

## *Supporting Information*

# The Morphology and Surface Charge-Dependent Cellular Uptake Efficiency of Upconversion Nanostructures Revealed by Single-Particle Optical Microscopy

Di Zhang,<sup>‡a</sup> Lin Wei,<sup>‡b</sup> Meile Zhong,<sup>b</sup> Lehui Xiao,<sup>\*,a,b</sup> Hung-Wing Li,<sup>c</sup> and Jianfang Wang<sup>d</sup>

<sup>a</sup> State Key Laboratory of Medicinal Chemical Biology, Tianjin Key Laboratory of Biosensing and Molecular Recognition, College of Chemistry, Nankai University, Tianjin, 300071, China;

<sup>b</sup> Key Laboratory of Chemical Biology & Traditional Chinese Medicine Research, Key Laboratory of Phytochemical R&D of Hunan Province, College of Chemistry and Chemical Engineering, Hunan Normal University, Changsha, 410082, China;

<sup>c</sup> Department of Chemistry, Hong Kong Baptist University, Kowloon Tong, Hong Kong, SAR China;

<sup>d</sup> Department of Physics, The Chinese University of Hong Kong, Shatin, Hong Kong SAR, China.

<sup>‡</sup> These authors contributed equally to this work.

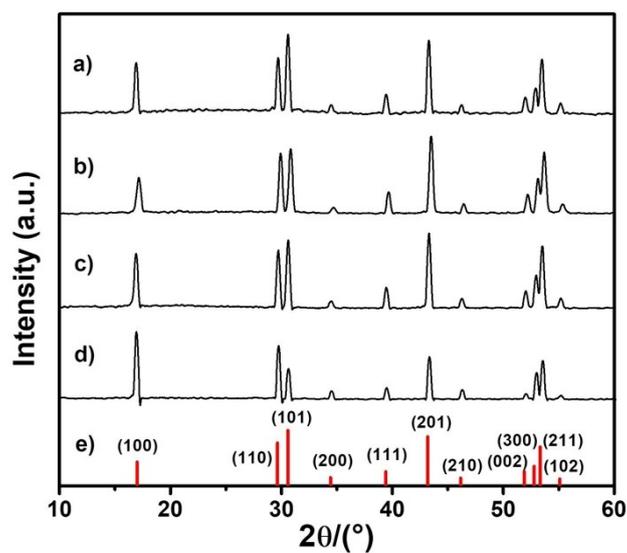
\* Corresponding author

Email: lehuixiao@nankai.edu.cn

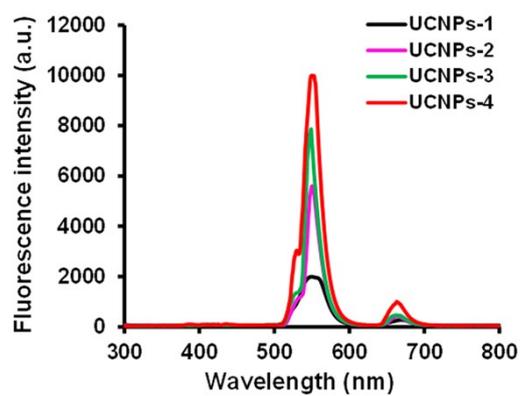
Fax: +86-022-23500201

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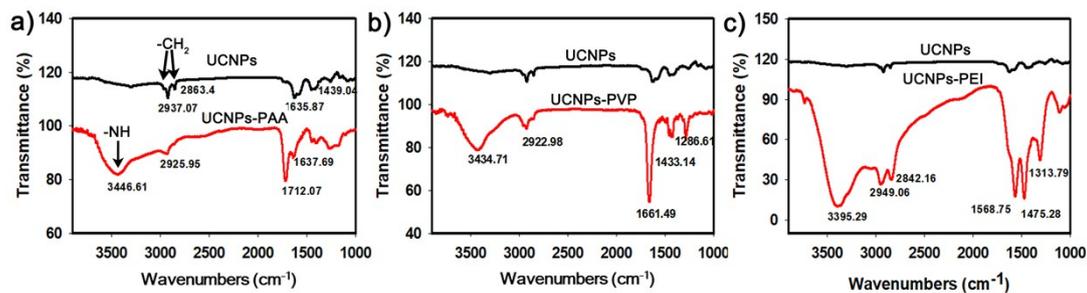
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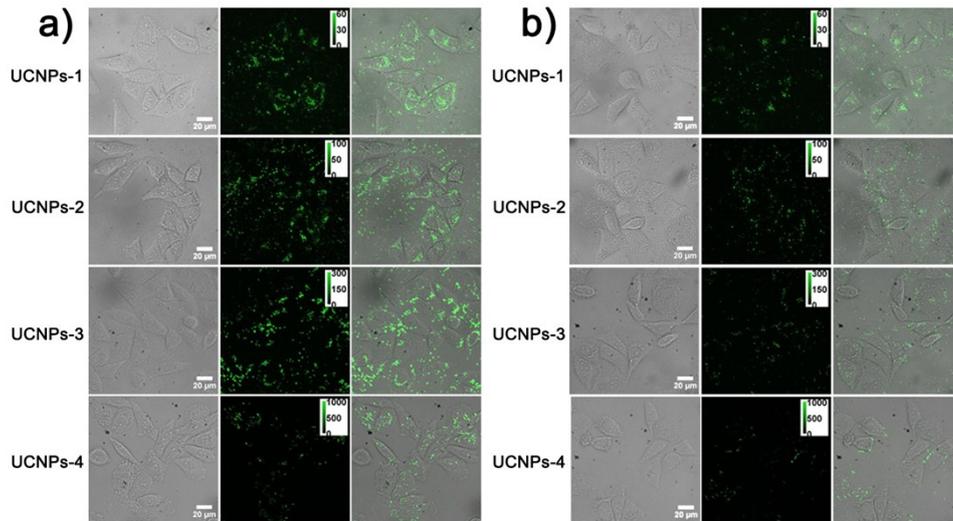
