

Supplementary Material

Water-soluble Cit-NaYbF₄:Tm³⁺ with enhanced 802 nm emission by Sr²⁺ ions doping for *in vivo* fluorescence molecular tomography

Xiaoli Luo^{a,b}, Qichen Chen^a, Hongbo Guo^a, Chen Cheng^a, Qianqian Ren^a, Heng Zhang^a, Xiaowei He^{a,*}, Wu Zhao^{a,*}

^aSchool of Information Science and Technology, Northwest University, Xi'an, Shaanxi 710127, China.

^bSchool of Mathematics and Computer Science, Ningxia Normal University, Guyuan, Ningxia 756099, China.

* Corresponding author: Dr. Wu Zhao, Tel: +86 29 88308280. E-mail: zhaowu@nwu.edu.cn

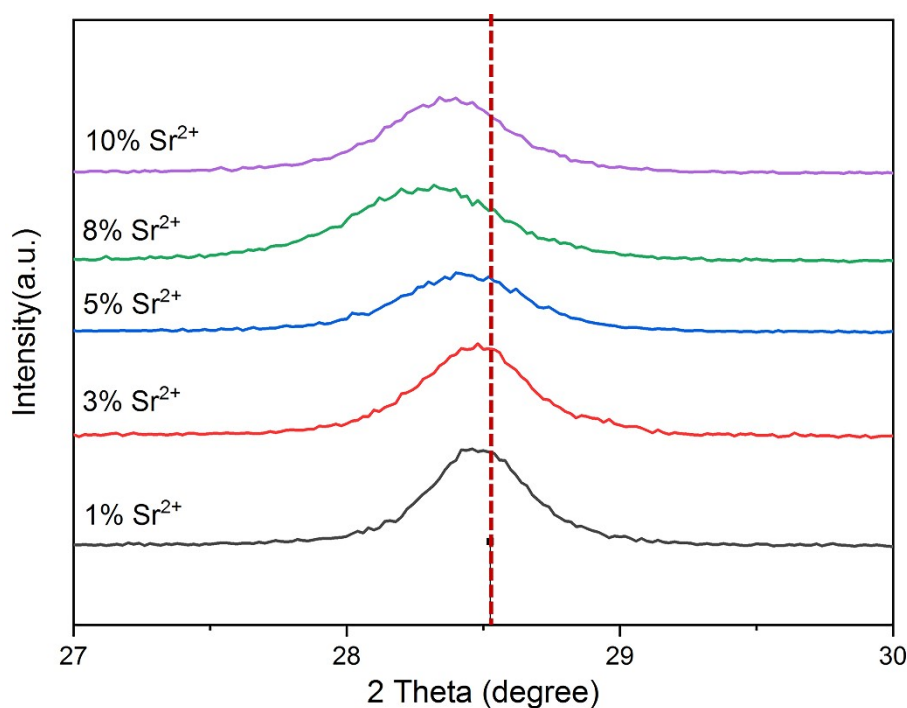


Figure S1: The magnified XRD patterns of NaYbF₄:2%Tm³⁺/x%Sr²⁺ (x = 1, 3, 5, 8, and 10).

