

## Supporting Information

### pH- and surfactant-mediated tunable morphology and upconversion of rare-earth doped fluoride microcrystals

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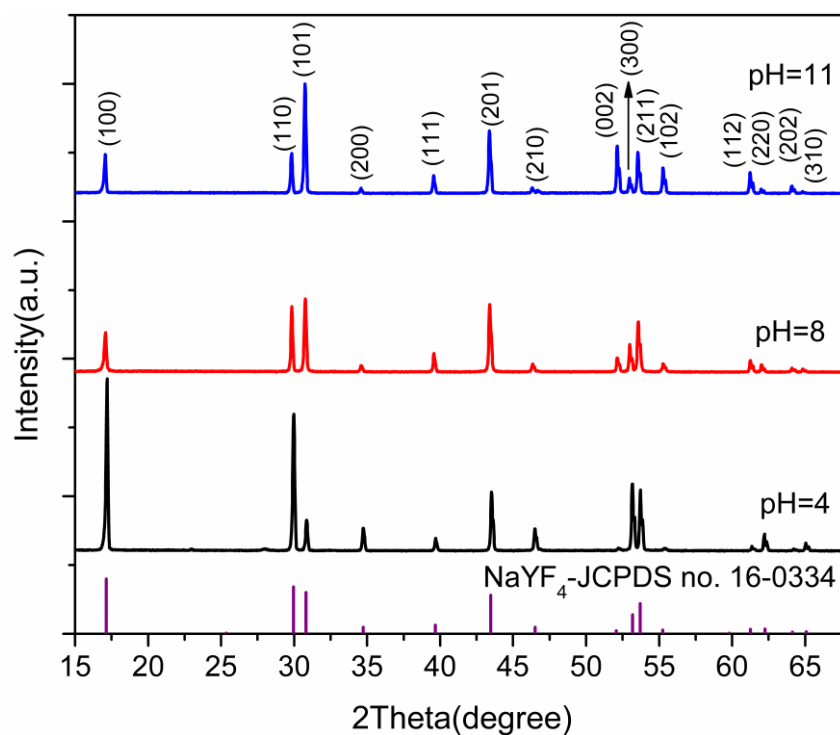


Fig. S1 XRD patterns of the NaYF<sub>4</sub> products at different pH values (2:1 molar ratio for Cit<sup>3+</sup>/Y<sup>3+</sup>). The standard peaks in the pure hexagonal β-NaYF<sub>4</sub> (JCPDS file number 16-0334) are used as a reference.

