**Supporting Information for** 

## X-ray recharged long afterglow luminescent nanoparticles MgGeO<sub>3</sub>: Mn<sup>2+</sup>, Yb<sup>3+</sup>, Li<sup>+</sup> in the first and second biological windows for longterm bioimaging

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Figure S1. (a)-(b) TEM images of as-prepared MGO nanoparticles without further centrifugal

screening.



**Figure S2.** (a) Persistent luminescence spectra of MgGeO<sub>3</sub>:Mn<sup>2+</sup>, Yb<sup>3+</sup>, Li<sup>+</sup> (red lines) and ZnGa<sub>2</sub>O<sub>4</sub>: Cr<sup>3+</sup> (purple lines) at 10 s after 2 min of UV irradiation. (b) Persistent luminescence decay curve of MgGeO<sub>3</sub>:  $Mn^{2+}$ , Yb<sup>3+</sup>, Li<sup>+</sup> (red lines) and ZnGa<sub>2</sub>O<sub>4</sub>: Cr<sup>3+</sup> (purple lines) after 5 min of UV irradiation.



Figure S3. The quantum efficiency of CCD camera.



**Figure S4.** (a) NIR-I persistent luminescence spectra of MgGeO<sub>3</sub>:Mn<sub>0.1%</sub>, Yb<sup>3+</sup><sub>0.5%</sub>, Li<sup>+</sup><sub>x</sub> (x=0- 3 mol %) monitored at 680 nm after 254 nm UV irradiation for 2 min. (b) NIR-II persistent luminescence emission



spectra recorded at 5 s after 2 min irradiation of 254 nm. (c) The relationship between persistent luminescence intensity of MGO and the concentration of Li<sup>+</sup>.

**Figure S5.** (a) Comparison of persistent luminescence intensity under X-ray (62.9 mW/cm<sup>2</sup>) and UV (80mW/cm<sup>2</sup>) irradiation for 2 min, respectively. (b) persistent luminescence signal beyond 980 nm after recharging by X-ray and UV. (c) persistent luminescence signal of MGO beyond 980 nm for three charged cycles with X-ray. (d)-(e) persistent luminescence signal of MGO beyond 980 nm after stopping initial X-ray irradiation for 2 h and then were *in situ* photo-stimulated by 650 nm and 808 nm laser for three imaging cycles.





**Figure S6.** (a) Thermoluminescence curves of MGO powder under X-ray (62.9 mW/cm<sup>2</sup>; black line) and UV excitation (80mW/cm<sup>2</sup>; red line), respectively. (b) The MGO sample under X-ray charging 2min with 130 min waiting (black line) and photo-stimulation by 650 nm laser for 3 min at 273 k after X-ray excitation with 120 min waiting (red line), photo-stimulation in the same condition with 808 nm laser (blue line). (c) The schematic diagram of long persistent luminescence process in the MGO matrix.



Figure S7. in vivo imaging excited by X-ray after intravenously injected in a normal mouse.