Supporting Information

Near-infrared optical and X-ray computed tomography dual-modal imaging probe based on novel lanthanide-Doped $K_{0.3}Bi_{0.7}F_{2.4}$ upconversion nanoparticles

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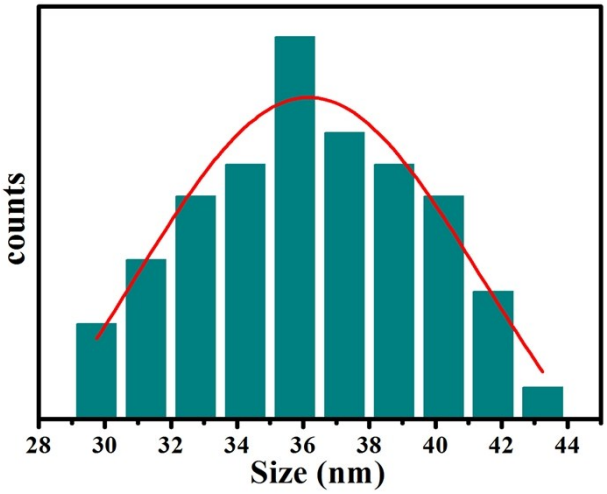

Fig. S1 The size histogram of BYT UCNPs.

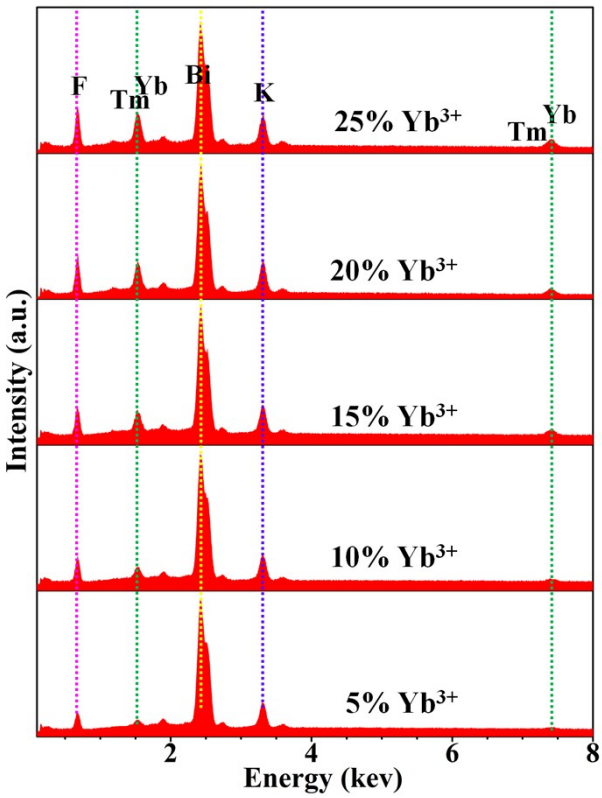

Fig. S2 Energy-dispersive X-ray (EDX) spectra of $K_{0.3}Bi_{0.7}F_{2.4}$:x% Yb^{3+}/0.5% Tm^{3+} UCNPs (x = 5, 10, 15, 20, 25).

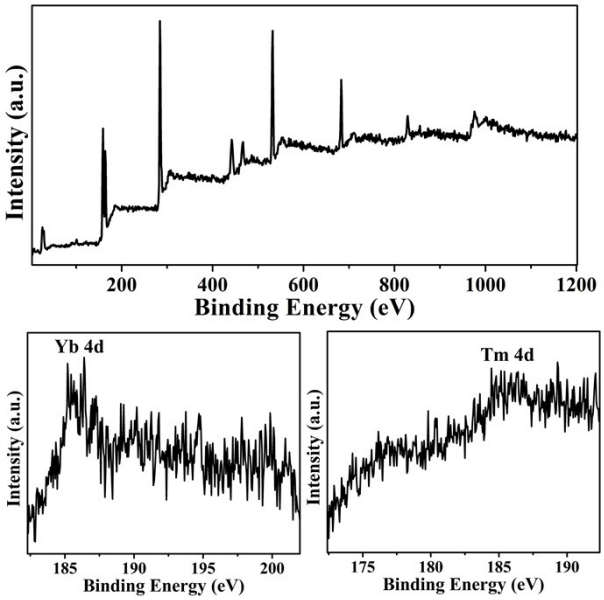

Fig. S3 X-ray photoelectron spectroscopy (XPS) analysis of BYT UCNPs. (a) survey, (b) Yb 4d, and (c) Tm 4d.

