## Supplementary Information

## Releasable antimicrobial polymer-silk coatings for combating multidrugresistant bacteria

Erna Wulandari,<sup>a</sup> Rachel Budhisatria,<sup>b</sup> Alexander H. Soeriyadi,<sup>b</sup> Mark Willcox,<sup>c</sup> Cyrille Boyer,\*<sup>a</sup> and Edgar H. H. Wong\*<sup>a</sup>

<sup>a</sup> Australian Centre for NanoMedicine (ACN), School of Chemical Engineering, University of New South Wales (UNSW), Sydney, NSW 2052, Australia. Emails: <u>edgar.wong@unsw.edu.au</u>; <u>cboyer@unsw.edu.au</u>

<sup>b</sup> Mochtar Riady Institute of Nanotechnology (MRIN), Banten 15810, Indonesia.

<sup>c</sup> School of Optometry and Vision Science, University of New South Wales (UNSW), Sydney NSW 2052, Australia.



Figure S1. <sup>1</sup>H NMR spectra of Boc-protected P1 in *d*-DMSO.



Figure S2.DMAc GPC-differential refractive index (DRI) chromatogram of the Boc-protected P1.



Figure S3. Evaluation of the formation of P1 films on glass coverslips with and without the presence of silk. (a) Silk-P1 and (b) silkless water-P1 mixtures immediately after casting on coverslips; (c) silk-P1 film (S<sub>1</sub>P1<sub>1.6</sub>) formed on glass coverslip after annealing; and (d) silkless water-P1 sample after annealing showing no stable film formation on glass coverslip.



Figure S4. Calibration curve of fluorescein labeled P1 for quantification of P1 release from  $S_{1}\text{P1}_{1.6}$ 



Figure S5.Nanolive images of *P. aeruginosa* PAO1 biofilm mass formed on S<sub>1</sub>P1<sub>1.6</sub>-coated glass coverslips.



Figure S6. Nanolive images of *P. aeruginosa* PAO1 biofilm mass formed on uncoated glass coverslips.



Figure S7. Nanolive images of P. aeruginosa PAO1 biofilm mass formed on silk-coated glass coverslip.



Figure S8. Antibacterial activity of S<sub>1</sub>P1<sub>1.6</sub> film against *P. aeruginosa* PAO1 (a) planktonic and (b) biofilm cells as determined by colony forming unit (CFU) analysis after 24 h of incubation. Data are representative of at least three independent experiments ± SD.



Figure S9. Zeta potential values of antimicrobial polymer P1 in water and the supernatants of 1 wt% silk (S<sub>1</sub>), S<sub>1</sub>P1<sub>1.6</sub>, 4 wt% silk (S<sub>4</sub>), S<sub>4</sub>P1<sub>1.6</sub>, and S<sub>1</sub>P1<sub>1.6</sub> with blank silk topcoat (S<sub>1</sub>P1<sub>1.6</sub>-S<sub>1</sub>) film after 6.5 hours of incubation in water. Data are representative of at least three independent experiments  $\pm$  SD.



Figure S10. (a) Schematic illustration of antibacterial activity experiment using 0.4 mg of P1 released from of  $S_1P1_{1.6}$  film. Antibacterial activity of pure P1 in solution (denoted as P1 (0.4 mg)) and 0.4 mg of P1 released from of  $S_1P1_{1.6}$  film (denoted as  $S_1P1_{1.6}$  (0.4 mg release)) against PAO1 as determined by CFU analysis after (b) 1 h and (c) 2 h of incubation. Data are representative of at least three independent experiments ± SD.



Figure S11. Comparison of uncoated and  $S_1P1_{1.6}$  film-coated cotton gauze. (a) Uncoated and (b)  $S_1P1_{1.6}$  film-coated cotton gauze with less frayed fibers in dry state; (c) uncoated cotton gauze after soaking in water for 1 minute; and (d)  $S_1P1_{1.6}$  film-coated cotton gauze after soaking in water for 1 minute showing the invisible film coating can maintain the gauze structure.



Figure S12.<sup>1</sup>H NMR spectra of P2 in D<sub>2</sub>O.



Figure S13. Calibration curve of fluorescein labeled P2 for quantification of P2 release from  $S_1P2_{1.6}$ .