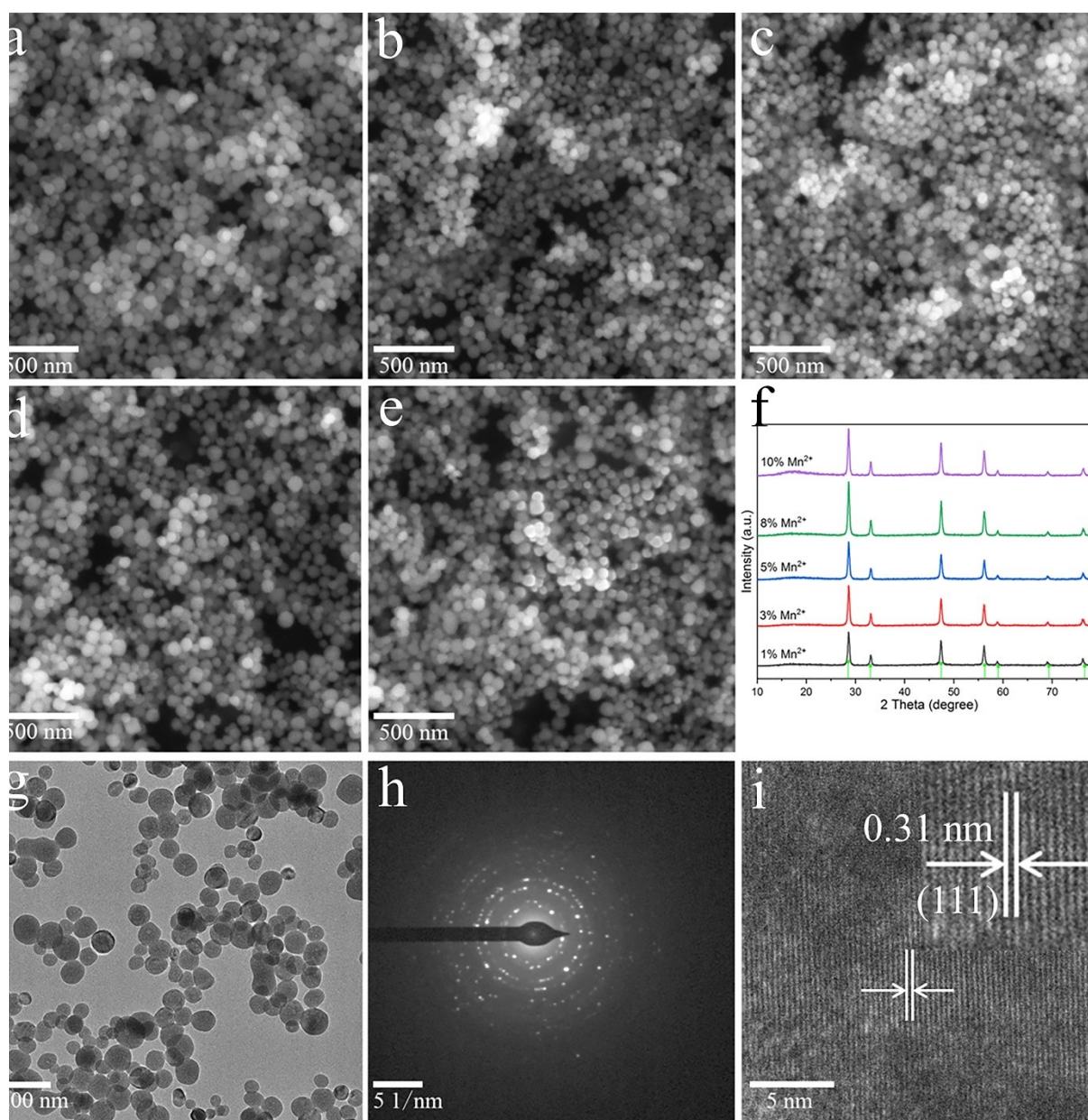


Figure S2: Analysis of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/x\%\text{Mn}^{2+}$ crystals. SEM images of $\text{NaYb}_{0.98-x}\text{F}_4:2\%\text{Tm}^{3+}/x\%\text{Mn}^{2+}$ crystals synthesized with (a) $x = 1$; (b) $x = 3$; (c) $x = 5$; (d) $x = 8$; (e) $x = 10$; (f) XRD patterns of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/x\%\text{Mn}^{2+}$ ($x = 1, 3, 5, 8,$ and 10). (g) the TEM image of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/5\%\text{Mn}^{2+}$; (h) the SAED pattern of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/5\%\text{Mn}^{2+}$; (i) the corresponding HRTEM image of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/5\%\text{Mn}^{2+}$.

