Supporting Information for

Surface Lanthanide Activators Doping for Constructing Highly Efficient Energy Transfer Based Nanoprobe for on-site Monitoring Atmospheric Sulfur Dioxide

Cuilan Zhang,^{a,⊥} Xiao Ling,^{a,⊥} Qingsong Mei,^{a,*} Hongbo He,^a Shengsong Deng,^a Yong

Zhang^{b,c,*}

^a School of Food and Biological Engineering, Hefei University of Technology, Anhui 230009, China

^b Department of Biomedical Engineering, Faculty of Engineering, National University of Singapore, Singapore 117575, Singapore

^c School of Environmental and Chemical Engineering, Shanghai University, Shanghai
200444, China

 $^{\perp}$ These authors contribute equally to this work.

Corresponding authors: qsmei@hfut.edu.cn (Qingsong Mei), biezy@nus.edu.sg (Yong Zhang).



Figure S1. TEM images and particle size distributions in length direction and width direction of NaYF₄:Yb/Tm (a, d, g), NaYF₄@NaYF₄:Yb/Tm (b, e, h), NaYF₄@NaYF₄:Yb/Tm@ NaYF₄ (c, f, i).



Figure S2. The high angle annular dark field (HAADF) image and corresponding elemental mapping of F⁻, Y^{3+} , Tm^{3+} and Yb^{3+} ions of the NaYF₄@NaYF₄:Yb/Tm@NaYF₄ nanoparticles, and the EDS line scan profiles of a single nanoparticle along the line marked in HAADF image.



Figure S3. XRD patterns of three types of upconversional nanocrystrals.



Figure S4. The luminescence quenching kinetics of the $NaYF_4$ @ $NaYF_4$:Yb/Tm nanoparticles after additions of different amounts of cyanine dye.



Figure S5. The absorption spectral evolutions of cyanine dye upon additions of different concentrations of bisulfite ions. From top to bottom, the concentrations of HSO_3^- ions are 0, 1×10^{-9} , 1×10^{-8} , 1×10^{-7} , 1×10^{-6} , 1×10^{-5} , 1×10^{-4} and 1×10^{-3} M, respectively.



Figure S6. The lifetime decay curves of upconversion luminescence at 475 nm of the NaYF₄:Yb/Tm nanoparticles (a), NaYF₄@NaYF₄:Yb/Tm nanoparticles (b) and NaYF₄@NaYF₄:Yb/Tm@NaYF₄ nanoparticles (c) before (black line) and after (red line) conjugation with the cyanine dye.



Figure S7. The influence of pH values on the luminescence enhancement of the nanoprobe, where I_0 and I respectively represent the luminescence intensity of the nanoprobe before and after additions of 1 mM of bisulfite ions.



Figure S8. The front view (a) and side view (b) of the smartphone-based detection platform.



Figure S9. The luminescence quenching of $NaYF_4@NaYF_4:Yb/Tm$ nanoparticles upon additions of different concentrations of cyanine dye. From top to bottom, the concentrations of cyanine dye are 0, 10, 20, 30, 40, 50, 60, 70 and 80 μ M.



Figure S10. The luminescence image of five different spots on the test paper, showing the homogeneity of test paper.



Figure S11. The luminescence images of test paper after addition of a drop of water in a period of 10 minutes with an interval of 2 min, demonstrating the luminescent stability of the test paper.



Figure S12. The luminescent images of the test paper in a period of 50 minutes after exposure

to N_2 atmosphere with an interval of 5 min.



Figure S13. The luminescent images of the test paper in a period of 50 minutes after exposure

to outdoor air with an interval of 5 min.