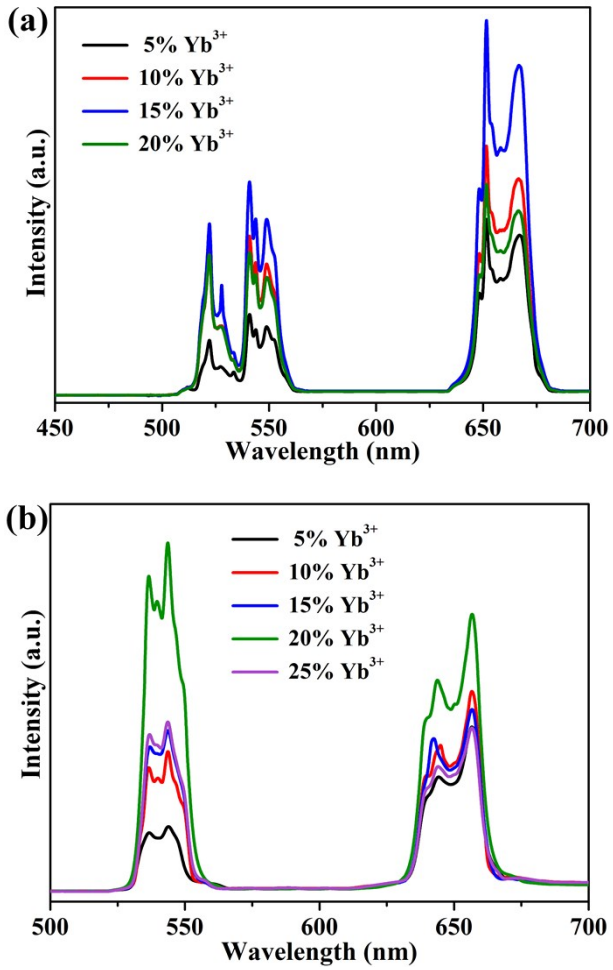

Fig. S4 (a) The UCL spectra of $K_{0.3}Bi_{0.7}F_{2.4}$:x% Yb^{3+}/2% Er^{3+} (x = 5, 10, 15, 20). (b) The UCL spectra of $K_{0.3}Bi_{0.7}F_{2.4}$:x% Yb^{3+}/2% Ho^{3+} (x = 5, 10, 15, 20, 25).

As shown in Fig. S4(a), $K_{0.3}Bi_{0.7}F_{2.4}$:x% Yb^{3+}/2% Er^{3+} (x = 5, 10, 15, 20, 25) exhibit two UC bands of green and red emissions in the spectroscopic range of 510-565 nm and 635-680 nm, which can be assigned to the $^2H_{11/2}/^4S_{3/2} \rightarrow {}^4I_{15/2}$ and $^4F_{9/2} \rightarrow {}^4I_{15/2}$ transitions of Er^{3+} ions. The intensity of emission peaks increases with Yb^{3+} ions concentration changing from 5 to 15%, and then decrease at the Yb^{3+} concentration of 20%. It is worthwhile to mention that a yellow emission was observed in $K_{0.3}Bi_{0.7}F_{2.4}$:15% Yb^{3+}/2% Er^{3+} by the naked eyes.

