

## Electronic Supplementary Information

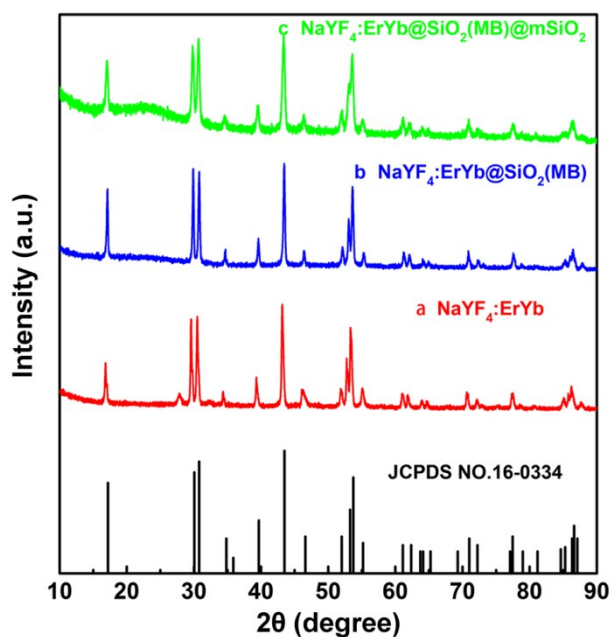
### **pH-responsive drug release and NIR-triggered singlet oxygen generation based on multifunctional core-shell-shell structure**

Renlu Han,<sup>a</sup> Haopeng Yi,<sup>a</sup> Jnhui Shi,<sup>a</sup> Zongjun Liu,<sup>b</sup> Hao Wang,<sup>a</sup> Yafei Hou<sup>a</sup> and You Wang<sup>\*a</sup>

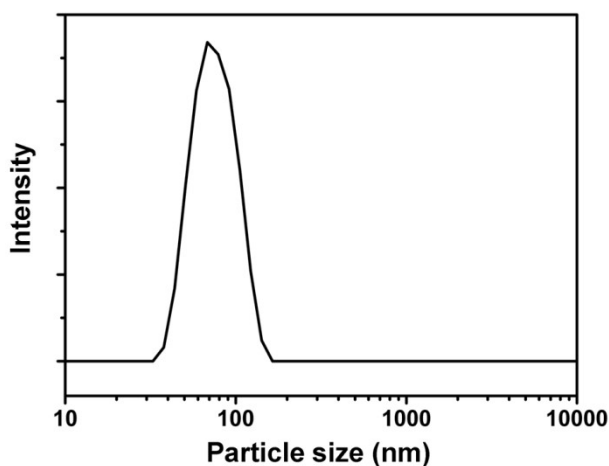
<sup>a</sup> School of Materials Science and Engineering, Harbin Institute of Technology, Harbin 150001, People's Republic of China.

<sup>b</sup> School of Chemical Engineering and Technology, Harbin Institute of Technology, Harbin 150001, People's Republic of China.

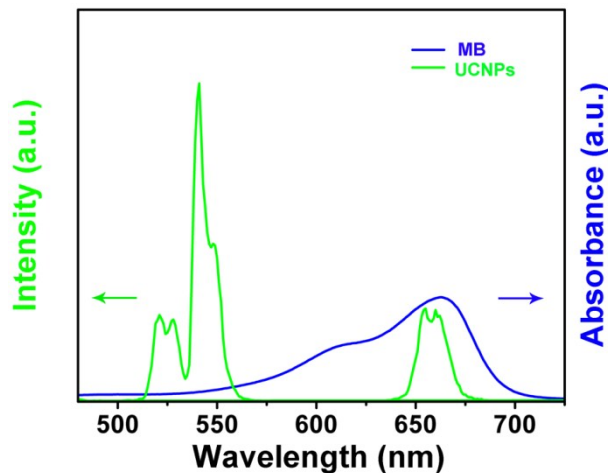
\* Corresponding author E-mail: [y-wang@hit.edu.cn](mailto:y-wang@hit.edu.cn).



