Supporting Information

Thermo-Enhanced Upconversion Luminescence in Inert-Core/Active-Shell UCNPs: Inert-Core Matters

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Fig. S1 The size distributions of (a) NaGdF₄ inert-core and (b) NaGdF₄@NaGdF₄: Yb/Tm inert-core/active-shell UCNPs.



Fig. S2 (a₁-c₁) TEM micrographs of the NaGdF₄ inert-core with different sizes about 17 nm, 27

nm, 43 nm, respectively. (a_2-c_2) The corresponding TEM micrographs after coating active-shell (about 1.5 nm thickness). (a_3-c_3) The integrated UCL intensities of NaGdF₄@NaGdF₄: Yb/Tm (20/1 mol%) UCNPs with different inert-core size.



Fig. S3 Temperature-dependent integrated UCL intensities for NaGdF₄: Yb/Tm (20/1 mol%, \sim 12 nm) and NaGdF₄@NaGdF₄: Yb/Tm (20/1 mol%, \sim 9@3 nm) UCNPs.



Fig. S4 Temperature-dependent integrated UCL intensities including 475 nm and 802 nm for NaGdF₄@NaGdF₄: Yb/Tm (20/1 mol%) UCNPs with different sizes of (a) ~9@3 nm, (b) ~17@3 nm, (c) ~27@3 nm (d) ~43@3 nm.



Fig. S5 Temperature-dependent UCL emission spectra ($\lambda_{ex} = 980$ nm) of NaGdF₄@NaGdF₄: Yb/Er (20/2 mol%) inert-core/active shell UCNPs with different sizes of (a) ~9@3 nm, (b) ~17@3 nm, (c) ~27@3 nm (d) ~43@3 nm. (e) Temperature-dependent integrated UCL intensities for NaGdF₄@NaGdF₄: Yb/Er (20/2 mol%) UCNPs with different inert-core sizes.



Fig. S6 Temperature-dependent decay lifetimes for ${}^{1}G_{4}$ excited state of Tm³⁺ in NaGdF₄@NaGdF₄: Yb/Tm (20/1 mol%) UCNPs with different sizes of (a) ~9@3 nm, (b) ~17@3 nm and (c) ~43@3 nm.



Fig. S7 Temperature-dependence DCL emission spectra of NaGdF₄@NaGdF₄: Yb/Tm (20/1 mol%) UCNPs with different sizes of (a) \sim 9@3 nm, (b) \sim 17@3 nm and (c) \sim 43@3 nm.



Fig. S8 The lifetime ratio for ${}^{2}F_{5/2}$ excited state of Yb³⁺ in NaGdF₄@NaGdF₄: Yb/Tm (20/1 mol%) UCNPs of different sizes.



Fig. S9 Simulation distribution diagram of rare earth ion in the inert-core/active-shell structure with a certain thickness of shell.



Fig. S10 Plots of the temperature-dependence of the interplanar spacing expansion factor $I_{\rm T}$ for the

(110) and (201) planes of the samples.