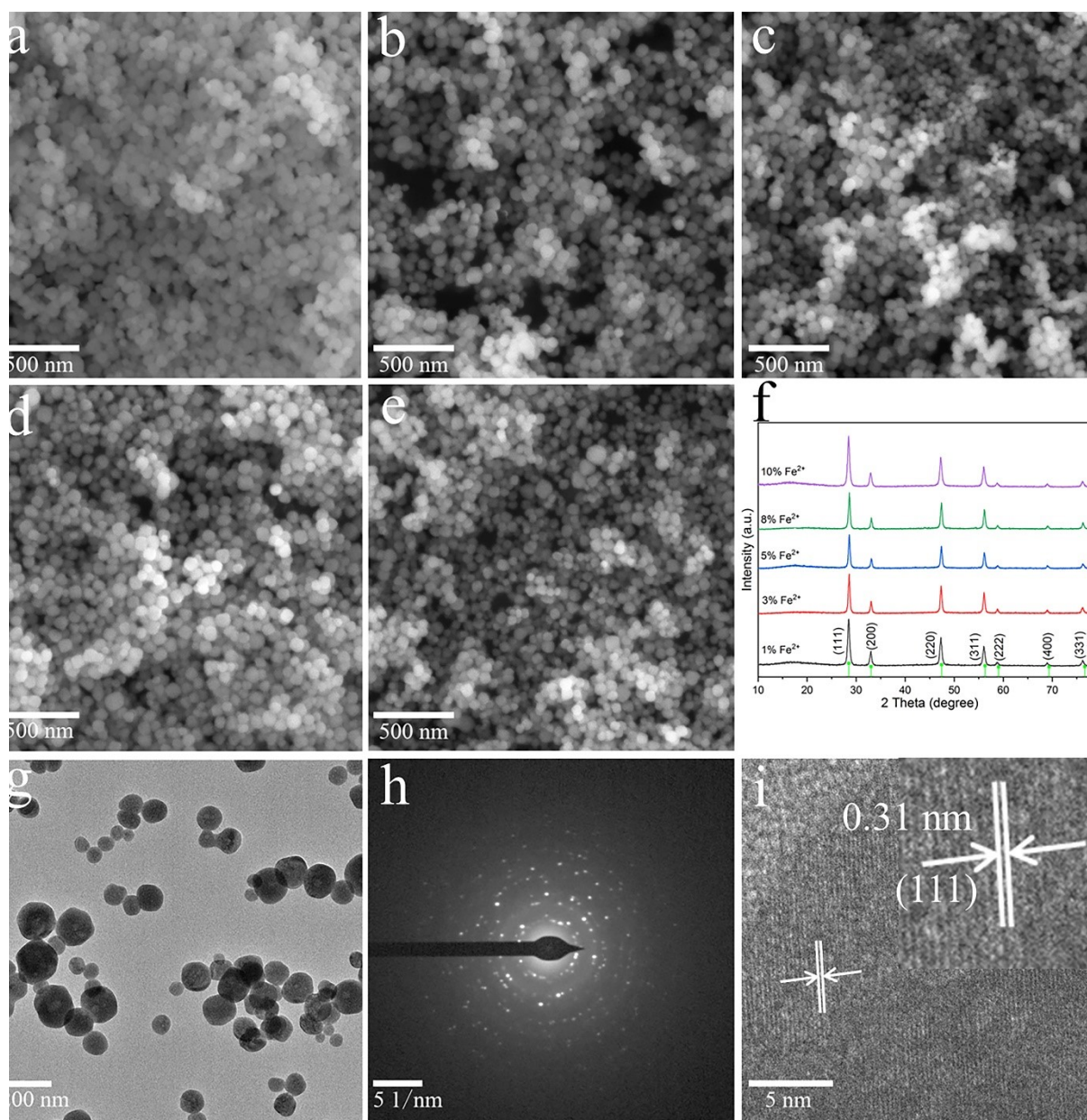


Figure S3: Analysis of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/x\%\text{Fe}^{2+}$ crystals. SEM images of $\text{NaYb}_{0.98-x}\text{F}_4:2\%\text{Tm}^{3+}/x\%\text{Fe}^{2+}$ crystals synthesized with (a) $x = 1$; (b) $x = 3$; (c) $x = 5$; (d) $x = 8$; (e) $x = 10$; (f) XRD patterns of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/x\%\text{Fe}^{2+}$ ($x = 1, 3, 5, 8,$ and 10). (g) the TEM image of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/5\%\text{Fe}^{2+}$; (h) the SAED pattern of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/5\%\text{Fe}^{2+}$; (i) the corresponding HRTEM image of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/5\%\text{Fe}^{2+}$.