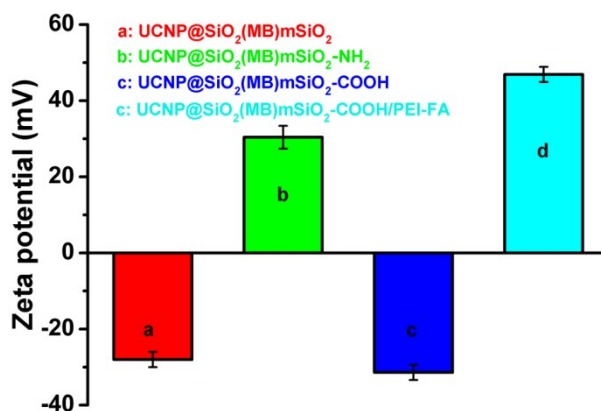
**Fig. S1** Powder X-ray diffraction (XRD) patterns for NaYF<sub>4</sub>:ErYb (a), NaYF<sub>4</sub>:ErYb@SiO<sub>2</sub>(MB) (b) and NaYF<sub>4</sub>:ErYb@SiO<sub>2</sub>(MB)@mSiO<sub>2</sub> (c). The standard card of β-NaYF<sub>4</sub> (JCPDS: 16-0334) was given as a reference.



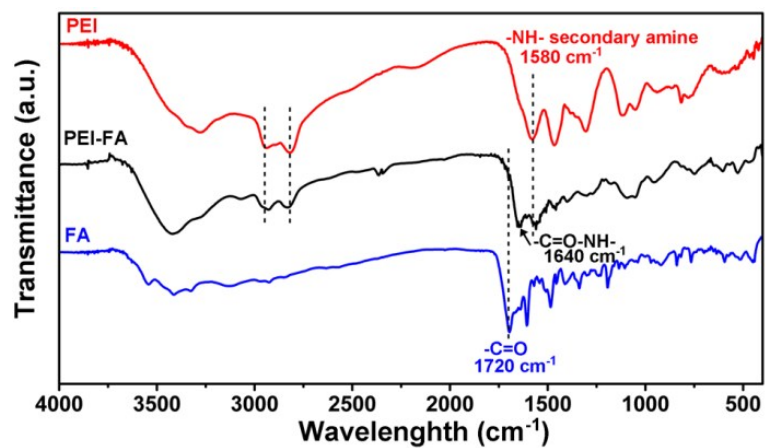
**Fig. S2** Particle size distribution of UCNP@SiO<sub>2</sub>(MB)@mSiO<sub>2</sub> nanoparticles obtained by DLS. The diameter of final core-shell-shell structure measured with DLS was 70 nm and larger than that obtained from TEM (Fig. S2), which would be attributed to the dynamic sizes of the nanoparticles.<sup>1,2</sup>



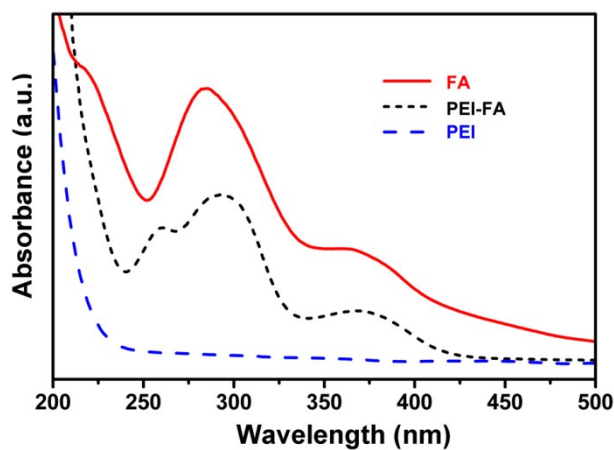
**Fig. S3** Upconversion fluorescent spectrum of UCNPs@SiO<sub>2</sub>@mSiO<sub>2</sub> solid powder under NIR laser excitation ( $\lambda=980\text{ nm}$ ,  $0.5\text{ W cm}^{-2}$ ) and UV/Vis absorbance spectrum of MB.



