Electronic Supplementary Information

Active-core/active-shell nanostructured design: An effective strategy to enhance Nd³⁺-Yb³⁺ cascade sensitized upconversion luminescence in lanthanide-doped

nanoparticles

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Figure S1. (a,b) TEM image and the corresponding size distribution of $NaYF_4:Nd/Yb/Er(1/29/0.5\%)$ nanopaticles. (c,d) TEM image and the corresponding size distribution of $NaYF_4:Nd/Yb/Er(1/39/0.5\%)$ nanopaticles.



Figure S2. (a,b) TEM image and the corresponding size distribution of NaYF₄:Nd/Yb/Er(1/19/0.5%)@NaYF₄:Nd/Yb(20/20%) core-shell nanopaticles. (c,d) TEM image and the corresponding size distribution of NaYF₄:Nd/Yb/Er(1/19/0.5%)@NaYF₄:Nd/Yb(20/40%) core-shell nanopaticles.



Figure S3. (a,b) TEM image and the corresponding size distribution of NaYF₄:Nd/Yb/Tm(1/19/0.5%) nanopaticles. (c,d) TEM image and the corresponding size distribution of NaYF₄:Nd/Yb/Tm(1/19/0.5%)@NaYF₄:Nd/Yb(20/10%) core-shell nanopaticles. The thickness of the active-shell is about 3.31 nm.



Figure S4. Upconversion emission spectra of the NaYF₄:Nd/Yb/Er(1/x/0.5%) core-only nanoparticles as a function of Yb³⁺ doping concentration (x = 19, 29, and 39 mol%) under 980 nm excitation.



Figure S5. Upconversion emission spectra for NaYF₄:Nd/Yb/Er(1/19/0.5%) nanoparticles and the corresponding core/shell nanoparticles coated with different active shells: NaYF₄:Nd(20%) shell, NaYF₄:Nd/Yb(20/10%) shell, NaYF₄:Nd/Yb(20/20%) shell, and NaYF₄:Nd/Yb(20/40%) shell. All of the spectra were recorded under excitation by a 980 CW laser. The inset shows the luminescence nm photographs of NaYF₄:Nd/Yb/Er(1/19/0.5%)@NaYF₄:Nd(20%) core-shell nanoparticles in cyclohexane solution upon irradiation of a 980 nm laser.



Figure S6. Upconversion emission spectra for NaYF₄:Nd/Yb/Tm(1/19/0.5%) nanoparticles and the corresponding core/shell nanoparticles coated with an inert NaYF₄ shell or an active NaYF₄:Nd/Yb(20/10%) shell. All of the spectra were recorded under excitation by a 980 nm CW laser. The inset shows the luminescence photographs of NaYF₄:Nd/Yb/Tm(1/19/0.5%)@NaYF₄ core/shell nanoparticles in cyclohexane solution upon irradiation of a 980 nm laser.