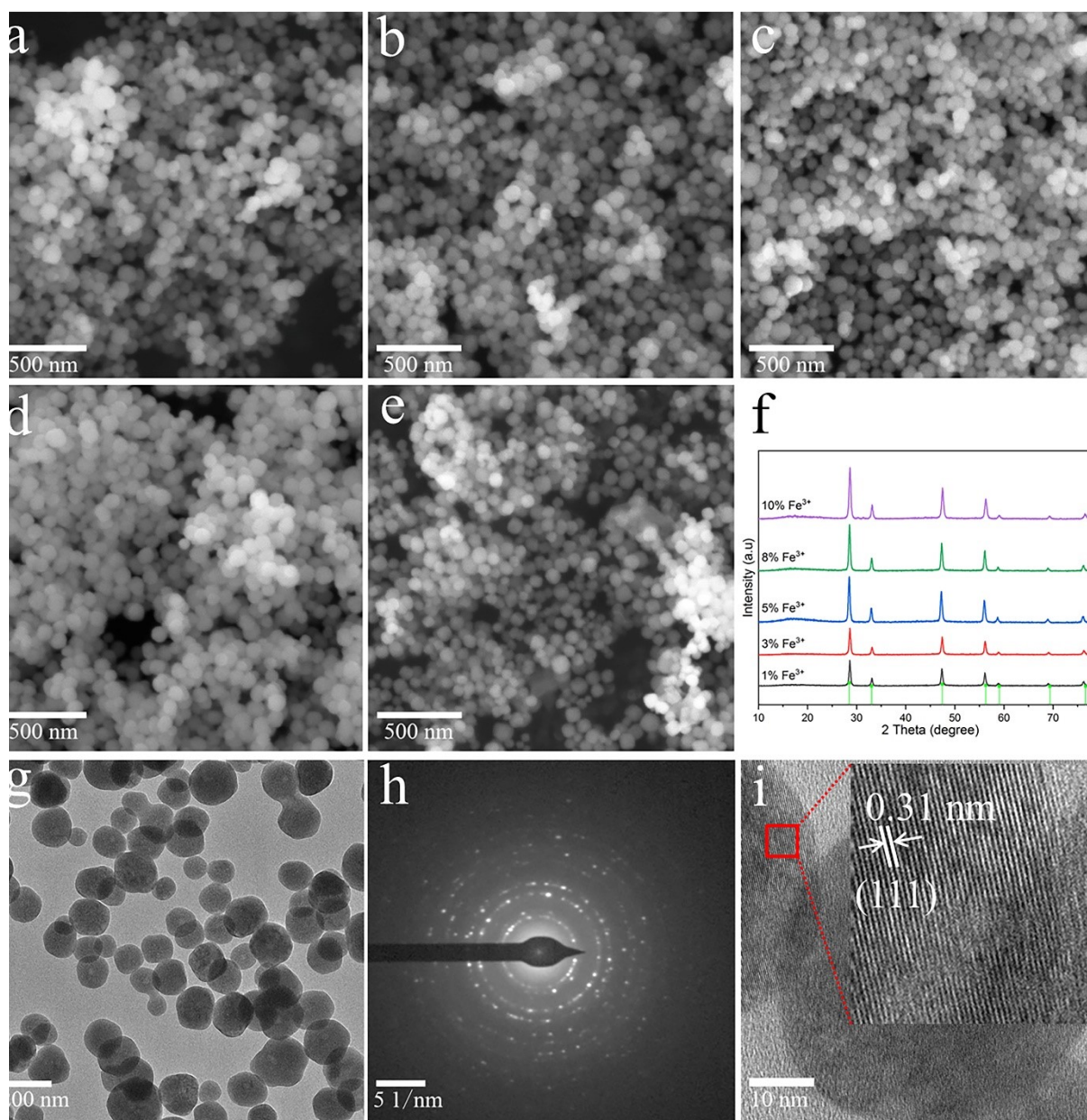


Figure S4: Analysis of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/x\%\text{Fe}^{3+}$ crystals. SEM images of $\text{NaYb}_{0.98-x}\text{F}_4:2\%\text{Tm}^{3+}/x\%\text{Fe}^{3+}$ crystals synthesized with (a) $x = 1$; (b) $x = 3$; (c) $x = 5$; (d) $x = 8$; (e) $x = 10$; (f) XRD patterns of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/x\%\text{Fe}^{3+}$ ($x = 1, 3, 5, 8,$ and 10). (g) the TEM image of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/5\%\text{Fe}^{3+}$; (h) the SAED pattern of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/5\%\text{Fe}^{3+}$; (i) the corresponding HRTEM image of $\text{NaYbF}_4:2\%\text{Tm}^{3+}/5\%\text{Fe}^{3+}$.

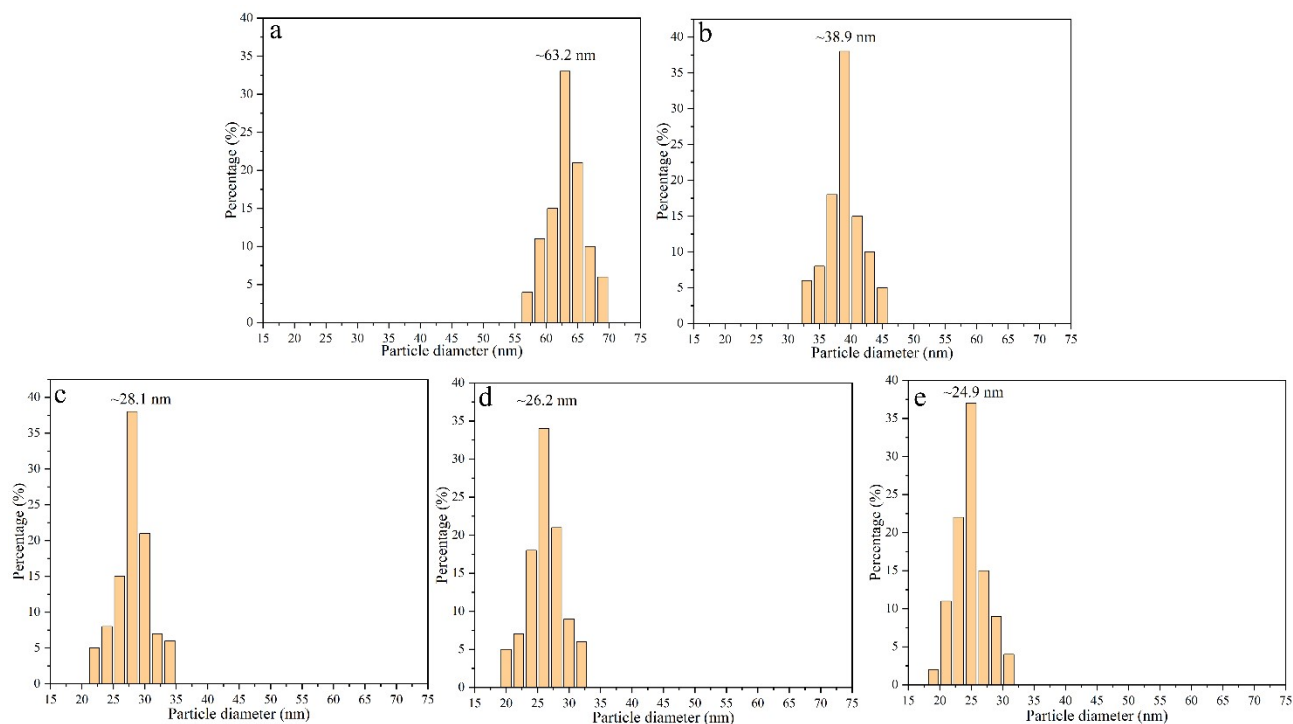


Fig. S5: Size histograms of NaYb_{0.98-x}F₄:2%Tm³⁺/x%Sr²⁺ crystals synthesized with (a) x = 1; (b) x = 3; (c) x = 5; (d) x = 8; (e) x = 10.

