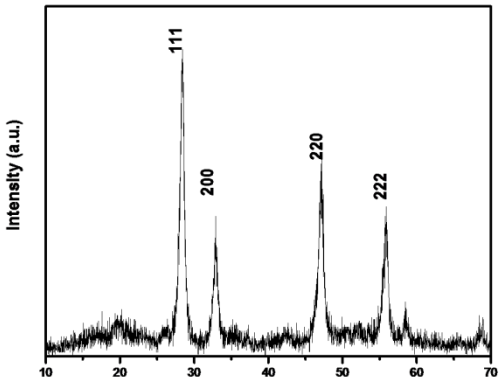

Figure S2 XRD patterns of cubic phase $NaYbF_4$, 2%Tm nanocrystals

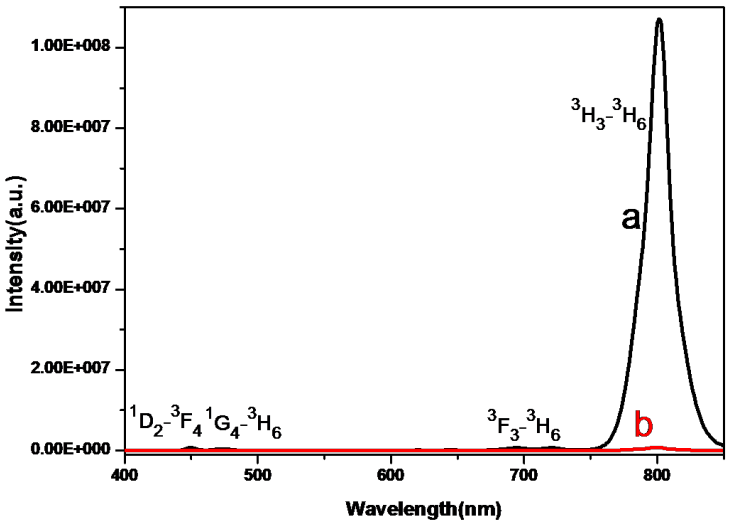

Figure S3. Room-temperature upconversion luminescence spectra of water-soluble hexagonal phase (a) $NaYbF_4$:25%Gd, 2%Tm nanocrystals with excess sodium oleate,(b) $NaYbF_4$: 25%Gd, 2%Tm nanocrystals

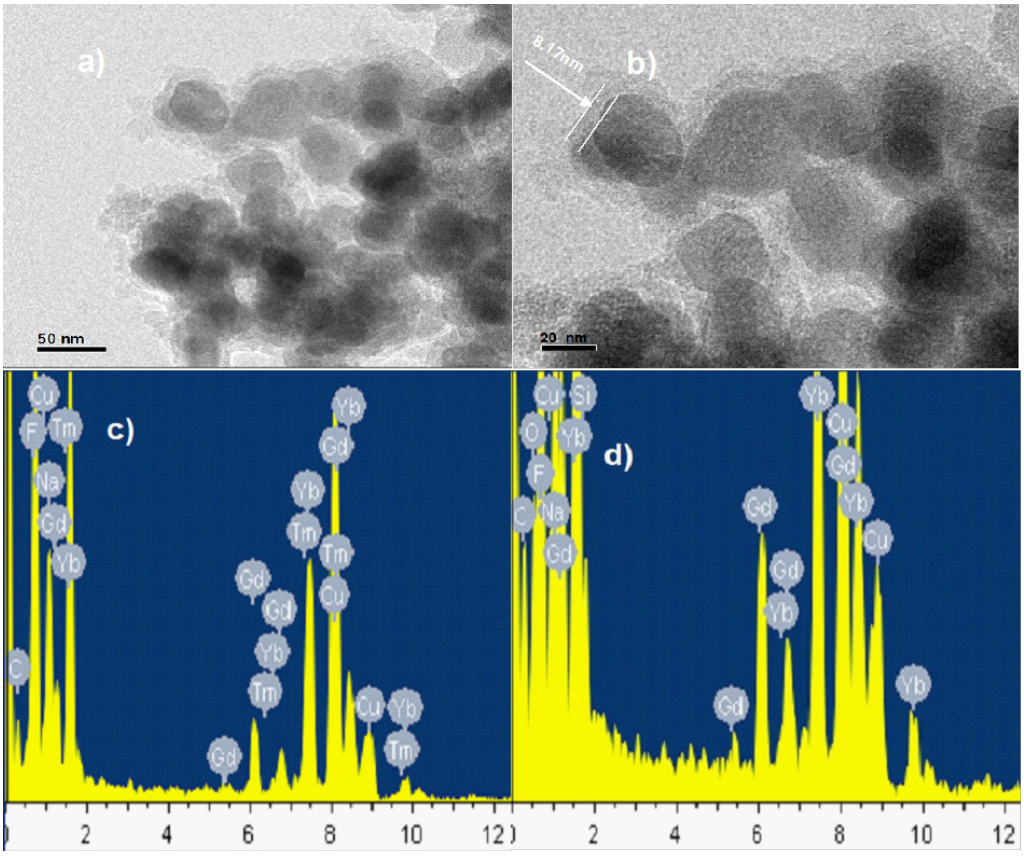

Figure S4 a) and b) TEM images with different magnifications of silica coating water-soluble hexagonal phase $NaYbF_4$:25% Gd, 2%Tm nanocrystals. c) and d) EDX spectrum for $NaYbF_4$: 25%Gd, 2%Tm and $NaYbF_4$: 25%Gd, 2%Tm@SiO_2, respectively.

