Supporting Information

## Balancing the Thickness of Sensitizing and Inert Layers in Neodymium-Sensitized Tetralayer Nanoconstructs for Optimal Ultraviolet Upconversion and Near-Infrared Cross-Linked Hydrogel Tissue Sealant

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Figure S1. HRTEM images of (a) Core, (b) CS, (c) CSS and (d) CSSS UCNPs.



Figure S2. XRD pattern of Core, CS, CSS and CSSS UCNPs.



Figure S3. Absorbance of CSS UCNPs with different reaction times from 15 min to 60 min with a concentration of 5mg ml<sup>-1</sup> in hexane.



Figure S4. Power dependence measurement of optimal CSSS UCNPs.



Figure S5. Zeta potential distribution of (a) GelMA, (b) UCNPs and (c) GelMA+UCNPs.



Figure S6. Fluorescence spectra of CSSS UCNPs before and after surface modification and digital pictures for (a) CSSS in hexane and (b) CSSS in DI water (power density: 10 W cm<sup>-2</sup>).



Figure S7. (a) Cell viability test under different concentrations of UCNPs. Corresponding live/dead staining of cells with different concentrations from (b) 0, (c) 1, (d) 9, (e) 36, (f) 81, (g) 144, (h) 225, (i) 400  $\mu$ g ml<sup>-1</sup>.



Figure S8. Mutagenesis test by measuring the percentage of micronuclei of control group TCP 2.68% and UCNPs 10.5%.



Figure S9. Degradation curves of GelMA and GelMA+UCNPs.



Figure S10. (a) Sample preparation and (b) measurement equipment for adhesive strength test.