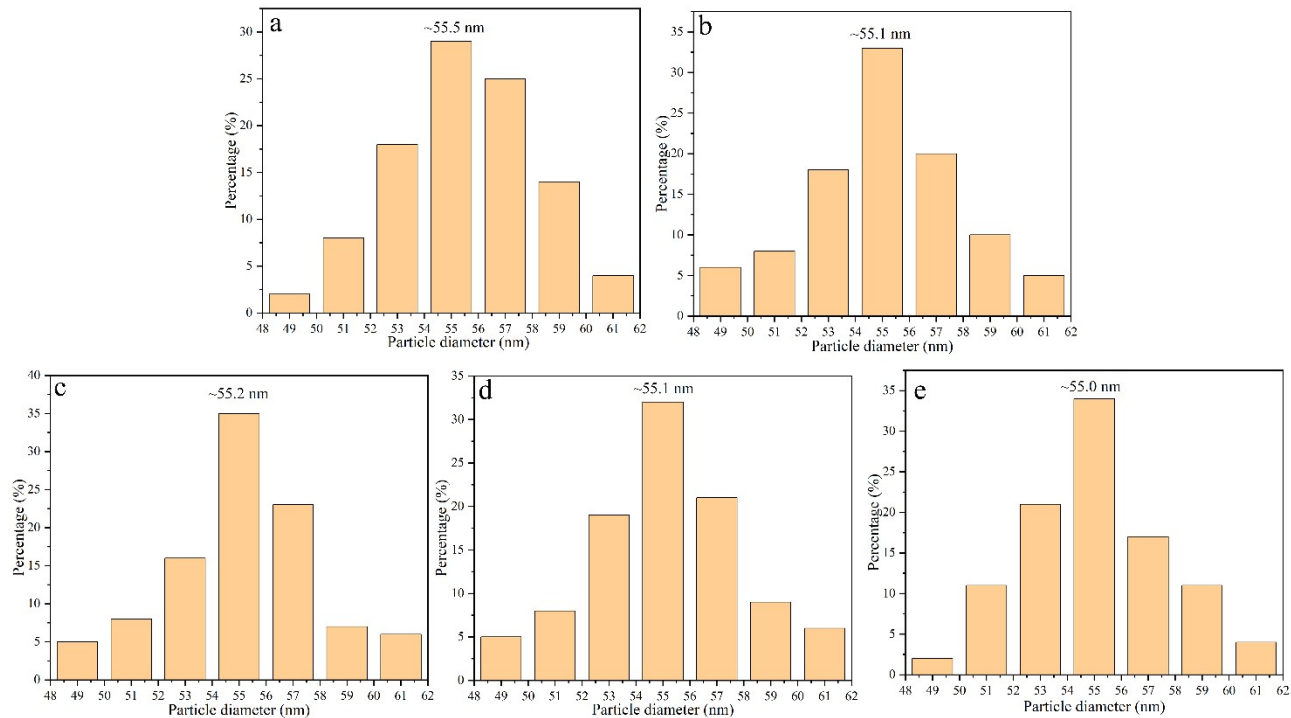


Fig. S6: Size histograms of NaYb_{0.98-x}F₄:2%Tm³⁺/x%Mn²⁺ crystals synthesized with (a) x = 1; (b) x = 3; (c) x = 5; (d) x = 8; (e) x = 10.

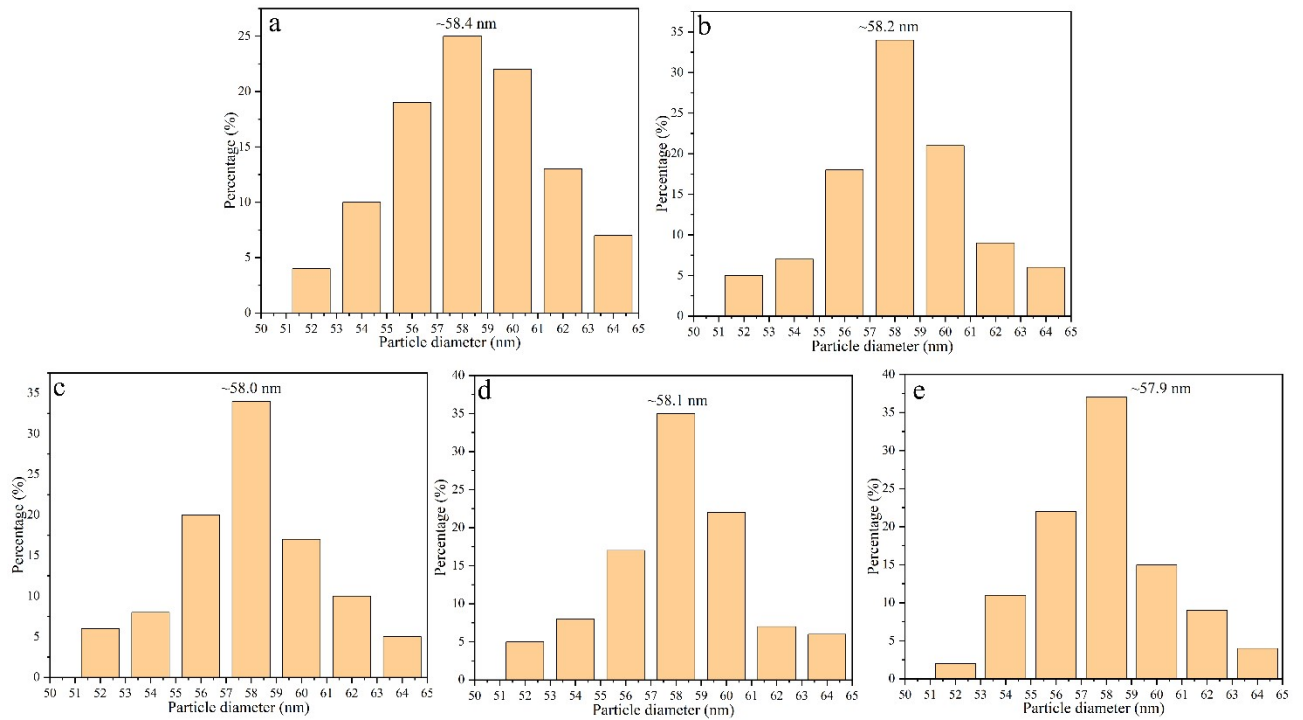


Fig. S7: Size histograms of NaYb_{0.98-x}F₄:2%Tm³⁺/x%Fe²⁺ crystals synthesized with (a) $x = 1$; (b) $x = 3$; (c) $x = 5$; (d) $x = 8$; (e) $x = 10$.