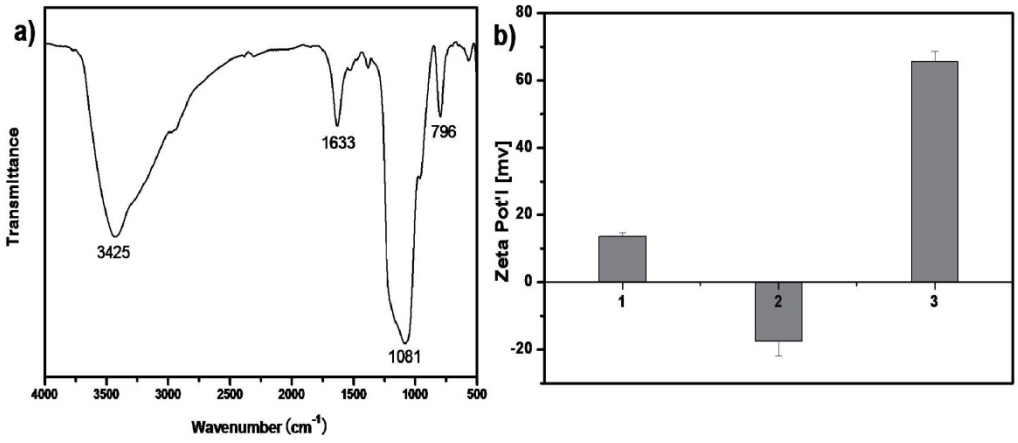

Figure S5. a) FT-IR spectrum of $NaYbF_4$: Gd, Tm UCNPs after silica coating. b) The bars of 1, 2, 3 stand for zeta potentials of $NaYbF_4$: 25%Gd, 2%Tm, $NaYbF_4$: 25%Gd, 2%Tm,@SiO_2

and $NaYbF_4$: 25%Gd, 2%Tm,@SiO_2-NH_2.

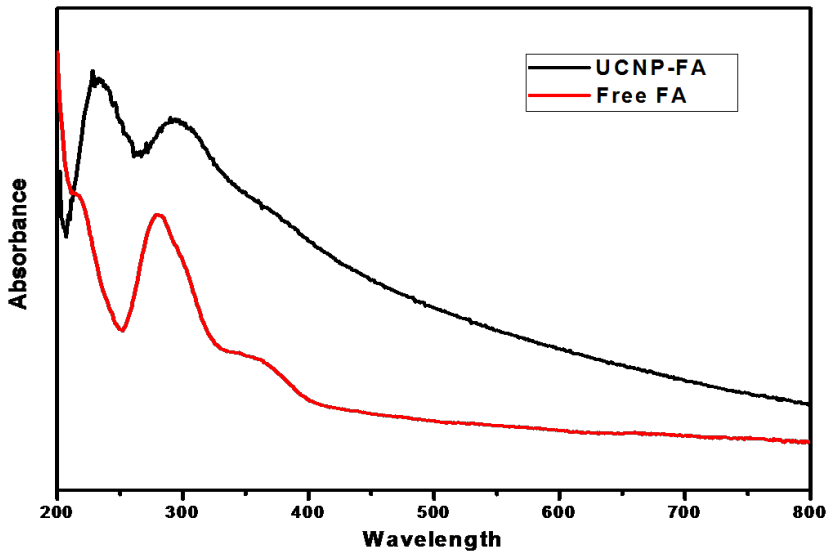

Figure S6. UV-vis absorbance spectra of UCNP-FA and free FA. The UCNP concentration was 1 mg/ml in this sample. The FA characteristic peaks in the UCNP-FA sample clearly evidenced the existence of FA conjugation on amine-functionalized UCNP@SiO_2-NH_2 core-shell nanoparticles.

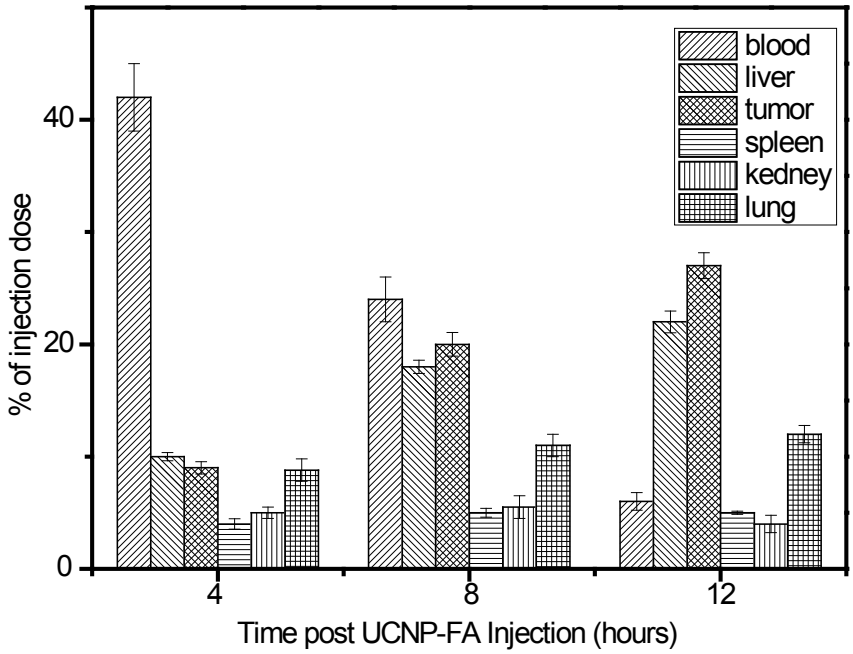

Figure S7. Biodistribution of targeted FA-UCNPs in mice after intravenous injection. Several time points after injection, UCNP amounts in tissue samples were evaluated by ICP mass spectrometry. Data represent mean values for n=3 and bars are standard deviations for means.