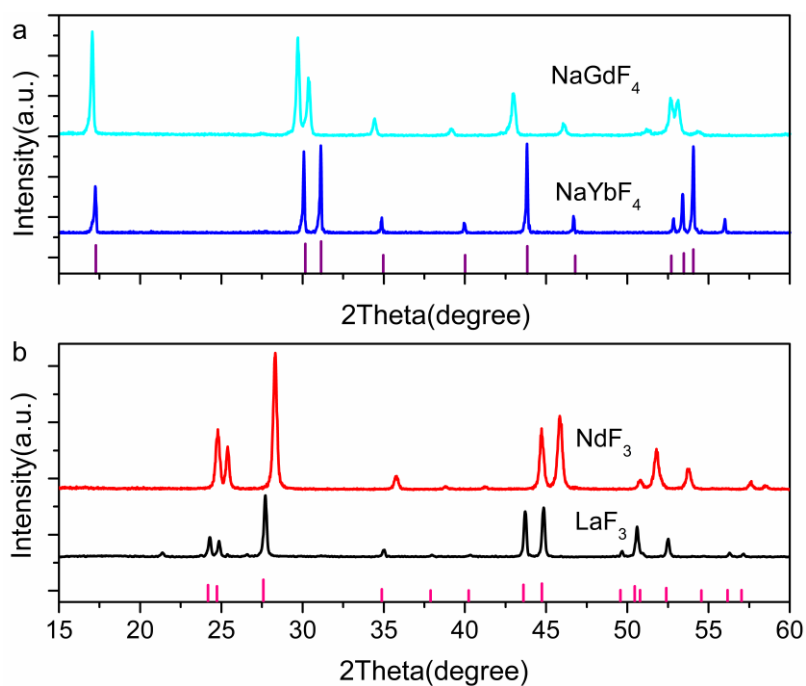


Fig. S2 XRD patterns of (a) NaYbF<sub>4</sub>, NaGdF<sub>4</sub> and (b) LaF<sub>3</sub>, NdF<sub>3</sub> products at the pH value of 8 (1:1 molar ratio for Cit<sup>3+</sup>/Y<sup>3+</sup>) and the standard peaks in the pure hexagonal β-NaYbF<sub>4</sub> (JCPDS file number 27-1427) (a) and β-LaF<sub>3</sub> (JCPDS file number 32-0483) (b) as references.

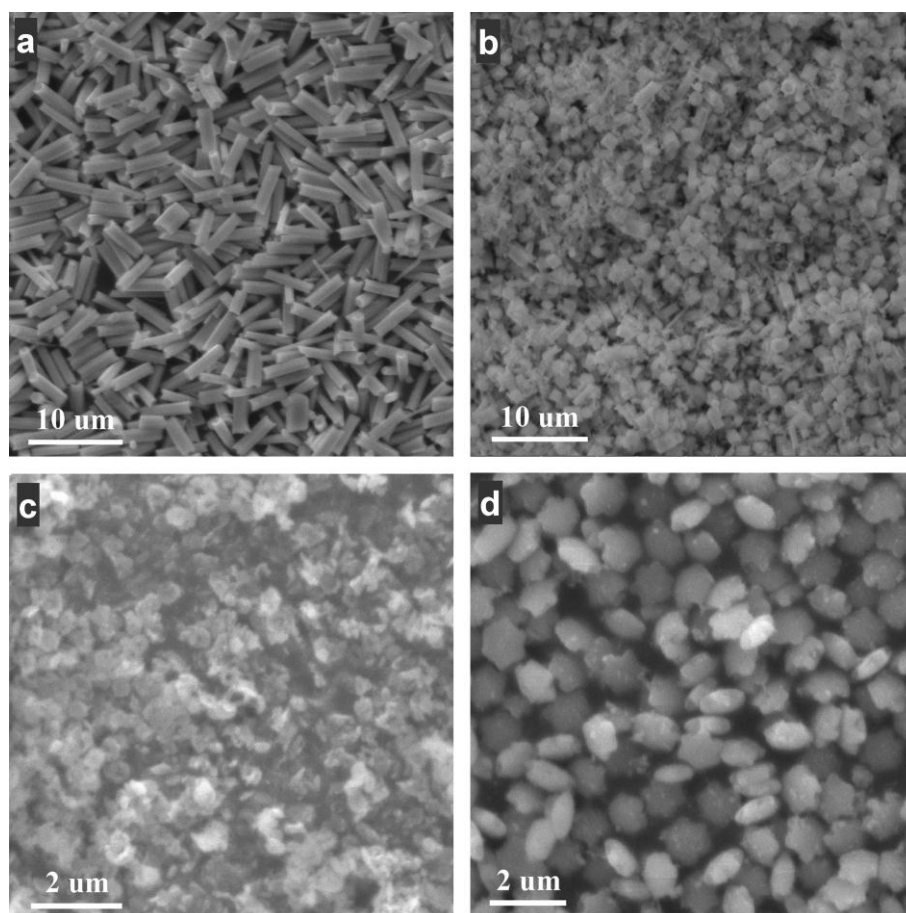


Fig. S3 SEM images of (a) NaYbF<sub>4</sub>, (b) NaGdF<sub>4</sub>, (c) LaF<sub>3</sub>, and (d) NdF<sub>3</sub> microcrystals obtained at pH 8. The molar ratio for Cit<sup>3+</sup>/Y<sup>3+</sup> was 1:1.