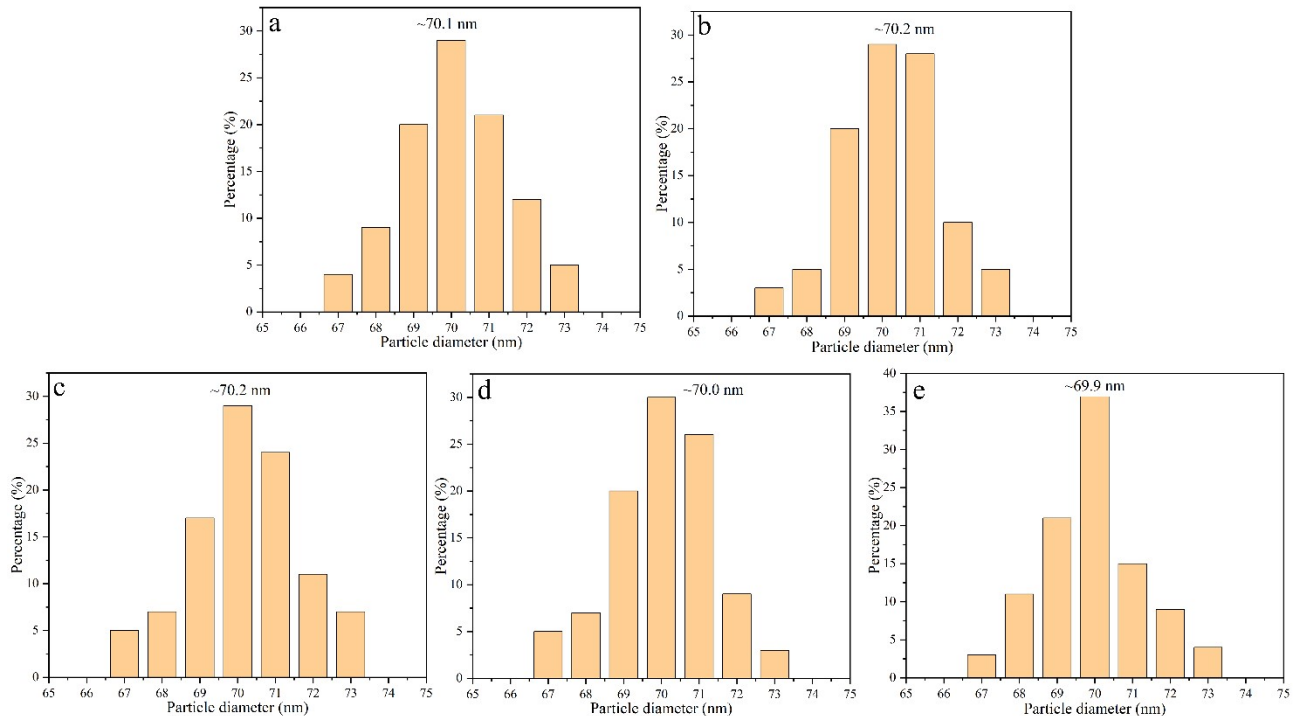


Fig. S8: Size histograms of NaYb_{0.98-x}F₄:2%Tm³⁺/x%Fe³⁺ crystals synthesized with (a) $x = 1$; (b) $x = 3$; (c) $x = 5$; (d) $x = 8$; (e) $x = 10$.

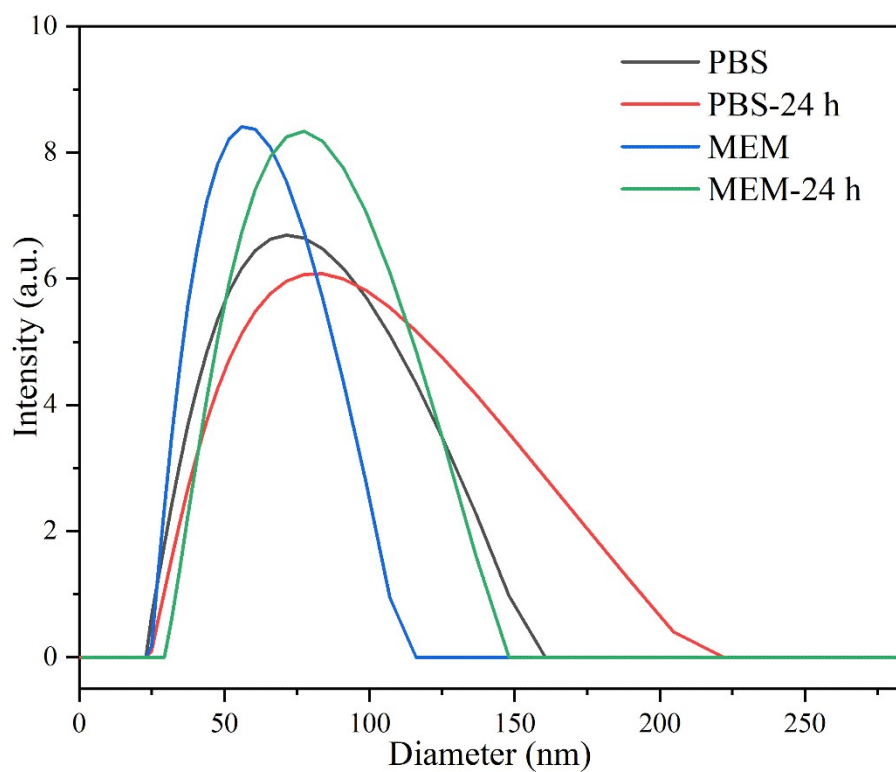


Fig. S9 DLS of prepared Cit-NaYbF₄:2%Tm³⁺/5%Sr²⁺.

