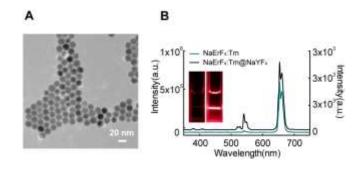
## **Supporting Information**

A Biosynthesized Near-Infrared-Responsive Nanocomposite Biomaterial for Antimicrobial and Antibiofilm Treatment

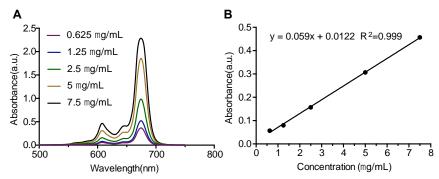
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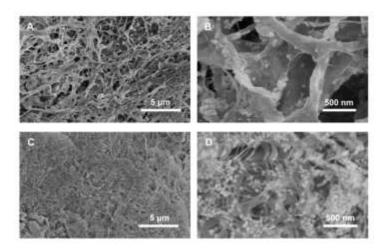
E-mail: biezy@nus.edu.sg, yihanw@shu.edu.cn



**Figure S1.** (A) TEM image of NaErF<sub>4</sub>:Tm(0.5%) UCNPs. (B) Upconversion fluorescence spectra of NaErF<sub>4</sub>:Tm and NaErF<sub>4</sub>:Tm@NaYF<sub>4</sub> UCNPs with 980 nm excitation. Inset: Representative photos showing the luminescence of NaErF<sub>4</sub>:Tm (left) and NaErF<sub>4</sub>:Tm@NaYF<sub>4</sub> (right) UCNPs.

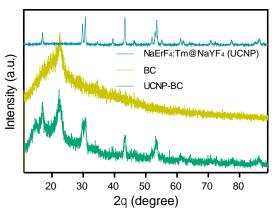


**Figure S2.** The UV-Vis absorption of ZnPc solutions with different concentrations (A). The calibration curve used to calculate the loading capacity of ZnPc to UCNPs (B).



**Figure S3.** SEM images of UCNP@mSiO<sub>2</sub>-BC film prepared by biosynthesis. UCNP@mSiO<sub>2</sub> were supplemented on the third day (A-B) or on the fourth day (C-D) to

the K. xylinus culture.



**Figure S4.** XRD patterns of UCNPs, unmodified BC film, and UCNP-BC film. UCNP-BC film exhibits X-ray diffraction peaks corresponding to UCNPs, suggesting the successful incorporation of UCNPs into the BC film.

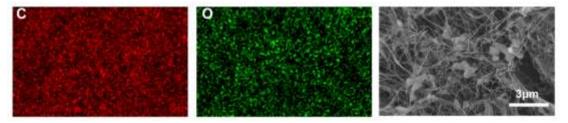
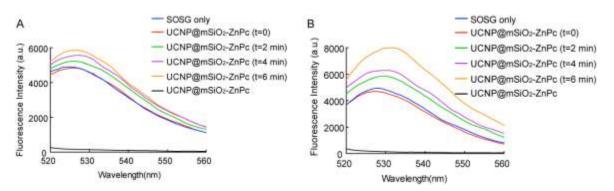
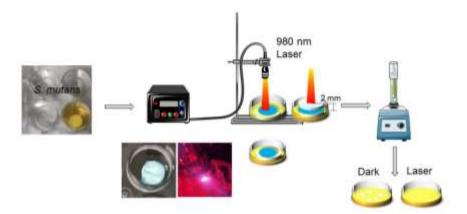


Figure S5. SEM elemental mapping analysis of pure BC film.

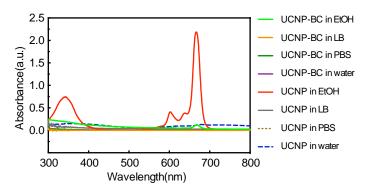


**Figure S6.** Singlet oxygen sensor green (SOSG) probe was used to monitor the singlet oxygen production of UCNPs and UCNP-BS materials. SOSG solution (2  $\mu$ M) was mixed with UCNP@mSiO<sub>2</sub>-ZnPc (200  $\mu$ g/mL) (A) or UCNP@mSiO<sub>2</sub>-ZnPC-BC (1 cm x 1 cm) (B), and irradiated by 980 nm laser light (2.72 W/cm<sup>2</sup>) for 6 min. Every 2 minutes, fluorescence of the solution was measured when excited by 504 nm light. Control groups

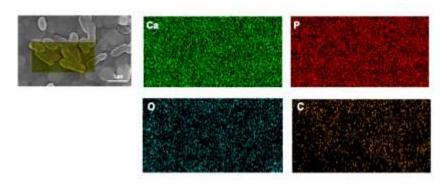
of SOSG only or nanomaterials only were included. The increase of the fluorescence at 530 nm confirmed the production of singlet oxygen.



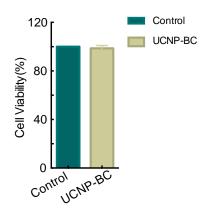
**Figure S7.** Scheme of the photodynamic antimicrobial treatment on the *in vitro* oral biofilm model.<sup>[1]</sup>



**Figure S8.** UCNP-BC nanocomposite film and UCNP nanoparticles were immersed in ethanol, LB media, PBS buffer, or water for 24 hours separately. The supernatant was collected and analyzed by UV-Vis spectrometer. No significant leakage of UCNPs from UCNP-BC film was observed in LB media, PBS buffer, or water.



**Figure S9.** SEM elemental mapping analysis of samples on HA disks after photodynamic antibacterial treatment using UCNP-BC film. No UCNP related elements were detected.



**Figure S10.** Biocompatibility assay of the biosynthesized UCNP-BC nanocomposite film. Compared to the control group, the addition of UCNP-BC film did not harm the cell viability. Results were measured and averaged from three biological replicates.

## References

[1] smart.servier.com