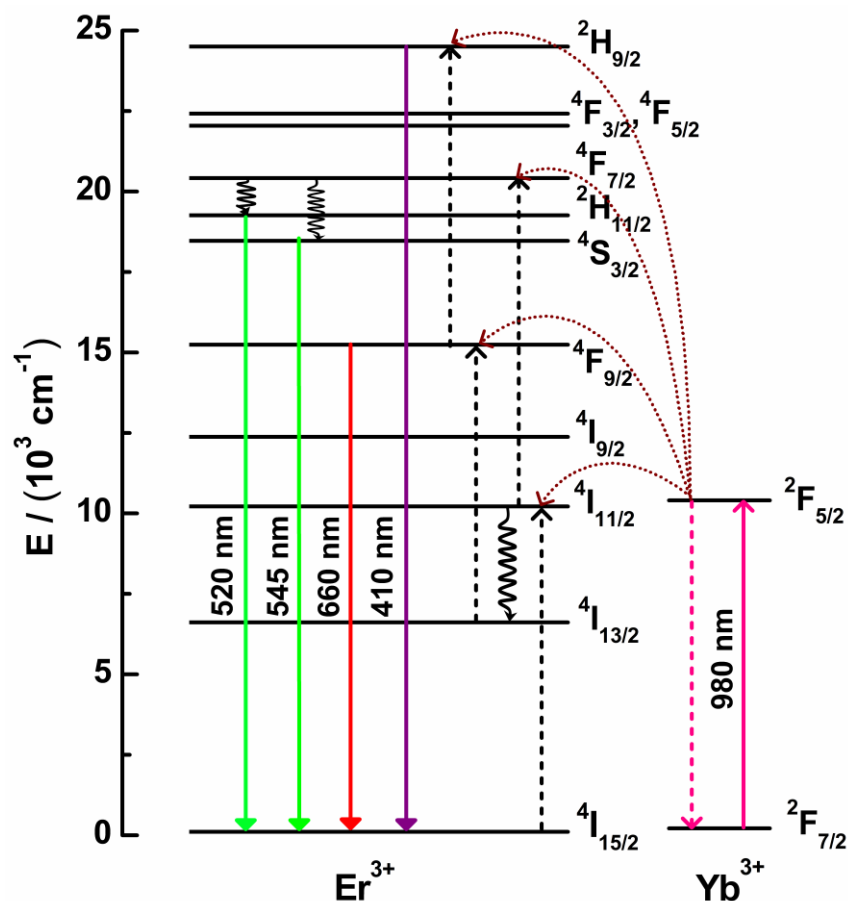


Fig. S4 The energy level diagrams of the  $\text{Er}^{3+}$  and  $\text{Yb}^{3+}$  dopant ions and upconversion mechanisms following 980 nm photons excitation. The full, dotted, and curly arrows represent emission, energy transfer, and multiphonon relaxation processes, respectively.

