Controllable Synthesis of $NaYF_4$:Yb,Er Upconversion Nanophosphors and Their Application to *in vivo* Imaging of *Caenorhabditis elegans*

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Hydrothermal preparation and characterization of β-$NaYF_4$:Yb,Er upconversion nanoparticles

In a typical green hydrothermal synthesis, a mixture containing 1.7614g Y_2O_3, 0.7882 g Yb_2O_3 , 0.07654 g Er_2O_3 was dissolved in about 8 mL concentrated nitric acid by heating and continuous stirring. When the solvent was slowly evaporated to dryness, rare earth nitrate powders were formed.[1] Then the powders were dissolved into 100 mL distilled water to get 0.2 M rare earth nitrates stock solution in which the molar ratio of Y^{3+}:Yb^{3+}:Er^{3+} was 0.78:0.20:0.02. Next, 5 mL of the as-prepared stock solution was added to a beaker containing 0.5645 g of [Bmim][BF_4], 1.6998 g of $NaNO_3$ and 10 mL of water. After stirring, the solution was adjusted to pH 10 by 1 M NaOH. Then the final solution was transferred to a Teflon-lined autoclave to conduct the reaction at 150 °C for 24 h. After the autoclave was cooled to room-temperature naturally, the product was separated by centrifugation, washed with distilled water and ethanol for three times, dried and collected for further characterization and application.

In this green hydrothermal synthesis, the dimension and morphology characterizations of the hydrothermally prepared UCNPs were performed by a JEM-2100HR transmission electron microscope (TEM, JEOL Ltd., Japan), using an acceleration voltage of 200 kV. Other apparatus and reagents are identical with our paper.

The TEM image, fluorescence spectrum and XRD pattern of the prepared UCNPs are shown in Fig. S1. It can be seen from Fig. S1a that the particles are irregularly spherical in shape with a diameter of approximately 150~200 nm, and relatively aggregated. Fig. S1b shows the fluorescence spectrum of nanocrystals obtained under the irradiation of a 980 nm laser, in which two green emissions at 521 and 540 nm correspond to $^2H_{11/2}\rightarrow{}^4I_{15/2}$ and $^4S_{3/2}\rightarrow{}^4I_{15/2}$ transitions of Er^{3+} ions, respectively, while a red emission at 654 nm corresponds to $^4F_{9/2}\rightarrow{}^4I_{15/2}$ transitions of Er^{3+} ions. The inset of Fig. S1b is the photograph of aqueous solution of the UCNPs under 980 nm excitation, showing the green emission from the solution. As shown in Fig. S1c, the peak positions and intensities of XRD are in good agreement with the calculated line pattern for β-phase $NaYF_4$ (the bottom curve, JCPDS NO. 028-1192).

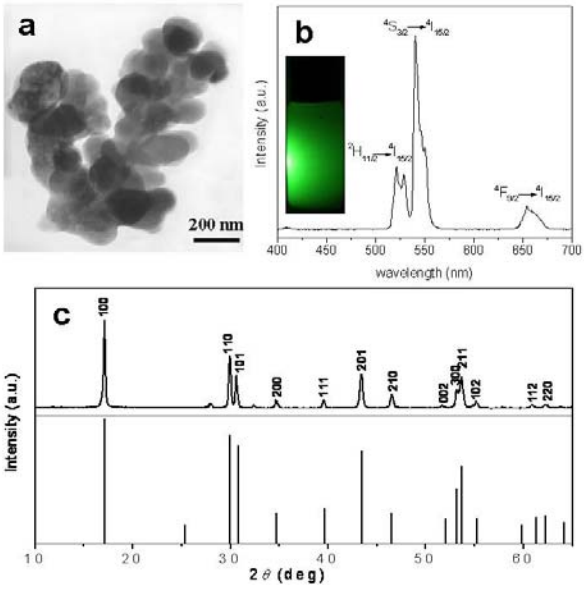

Fig. S1 Characteristics of the hydrothermal synthesized $NaYF_4$:Yb,Er upconversion nanoparticles. (a) TEM image. (b) Upconversion fluorescence spectrum. Inset is the photo of the UCNPs aqueous colloidal solution under 980 nm exciter. (c) Powder XRD pattern (top plot) and the calculated line pattern (bottom plot) for β-$NaYF_4$ (JCPDS No. 028-1192).

References

1 M. Wang, J. L. Liu, Y. X. Zhang, W. Hou, X. L. Wu and S. K. Xu, Mater. Lett., 2009, 63, 325-327.

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