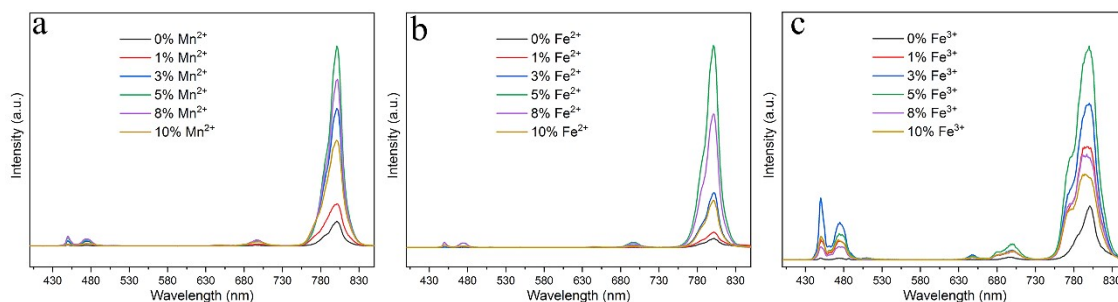


Figure S10: The UC luminescence spectra of the samples generated with different ions doping at different doping molar ratios. (a) NaYbF₄:2%Tm³⁺/x%Mn²⁺ (x = 0, 1, 3, 5, 8, and 10), (b) NaYbF₄:2%Tm³⁺/x%Fe²⁺ (x = 0, 1, 3, 5, 8, and 10), (c) NaYbF₄:2%Tm³⁺/x%Fe³⁺ (x = 0, 1, 3, 5, 8, and 10).

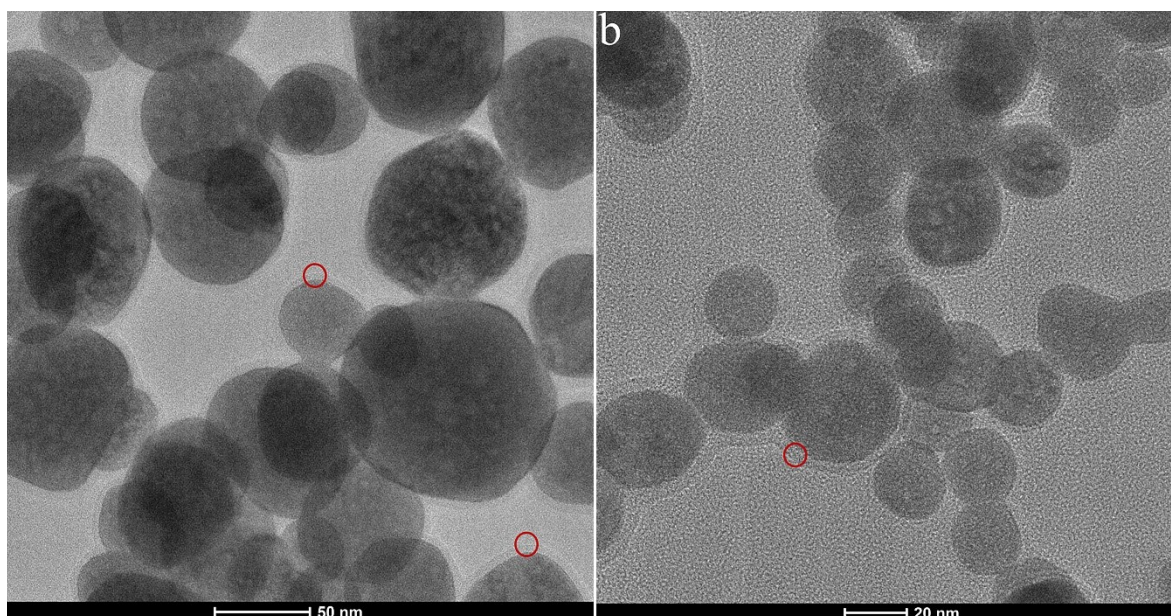


Figure S11: Higher-magnification TEM image of (a) $\text{NaYbF}_4:2\%\text{Tm}^{3+}$; (b) $\text{NaYbF}_4:2\%\text{Tm}^{3+}/5\%\text{Sr}^{2+}$. The transparent areas circled in red represent the surface of these UCNPs coated with water-soluble organic ligands



Figure S12: Aqueous solution of UCNPs at a concentration of 400 $\mu\text{g}/\text{mL}$ at room temperature. (left) $\text{NaYbF}_4:2\%\text{Tm}^{3+}/5\%\text{Sr}^{2+}$; (right) $\text{NaYbF}_4:2\%\text{Tm}^{3+}$.