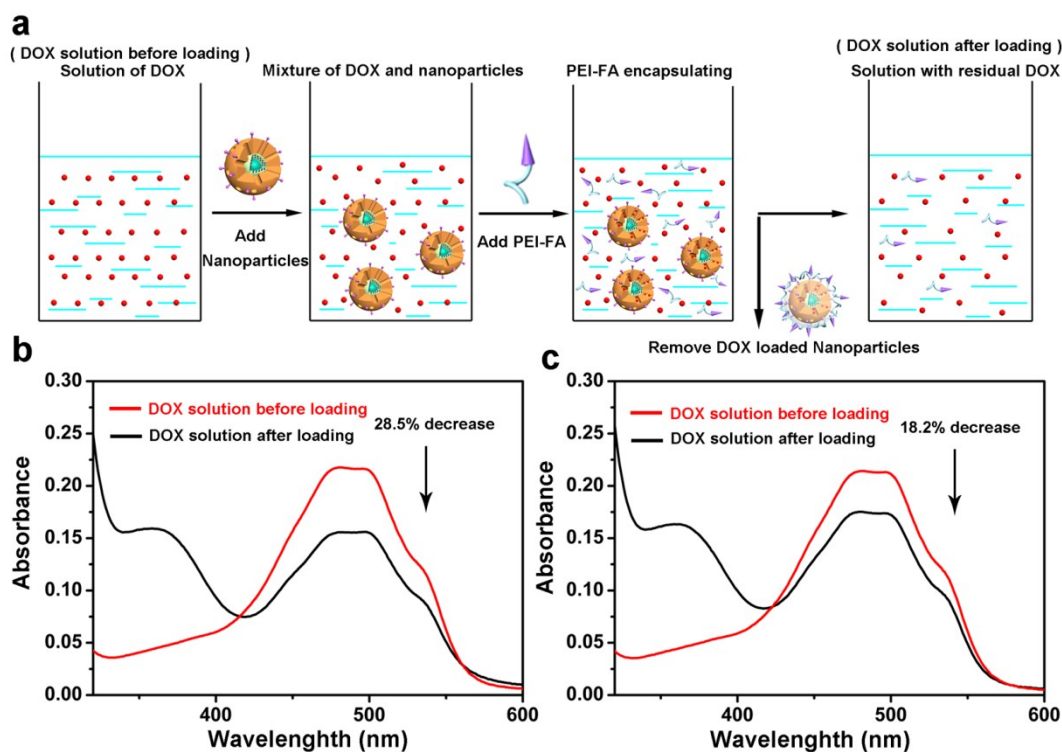
**Fig. S4** Zeta potentials of UCNPs@SiO<sub>2</sub>(MB)@mSiO<sub>2</sub> (a), UCNPs@SiO<sub>2</sub>(MB)@mSiO<sub>2</sub>-NH<sub>2</sub> (b), UCNPs@SiO<sub>2</sub>(MB)@mSiO<sub>2</sub>-COOH (c), and UCNPs@SiO<sub>2</sub>(MB)@mSiO<sub>2</sub>-COOH/PEI-FA (d).



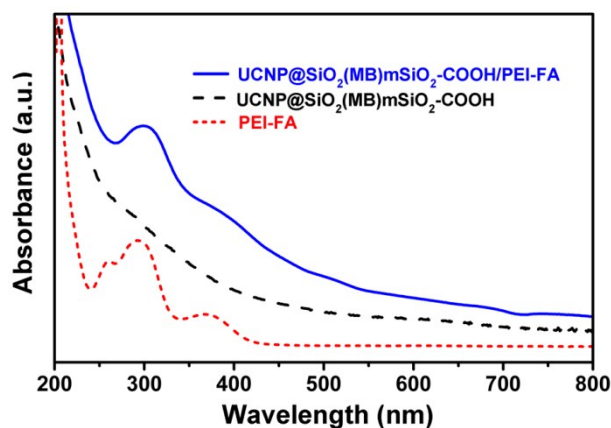
**Fig. S5** FTIR spectra of PEI (red) , PEI-FA (black) and FA (blue).



