## **Supporting Information**

## An Injectable Hybrid Hydrogel Based on Genetically Engineered Polypeptide for Second Near-infrared Fluorescence/Photoacoustic Imaging-monitored Sustained Chemo-photothermal Therapy

Ruimei Jin,<sup>a</sup> Xiaoquan Yang,<sup>a</sup> Donghui Zhao,<sup>a</sup> Xiaolin Hou,<sup>a</sup> Chaoqing Li,<sup>a</sup> Xianlin Song,<sup>a</sup> Qiong Wang,<sup>c</sup>\* Wei Chen,<sup>a</sup> Yuandi Zhao,<sup>a,b</sup> Bo Liu<sup>a,b</sup>\*

<sup>a</sup> Britton Chance Center for Biomedical Photonics at Wuhan National Laboratory for Optoelectronics – Hubei Bioinformatics & Molecular Imaging Key Laboratory, Department of Biomedical Engineering, College of Life Science and Technology, Huazhong University of Science and Technology, Wuhan 430074, Hubei, P. R. China

<sup>b</sup> Key Laboratory of Biomedical Photonics (HUST), Ministry of Education, Huazhong University of Science and Technology, Wuhan 430074, Hubei, P. R. China

<sup>c</sup> Cancer center, Union Hospital, Tongji Medical College, Huazhong University of Science and Technology, Wuhan 430022, Hubei, P. R. China.



Figure S1. The photograph of self-healing  $PC_{10}A$  hydrogels.



Figure S2. Thermal imaging of  $PC_{10}A/Ag_2S$  QD/PTX hydrogels ( $PC_{10}A$ : 3% w/w, PTX: 200  $\mu$ g mL<sup>-1</sup>) with different concentrations of Ag<sub>2</sub>S QD (0, 0.1, 0.2, 0.4, 0.6, 0.8, and 1 mg mL<sup>-1</sup>) under irradiation with an 808 nm laser for 8 min (2.5 W cm<sup>-2</sup>).



Figure S3. Thermal imaging of  $PC_{10}A/Ag_2S$  QD/PTX hydrogels ( $PC_{10}A$ : 3% w/w,  $Ag_2S$  QD: 1 mg mL<sup>-1</sup>, PTX: 200 µg mL<sup>-1</sup>) with different power densities.



Figure S4. (A)  $PC_{10}A/Ag_2S$  QD/PTX hydrogel ( $PC_{10}A$ : 3% w/w,  $Ag_2S$  QD: 1 mg mL<sup>-1</sup>, PTX: 200 µg mL<sup>-1</sup>) was exposed with an 808 nm laser at a power density of 2.5 W cm<sup>-2</sup> for 8 min (B) and cooling down under room temperature (C).



Figure S5. The cell viability of SKOV3 cells incubation with different concentrations of  $PC_{10}A$  nanogels.



Figure S6. The cell viability of SKOV3 cells incubation with different concentrations of  $PC_{10}A/Ag_2S$  QD ( $PC_{10}A$ : 0.1%; Ag<sub>2</sub>S QD: 1, 0.5, 0.25, 0.125, and 0.063 mg mL<sup>-1</sup>) nanogels.



Figure S7. The cell viability of SKOV3 cells incubated in  $PC_{10}A$  hydrogel and  $PC_{10}A/Ag_2S$  QD hydrogel.



Figure S8. The cell viability of SKOV3 cells suffer from different treatments.



Figure S9. Standard curve of  $PC_{10}A$  nanogel measured with absorption spectrum at 278 nm.



Figure S10. PTX analysis through high performance liquid chromatography (HPLC) at the absorption of 227 nm.



Figure S11. Standard curve of PTX measured by HPLC.



Figure S12. The fluorescence intensity of  $PC_{10}A/Ag_2S$  QD/PTX hydrogels contained different concentrations of  $Ag_2S$  QD (left to right: 20, 40, 60, 80, 100, and 120 µg mL<sup>-1</sup>).



Figure S13. The linear relationship between PA signal intensity and  $PC_{10}A/Ag_2S$  QD ( $PC_{10}A$ : 0.1% w/w, Ag<sub>2</sub>S QD: 1.2, 0.8, 0.4, 0.2, 0.1, and 0.05 mg mL<sup>-1</sup>, PTX: 200 µg mL<sup>-1</sup>).