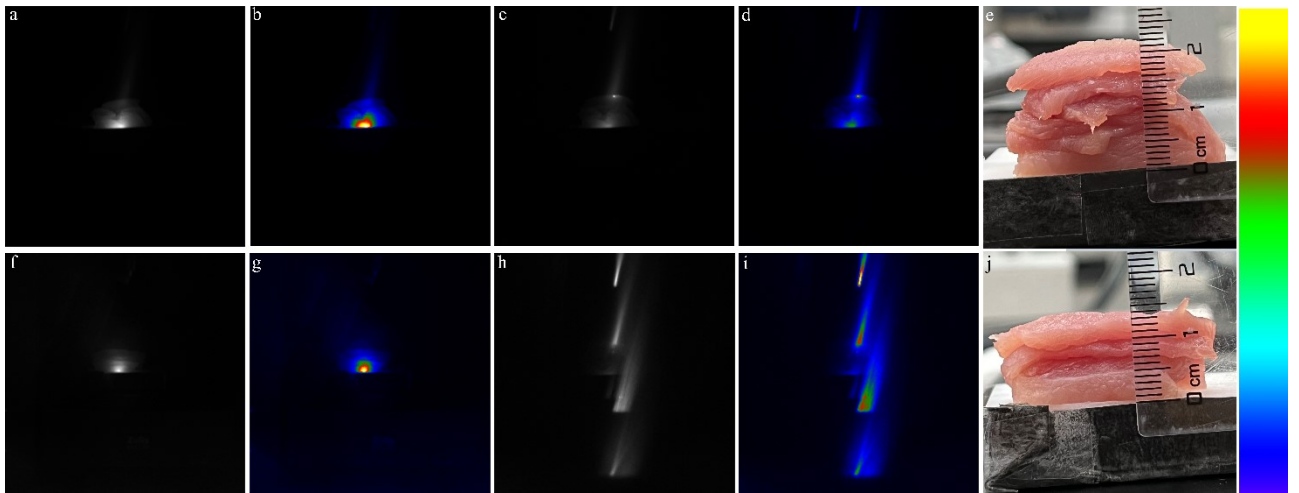


Fig. S13 Fluorescence signal detection of pork muscle tissues treated with different fluorescence probes in different depths. **(a-e)** 2 mg/mL Cit-NaYbF₄:2%Tm³⁺/5%Sr²⁺; **(f-j)** 0.1 mg/mL Indocyanine Green (ICG). **(a)** 20 mm; **(b)** 20 mm with energy; **(c)** 21 mm; **(d)** 21 mm with energy; **(e)** 20 mm of pork muscle tissues treated with 2 mg/mL Cit-NaYbF₄:2%Tm³⁺/5%Sr²⁺; **(f)** 13 mm; **(g)** 13 mm with energy; **(h)** 14 mm; **(i)** 14 mm with energy; **(j)** 13 mm of pork muscle tissues treated with 0.1 mg/mL ICG.

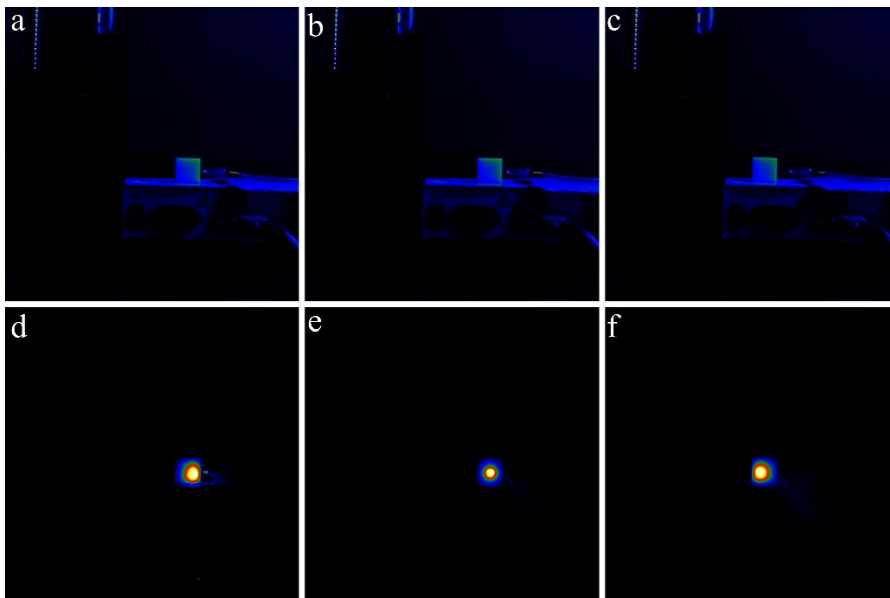


Figure S14: Fluorescence molecular tomography (FMT) imaging in phantom at different faces. **(a-c)** Bright field images. **(d-e)** Upconversion luminescence images.

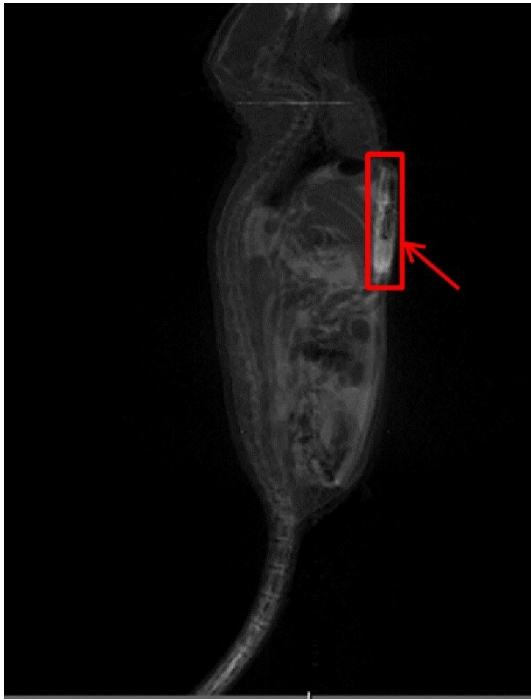


Figure S15: CT image.