## **Electronic Supplementary Information**

## All-in-One Theranostic Nanoplatform Based on Upconversion Dendritic Mesoporous Silica Nanocomposites for Synergistic Chemodynamic/Photodynamic/Gas Therapy

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Fig. S1. TEM images of a) UCNPs and b) CuO<sub>2</sub> at different magnifications.



Fig. S2. EDS spectrum of UMNO nanoparticles.



Fig. S3. a)  $N_2$  absorption-desorption isotherms and b) corresponding pore-size distribution of UMNOCC-PEG.



Fig. S4. The high resolution a) O 1s and b) Cu 2p XPS spectra of UMNOCC-PEG.



Fig. S5. Colorimetric analysis demonstrating the presence of peroxo groups in UMNOCC-PEG. Peroxo groups can reduce  $MnO_4^-$  to colorless  $Mn^{2+}$  in strong acidic media (0.1 M H<sub>2</sub>SO<sub>4</sub>).



**Fig. S6.** a) The particle size distributions of UMNOCC-PEG NPs in different solvents (including  $H_2O$ , phosphate buffered solution (PBS) and culture medium) measured by dynamic light scattering (DLS) and b) the supernatant obtained from the UMNOCC-PEG solutions in  $H_2O$ , PBS (pH 7.4) and culture medium after two days standing.



**Fig. S7.** Upconversion emission spectra of UMNOCC-PEG in aqueous solutions at different time periods.



**Fig. S8.** a) The standard curves for Ce6. b) The absorbance spectra of the initial Ce6 solution and the supernatant obtained after UMNOCC-PEG preparation.



Fig. S9. GSH consumption after treating with UMNOCC-PEG nanoparticles (100  $\mu$ g mL<sup>-1</sup>) at pH 6.5.



**Fig. S10.** Time-dependent UV-vis absorption spectra of MB contained UMNOCC-PEG solution at pH 6.5 a) and 7.4 b), respectively.



**Fig. S11.** Hydrogen peroxide generation capability of UMNOCC-PEG with BSA added at pH 7.4 and 6.5 (inset: digital photos, 200  $\mu$ g mL<sup>-1</sup> of UMNOCC-PEG, 1 mM of GSH and BSA in 3 mL solution).



Fig. S12. Absorbance spectrum of the titanium peroxide complex in the presence of UMNOCC-PEG or  $H_2O_2$ .



Fig. S13. The standard curves of NO and measured the UV-vis absorption spectra at 540 nm.



Fig. S14. Rhodamine 123 (RH) UV-vis absorbance spectrum after various treatments.



**Fig. S15.** Fluorescence of L-Tyrosine in the system before and after adding UMNOCC-PEG and illuminated with NIR for 10 min.



Fig. S16. In vivo  $T_1$ -weighted MRI of tumor-bearing mice before and after in situ injection.



Fig. S17. Microscopy images of HeLa cells incubated with UMNOCC-PEG NPs at 37 °C for 0.5 h, and 1 h, respectively. Scale bar: 100  $\mu$ m.



Fig. S18. CLSM images of HeLa cells incubated with UMNOCC-PEG-FITC for 0.5, 1 and 3 h at 37 °C. Scale bar: 50  $\mu$ m.



Fig. S19. Time-dependent cellular uptake of UMNOCC-PEG (50  $\mu$ g mL<sup>-1</sup>) determined by ICP-MS after incubation.



Fig. S20. Time-dependent concentrations of Cu in the major organs measured by ICP-MS.



Fig. S21. Survival curves of different groups of mice after various treatments.



**Fig. S22.** The H&E stained images of heart, liver, spleen, lung and kidney obtained from different groups after 20 days treatment. Scale bar: 50 μm.



**Fig. S23.** The Blood biochemistry and hematology data of female Kunming mice treated with UMNOCC-PEG nanocomposites in different times were as follow: liver function indicators a), BUN b), WBC c), RBC d), HCT e), HGB f), PLT g), MCH h), and MCHC i).