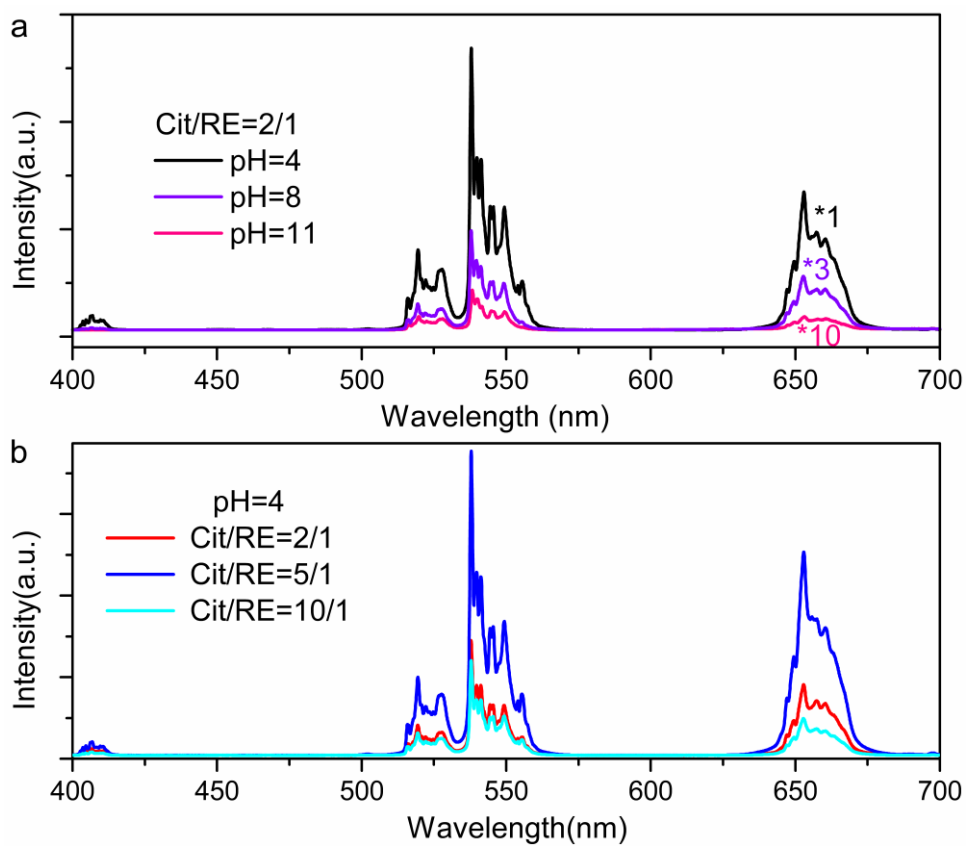


Fig. S5 Upconversion spectra for NaYF<sub>4</sub>: Yb/Er (18/2 mol%) microcrystals synthesized at the pH values of 4, 8 and 11 (a) and at Cit<sup>3+</sup>:RE<sup>3+</sup> ratios of 2:1, 5:1, and 10:1 (pH=4) (b). The samples were excited with a 977 nm laser operating at 400 mW.

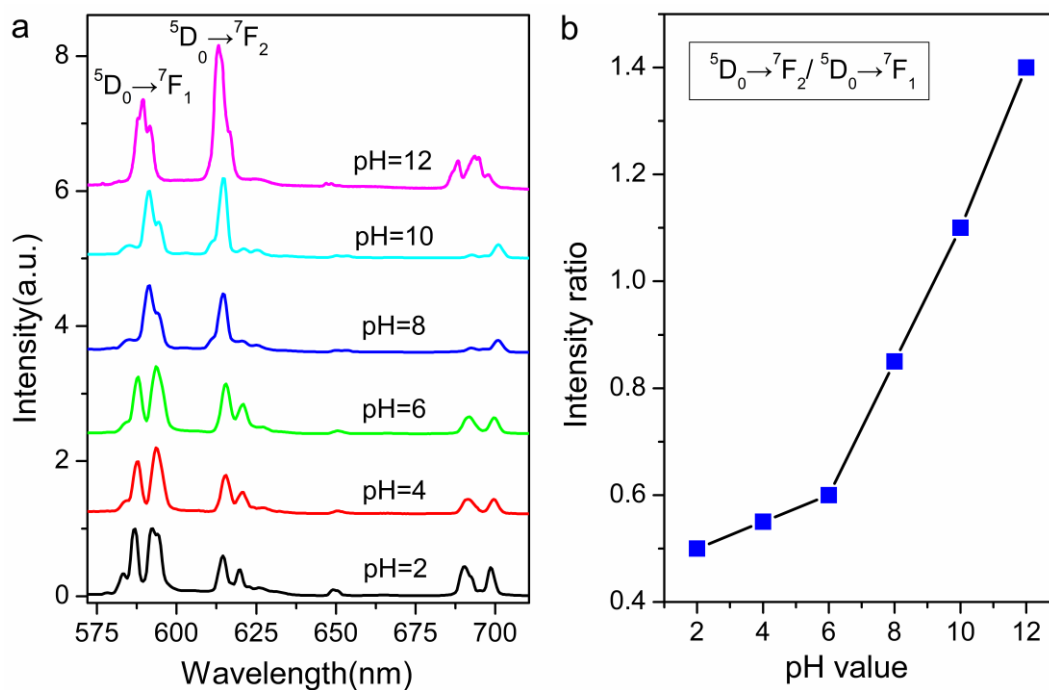


Fig. S6 Luminescence emission spectra of Eu<sup>3+</sup> doped fluoride samples prepared at different pH values in (a) and the ratio of luminescence intensities of  $^5D_0 \rightarrow ^7F_2$  to  $^5D_0 \rightarrow ^7F_1$  in (b). The samples were excited with a 532 nm laser.