**Fig. S1.** X-ray power diffraction (XRD) spectra of NaYF<sub>4</sub>:Yb<sup>3+</sup>, Er<sup>3+</sup> UCNPs with various shapes a) UCNPs-1, b) UCNPs-2, c) UCNPs-3, d) UCNPs-4 and e) Line pattern of the calculated hexagonal phase NaYF<sub>4</sub> (PDF No. 16-0334).



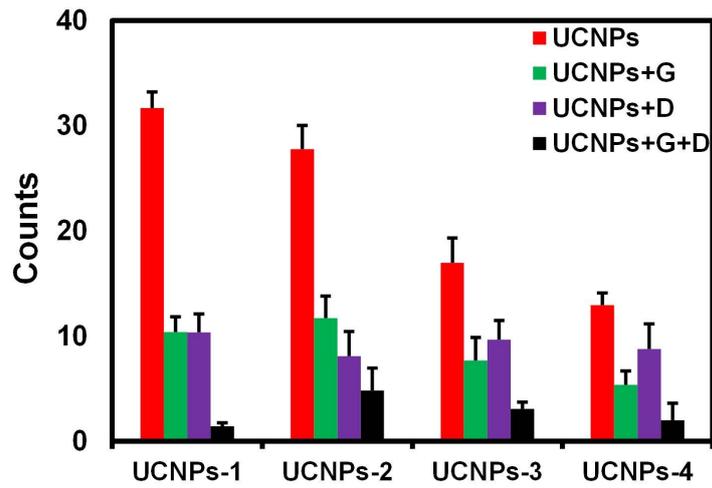
**Figure S2.** The fluorescence emission spectra of polymer-coated UCNPs dispersed in deionized water at the concentration of 1 mg/mL under excitation at 980 nm.



**Figure S3.** The FT-IR spectra of a) PAA, b) PVP, c) PEI modified UCNPs and untreated UCNPs. Absorption peaks at 2937 and 2863  $\text{cm}^{-1}$  correspond to the stretching vibration of methylene groups, which are the result of sodium citrate on the surface of UCNPs. Absorption peaks at 3446 and 1712  $\text{cm}^{-1}$  in a) are assigned to the vibrational absorption of hydroxyl groups and carbonyl groups, respectively, indicating the presence of carboxylic acid in PAA. An absorption peak at 1661  $\text{cm}^{-1}$  in b) reveals the stretching vibration of C=O in PVP. Absorption peaks at 3395 and 1568  $\text{cm}^{-1}$  in c) are the results of imino groups in polymer PEI.



**Figure S4.** a) and b) are the fluorescence microscopic characterizations of HeLa cells co-cultured with PEI-modified UCNPs-1, -2, -3 and -4 at 37 and 4 °C for 4h respectively. The images from left to right are the bright-field, fluorescence and merged microscopic images respectively.



**Figure S5.** The statistically counted number of UCNPs-PEI with various morphologies in single HeLa cells co-incubated without drug treatment (red), with Genistein (green), Dynasore (purple), and both of them (black) respectively.