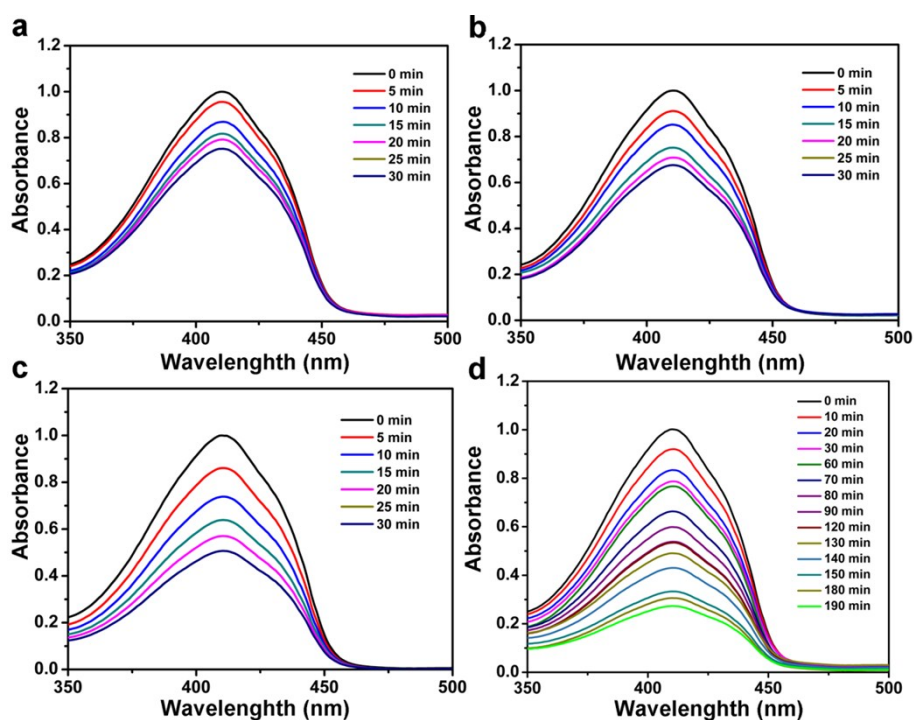
**Fig. S6** UV/Vis absorption spectra of FA (red), PEI-FA (black) and PEI (blue). All the samples were dissolved in DMSO.



**Fig. S7 (a)** Schematic illustration of the procedure to determine the DOX loading efficiency in as-prepared nanoparticles. (b) UV/Vis absorption spectra of DOX solution before and after loading for UCNP@SiO<sub>2</sub>(MB)@mSiO<sub>2</sub>-COOH. (c) UV/Vis absorption spectra of DOX solution before and after loading for UCNP@SiO<sub>2</sub>(MB)@mSiO<sub>2</sub>. Using Beer's Law, it can be calculated the DOX loading efficacy is 1.7 % (17 μg DOX in 1 mg nanoparticles) for UCNP@SiO<sub>2</sub>(MB)@mSiO<sub>2</sub>-COOH and 1.07 % (10.7 μg DOX in 1 mg nanoparticles) for UCNP@SiO<sub>2</sub>(MB)@mSiO<sub>2</sub>, respectively.<sup>3</sup>



**Fig. S8** UV/Vis absorption spectra of UCNP@SiO<sub>2</sub>(MB)@mSiO<sub>2</sub>-COOH/PEI-FA (blue), UCNP@SiO<sub>2</sub>(MB)@mSiO<sub>2</sub>-COOH (black) and PEI-FA (red). All the samples were suspended or dissolved in deionized water.



**Fig. S9** Original UV/Vis spectra of DPBF upon continuous 980 nm laser irradiation at a power density of 1.0 W cm<sup>-2</sup> (a), 2.0 W cm<sup>-2</sup> (b), and 3.5 W cm<sup>-2</sup> (c) for UCNP@SiO<sub>2</sub>(MB)@mSiO<sub>2</sub> nanoparticles in Fig. 6a. (d) Original UV-Vis spectrum of DPBF upon 980 nm laser ON/OFF irradiation at a power density of 0.6 W cm<sup>-2</sup> for UCNP@SiO<sub>2</sub>(MB)@mSiO<sub>2</sub> nanoparticles in Fig. 6b.

- 1 L. Xing, H. Zheng, Y. Cao and S. Che, *Adv. Mater*, 2012, **24**, 6433.
- 2 X. Ma, S. Sreejith and Y. Zhao, *ACS Appl. Mater. Interfaces*, 2013, **5**, 12860.
- 3 He, K. Krippes, S. Ritz, Z. Chen, A. Best, H. J. Butt, V. Mailander and S. Wu, *Chem, Commun.*, 2015, **51**, 431.