

**Supporting Information****Silk Macromolecules with Amino Acid-Poly(Ethylene Glycol) Grafts for Controlling Layer-by-Layer Encapsulation and Aggregation of Recombinant Bacterial Cells**

*Irina Drachuk<sup>a</sup>, Rossella Calabrese<sup>b</sup>, Svetlana Harbaugh<sup>c</sup>, Nancy Kelley-Loughnane<sup>c</sup>, David L. Kaplan<sup>b</sup>, Morley Stone<sup>c</sup>, Vladimir V. Tsukruk<sup>a,\*</sup>*

<sup>a</sup> School of Materials Science and Engineering, Georgia Institute of Technology, Atlanta, Georgia 30332 United States

<sup>b</sup> Department of Biomedical Engineering, Tufts University, Medford, Massachusetts 02155 United States

<sup>c</sup> Air Force Research Laboratory, Directorate of Human Effectiveness, Wright-Patterson AFB, Dayton, Ohio 45433 United States

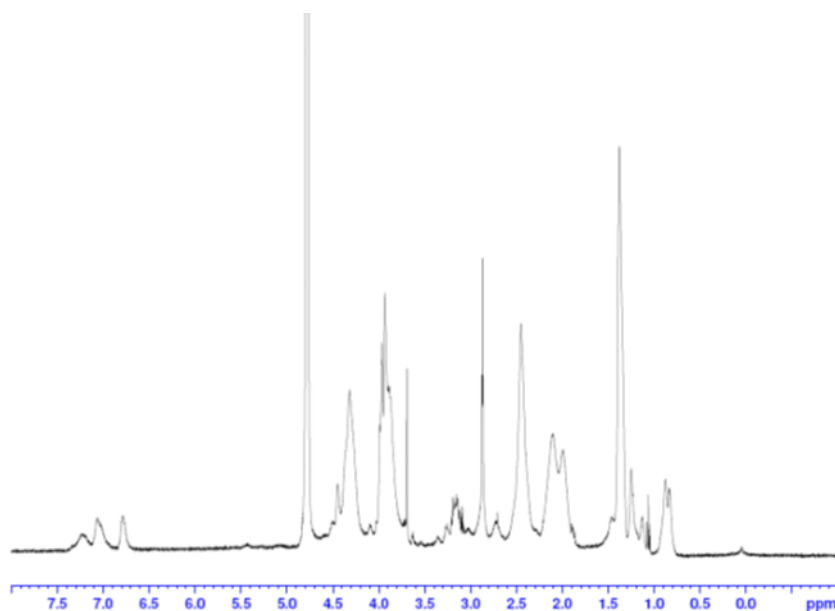


Figure S1. <sup>1</sup>H NMR spectrum of low charge density SF(S)-PGA silk ionomer.

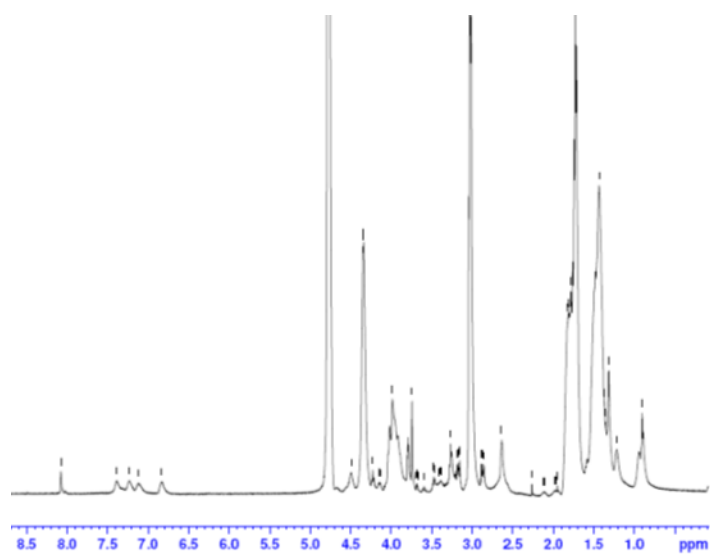


Figure S2:  $^1\text{H}$ NMR spectrum of low charge density SF(S)-PLL silk ionomer.