In the emission spectra of $K_{0.3}Bi_{0.7}F_{2.4}$:x% Yb^{3+}/2% Ho^{3+} (x = 5, 10, 15, 20, 25) (Fig. S4 (b)), the green emission at 525-562 nm corresponds to $^5S_2 \rightarrow {}^5I_8$ transitions and the red

emission region at 628-677 nm is ascribed to $^5F_5 \rightarrow {}^5I_8$ transition of Ho^{3+}. Variations in the Yb^{3+} concentration (5-25%) lead to corresponding changes in the green ($^5S_2 \rightarrow {}^5I_8$) and red ($^5F_5 \rightarrow {}^5I_8$) spectral region, accompanied by changes of fluorescence emission. The UCL intensity achieved the maximum when the Yb^{3+} ions concentration reaches at 20%.

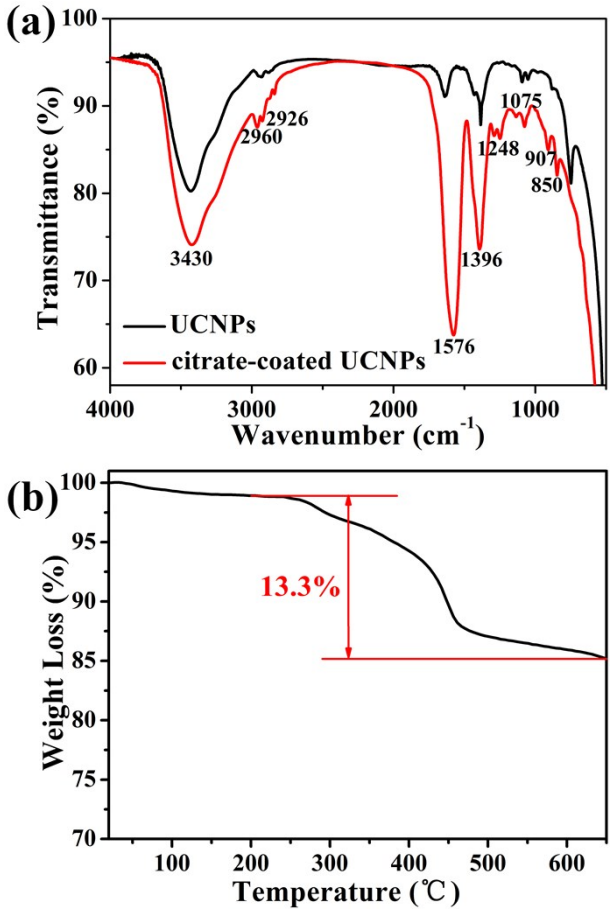

Fig. S5 (a) FTIR spectra of BYT UCNPs and citrate-coated BYT UCNPs. (b) Thermogravimetric analysis (TGA) of citrate-coated BYT UCNPs.

Fig. S5(a) shows the BYT UCNPs and citrate-coated BYT UCNPs both have the broad absorption band located at 3430 cm^{-1} corresponding to the O–H stretching vibration of water on the surface. For the citrate-coated BYT UCNPs, the weak absorption peaks (2960 and 2926 cm^{-1}) and the strong absorption peaks (1576 and 1396 cm^{-1}) are attributed to the C−H bond vibration of the surface coated citric acid molecules. The weak absorption peaks at 1248 and 1075 cm^{-1} are attributed to the C-C bond vibration of citric acid molecules. In addition, the O-H bond vibration of carboxylic group was displayed at 907 and 850 cm^{-1}. On the basis of the above results, the citric acid ligands have been successfully coated on the surface of UCNPs. The weight loss of citrate-coated BYT UCNPs was about 13.3 wt%, indicating the

amount of citric acid molecules on the surface of UCNPs (Fig. S5(b)).

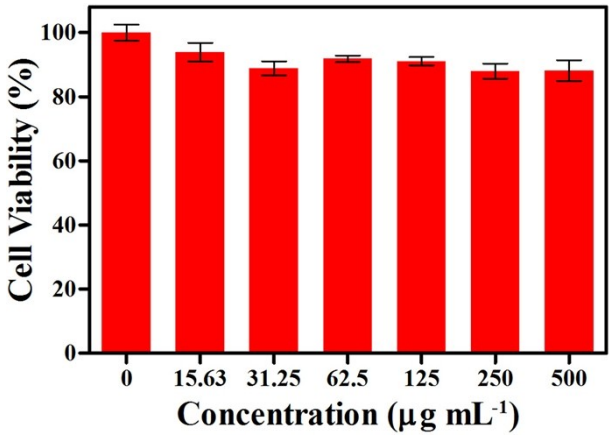

Fig. S6 *In vitro* cell viability of HeLa cells after incubation with citrate-coated BYT UCNPs for 24 h using standard MTT colorimetric assay.

