Supporting Information for

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

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Figure S1. TEM images and particle size distributions in length direction and width direction of NaYF$_4$:Yb/Tm (a, d, g), NaYF$_4$@NaYF$_4$:Yb/Tm (b, e, h), NaYF$_4$@NaYF$_4$:Yb/Tm@NaYF$_4$ (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 NaYF\(_4\)@NaYF\(_4\):Yb/Tm@NaYF\(_4\) 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 nanocrystals.

Figure S4. The luminescence quenching kinetics of the NaYF₄@NaYF₄: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 NaYF$_4$:Yb/Tm nanoparticles (a), NaYF$_4$@NaYF$_4$:Yb/Tm nanoparticles (b) and NaYF$_4$@NaYF$_4$:Yb/Tm@NaYF$_4$ 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 μM.

**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\textsubscript{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.