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## SUPPORTING INFORMATION

## Role of Inert Shell in improving Energy Utilization in Lanthanide-doped Upconversion Nanoparticles

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**Figure S1.** TEM images of NaYF<sub>4</sub>:Yb/Er(40/2%) core and NaYF<sub>4</sub>:Yb/Er(40/2%)@NaYF<sub>4</sub> coreshell nanoparticles. Scale bars are 100 nm.



**Figure S2.** XRD of NaYF<sub>4</sub>:Yb/Er(40/2%) core (A) and NaYF<sub>4</sub>:Yb/Er(40/2%)@NaYF<sub>4</sub> core-shell (B) nanoparticles. The diffraction pattern at the bottom is the literature reference for hexagonal NaYF<sub>4</sub> crystal (Joint Committee on Powder Diffraction Standards file number 16-0334).



Figure S3. Integrating sphere setup for quantum yield measurement.



**Figure S4.** Schematic presentation showing the synthetic process of core-shell structure nanoparticles and TEM images of the as-synthesized nanoparticles.  $Yb^{3+}$  concentration is 40% in each doping area. Scale bar is 50 nm.



Figure S5. Scattered 975 nm laser beam by Yb-doped different structured nanoparticles: (a)  $NaYF_4@NaYF_4$ , (b)  $NaYF_4:Yb(40\%)@NaYF_4$ , (c)  $NaYF_4@NaYF_4:Yb(40\%)$ , (d)  $NaYF_4:Yb(40\%)@NaYF_4:Yb(40\%)$ , collected by integrating sphere.



**Figure S6.** Decay curves of 985 nm luminescence from Yb<sup>3+</sup> ions in NaYF<sub>4</sub>:Yb/Tm@NaYF<sub>4</sub> UCNPs with 20 %Yb<sup>3+</sup> (a), 40 %Yb<sup>3+</sup> (b), 60 %Yb<sup>3+</sup> (c) and 80 %Yb<sup>3+</sup> (d) doping in core upon 975 nm laser excitation. The doping concentration of Tm<sup>3+</sup> in each groups varies from 0 to 2%.

$\bigcirc \longrightarrow \bigcirc$			
NaYF <sub>4</sub> :Yb/Er		NaYF <sub>4</sub> :Yb/Er@NaYF <sub>4</sub>	
	Yb: 0 %	Yb: 20 %	Yb: 40 %
Er: 0.5 %	3.0	9.1	20.1
Er: 1 %	4.7	16.3	67.4
Er: 2 %	5.7	20.5	69.0
Er: 4 %	6.4	21.0	76.8
Er: 8 %	21.1	29.0	86.0
Er: 16 %	30.5	92.4	227.7
Er: 32 %	166.7	354.9	
Er: 60 %	547.2		> 20000

**Figure S7.** Photoluminescence enhancement factor of NaYF<sub>4</sub>:Yb/Er@NaYF<sub>4</sub> compare to the corresponding core counterparts NaYF<sub>4</sub>:Yb/Er nanoparticles upon 975 nm laser excitation with power density of 50 W/cm<sup>2</sup>.



**Figure S8.** NIR (975 nm) excited Upconversion luminescence spectra of NaYF<sub>4</sub>:Yb/Er(20/2%) UCNPs with different NaYF<sub>4</sub> shell thickness. The spectra are normalized to  $Er^{3+}$  emission at 540 nm.



Figure S9. Average sizes of different NaYF<sub>4</sub>:Yb/Er@NaYF<sub>4</sub> UCNPs shown in Figure 5.



**Figure S10.** Schematic of the growing process of large length-to-width ratio core-shell structured  $NaYF_4$ :Yb/Er@NaYF<sub>4</sub> nanorods and TEM images of UCNPs with reaction time of 0 h (a), 2 h (b) 4 h (c) and 6 h (d).



**Figure S11.** Schematic illustration of the route for energy loss in sphere-like and rod-like core-shell structure UCNPs.