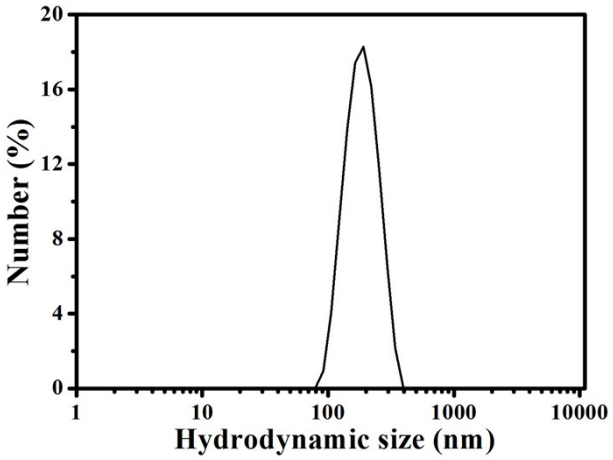

Fig. S7 The hydrodynamic size of BYT UCNPs.

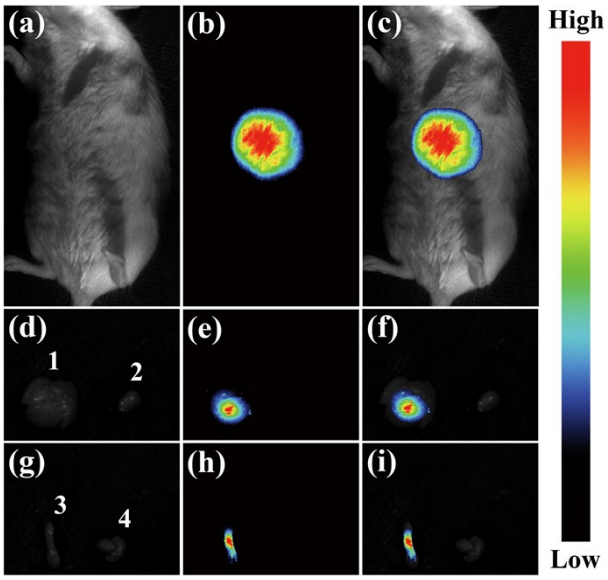

Fig. S8 Whole-body NIR-to-NIR UCL imaging of a mouse after intravenous injection of the citrate-coated BYT UCNPs. (a, d, g) bright-field images. (b, e, h) UCL images. (c, f, i) corresponding overlay bright-field and UCL images of (a, d, g and b, e, h). The (d-i) images are *ex vivo* UCL imaging after injection for 2 h. 1, liver; 2, heart; 3, spleen; 4, kidney.

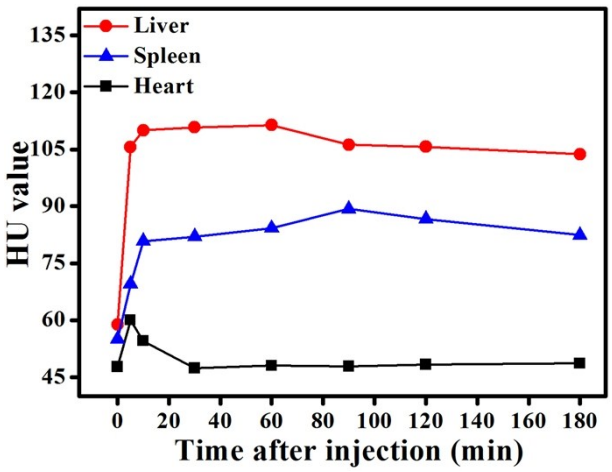

Fig. S9 The HU values of different organs of a mouse after intravenous injection of citrate-coated BYT UCNPs at timed intervals.