## **Electronic Supplementary Information**

Simultaneous realization of Hg<sup>2+</sup> sensing, magnetic resonance imaging and upconversion luminescence *in vitro* and *in vivo* bioimaging based on hollow mesoporous silica coating UCNPs and ruthenium complex

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Fig. S1. Dynamic light scattering (DLS) of Ru-UCNPs@HmSiO<sub>2</sub>-PEI in water.



**Fig. S2**. The zeta potential of UCNPs@mSiO<sub>2</sub>, UCNPs@mSiO<sub>2</sub>-PEI, UCNPs@HmSiO<sub>2</sub>-PEI, and Ru-UCNPs@HmSiO<sub>2</sub>-PEI.



Fig. S3. The XRD patterns of UCNPs, UCNPs@mSiO<sub>2</sub>, UCNPs@HmSiO<sub>2</sub>-PEI, Ru-UCNPs@HmSiO<sub>2</sub>-PEI, and the standard card of  $\beta$ -NaYF<sub>4</sub> (JCPDS: 16-0334).



Fig. S4. Energy dispersive X-ray (EDX) spectrum of Ru-UCNPs@HmSiO<sub>2</sub>-PEI.



Fig. S5. FT-IR spectra of the UCNPs@mSiO<sub>2</sub>, UCNPs@HmSiO<sub>2</sub>-PEI, and Ru-UCNPs@HmSiO<sub>2</sub>-PEI.



Fig. S6. (A)  $N_2$  adsorption-desorption isotherm of Ru-UCNP@HmSiO<sub>2</sub>-PEI; (B) The pore size distribution of Ru-UCNPs@HmSiO<sub>2</sub>-PEI.



**Fig. S7.** (A) The photos of Ru-UCNPs@HmSiO<sub>2</sub>-PEI in water, PBS and DMEM culture solution; (B) The photos of Ru-UCNPs@HmSiO<sub>2</sub>-PEI placed in water, PBS and DMEM culture solution after one week; (C) UV-Vis aborption spectra of Ru-UCNPs@HmSiO<sub>2</sub>-PEI in PBS solution on first day and after one week; (D) UCL spectra of Ru-UCNPs@HmSiO<sub>2</sub>-PEI in PBS solution on first day and after one week.



Fig. S8. Thermogravimetry (TG) curves of UCNPs@HmSiO<sub>2</sub>-PEI and Ru-UCNPs@HmSiO<sub>2</sub>-PEI.



**Fig. S9.** The Ru complex concentrations of Ru-UCNPs@HmSiO<sub>2</sub>-PEI and Ru-UCNPs@mSiO<sub>2</sub>-PEI were calculated by using the detailed titration spectra. (A) UV/Vis absorption spectra of Ru complex with different concentrations. (B) The absorption intensity at 532 nm as a function of Ru complex concentration. The Ru complex contents of Ru-UCNPs@HmSiO<sub>2</sub>-PEI and Ru-UCNPs@mSiO<sub>2</sub>-PEI were determined as 23.4  $\mu$ M (15.7 wt%) and 15.0  $\mu$ M (10.1 wt%), respectively. The concentration of Ru-UCNPs@HmSiO<sub>2</sub>-PEI and Ru-UCNPs@mSiO<sub>2</sub>-PEI are both 0.15 mg·mL<sup>-1</sup>.



Fig. S10. The sensitivity test of Ru-UCNPs@HmSiO<sub>2</sub>-PEI towards Hg<sup>2+</sup> by using upconversion luminescence emission technique. The limit of detection limit (LOD) was given by the equation LOD=  $3S_0/S$ ; where 3 is the factor at the 99% confidence level,  $S_0$  the standard deviation of the blank measurements (n = 10,  $S_0$ =0.001392), and S is the slope of the calibration curve. The limit of detection (LOD) was determined to be 0.16  $\mu$ M.



**Fig. S11.** *In vitro* cell viabilities of HeLa cells incubated with Ru-UCNPs@HmSiO<sub>2</sub>-PEI at different concentrations (0, 50, 100, 200, 300, 400  $\mu$ g·mL<sup>-1</sup>) for 24 h.