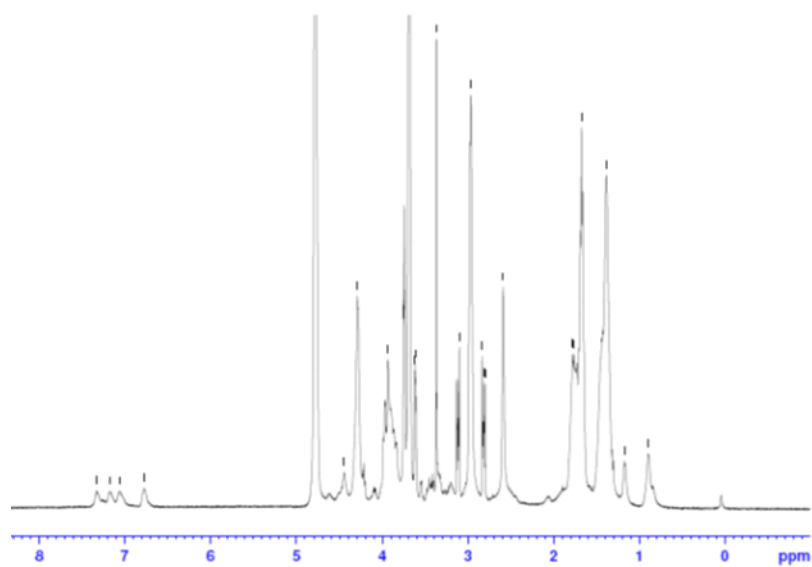


Figure S3.  $^1\text{H}$ NMR spectrum of block SF(S)-PLL-PEG silk copolymer

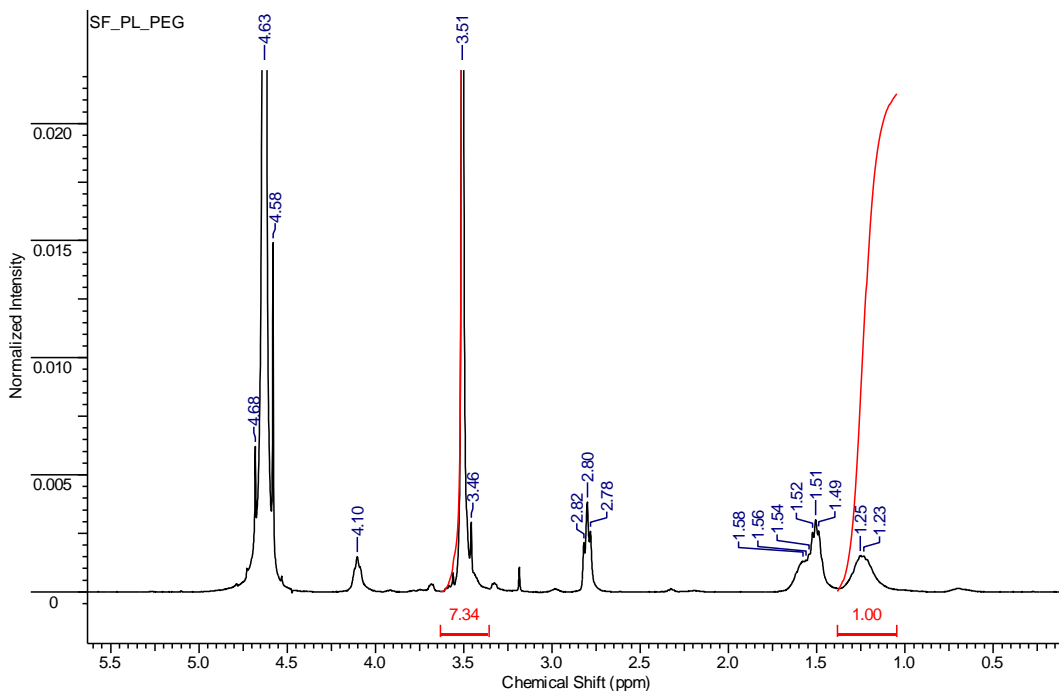


Figure S4.  $^1\text{H}$ NMR spectrum of double-brush PEG-grafted SF(Y,S)-PLL-g[D]-PEG silk ionomer with  $D=9$ .

