## Fe<sup>3+</sup>-codoped Ultra-small NaGdF<sub>4</sub>:Nd<sup>3+</sup> Nanophosphors:Enhanced Nearinfrared Luminescence, Reduced Particle Size and Bioimaging Applications

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Figure S1 EDS spectrum of the NaGdF<sub>4</sub>:Nd<sup>3+</sup>,Fe<sup>3+</sup> NPs (20 mol% of Fe<sup>3+</sup>).



Figure S2 Change of particle size of NaGdF<sub>4</sub>:Nd<sup>3+</sup>,Fe<sup>3+</sup> NPs with varying Fe<sup>3+</sup> concentration.



**Figure S3** Comparison of NIR luminescence intensity of the NaGdF<sub>4</sub>:Nd<sup>3+</sup> NPs before and after being doped by 20 mol% of Fe<sup>3+</sup>.



**Figure S4** Zeta potential distributions of PEG-NPs (green line) and PEG-NPs-FA (red line) in aqueous solution.



**Figure S5** TEM images of PEG-NPs stored in PBS buffer solution for different time, (a) 1 day, (b) four days, (c) seven days.



Figure S6 DLS of PEG-NPs in PBS buffer solution at different time points.



**Figure S7** The photographs of PBS buffer solution of PEG-NPs at different storing time: (a) 1 day, (b) four days, and (c) seven days.



**Figure S8** The cytotoxicity of the NPs assessed by cell viability based on the MTT assay: (a) HeLa cell viabilities upon incubation with various concentrations of PEG-NPs-FA (0, 20, 50, 100, 200, 300, 400, 500, and 600  $\mu$ g mL<sup>-1</sup>, respectively) for 24 h, (b) HeLa cell viabilities after incubating for 0, 12, 24, 36 and 48 h with 300  $\mu$ g mL<sup>-1</sup> of PEG-NPs-FA.



Figure S9 The molecular structure of DSPE-PEG-NH2.



Figure S10 The TGA curves of OA-NPs, PEG-NPs and PEG-NPs-FA.



**Figure S11** TEM images of  $NaYF_4$ :Nd (a) before and (b) after targeted modification, and (c) DLS of  $NaYF_4$ :Nd after FA modification.



Figure S12 Near-infrared living imaging using the targeting PEG- NPs-FA.



**Figure S13** Near-infrared living imaging using the non-targeting PEG-NPs: (a) intraperitoneal injection and (b) tail vein injection. The red circle in the figure marks the location of the tumor



**Figure S14** (a) T2-weighted MR image of NaGdF<sub>4</sub>:Nd<sup>3+</sup> NPs without doping of Fe<sup>3+</sup>. (b) plots of 1/T1 (black) and 1/T2 (red) as a function of the Gd<sup>3+</sup> concentration for NaGdF<sub>4</sub>:Nd<sup>3+</sup> NPs.