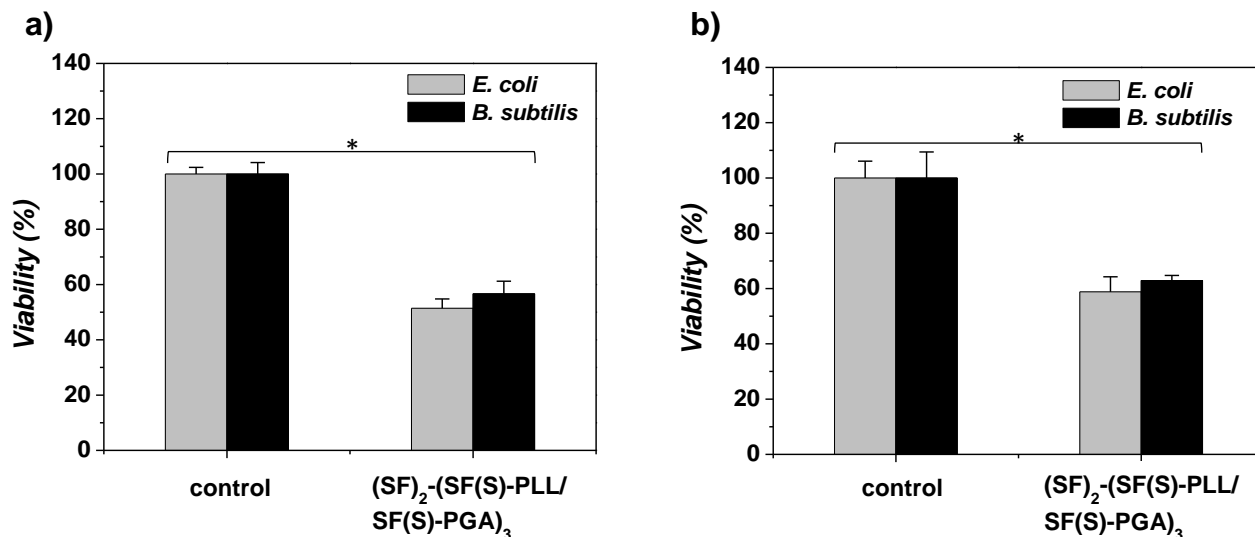


Figure S5. Viability of bacterial cells encapsulated in 3 bilayers of low charge density silk polyelectrolytes that were initially primed with a bilayer of SF,  $(\text{SF})_2$ -(SF(S)-PLL/SF(S)-PGA)<sub>3</sub> shells. Viability was assessed with resazurin (a) and MTT (b) assays. Experimental values are represented as means  $\pm$  SD,  $n = 6$ . Paired  $t$  test,  $*p < 0.01$ .

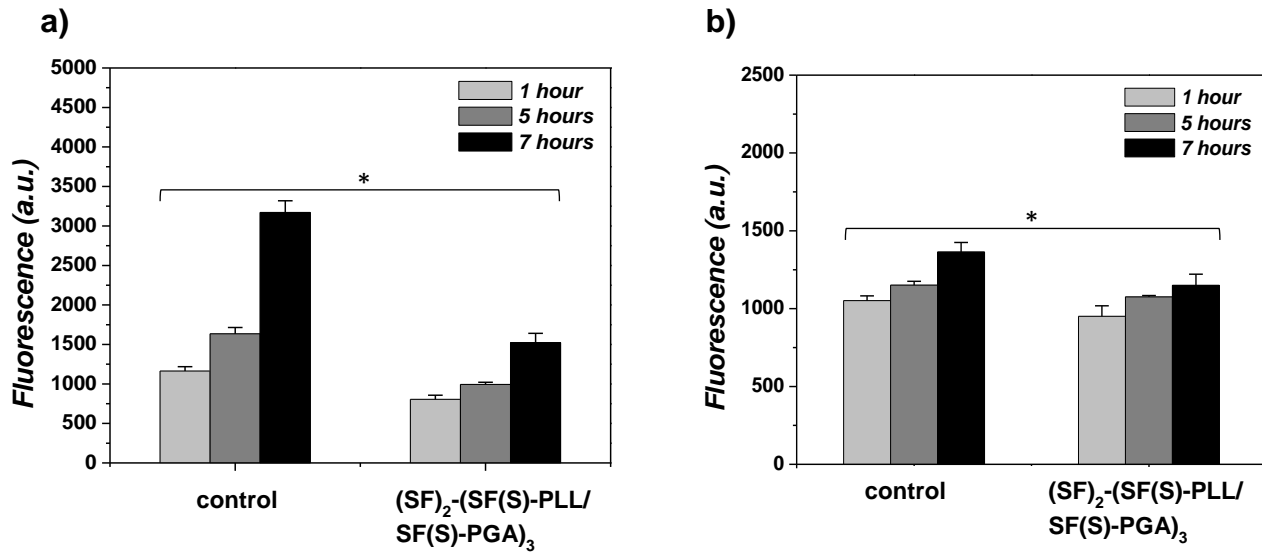


Figure S6. Kinetics of GFP expression in *E. coli* (a) and *B. subtilis* (b) cells encapsulated in 3 bilayers of low charge density silk polyelectrolytes that were initially primed with a bilayer of SF, (SF)<sub>2</sub>-(SF(S)-PLL/SF(S)-PGA)<sub>3</sub> shells. Experimental values are represented as means  $\pm$  SD,  $n = 6$ . Paired  $t$  test,  $*p < 0.05$ .