Supporting Information

Mussel-Inspired Multifunctional Hydrogel Coating for Prevention of Infections and Enhanced Osteogenesis

Hao Cheng,[a,b,c,#] *Kan Yue,*[a,b,#] *Mehdi Kazemzadeh-Narbat,*[a,b,#] *Yanhui Liu,*[a,b,d] *Akbar Khalilpour,*[a,b] *Bingyun Li,*[e] *Yu Shrike Zhang,*[a,b] *Nasim Annabi,*[a,b,f,*] *Ali Khademhosseini*[a,b,g,h,*]

[a] Biomaterials Innovation Research Center, Division of Biomedical Engineering, Department of Medicine, Brigham and Women's Hospital, Harvard Medical School, Cambridge, MA, 02139, USA

[b] Harvard-MIT Division of Health Sciences and Technology, Massachusetts Institute of Technology, Cambridge, MA, 02139, USA

[c] Othopeadic Department, Tongji Hospital, Tongji Medical College, Huazhong University of Science and Technology, Wuhan 430030, China

[d] College of Textiles, Donghua University, Shanghai, 201620, China

[e] Department of Orthopaedics, School of Medicine, West Virginia University, Morgantown, WV 26506, USA

[f] Department of Chemical Engineering, Northeastern University, Boston, MA, 02115-5000, USA

[g] Department of Bioindustrial Technologies, College of Animal Bioscience and Technology, Konkuk University, Seoul, 143-701, the Republic of Korea

[h] Department of Physics, King Abdulaziz University, Jeddah, 21569, Saudi Arabia

[#] These authors contributed equally to this work.

[*] Corresponding authors: Prof. Ali Khademhosseini (alik@bwh.harvard.edu), Prof. Nasim Annabi (n.annabi@neu.edu)

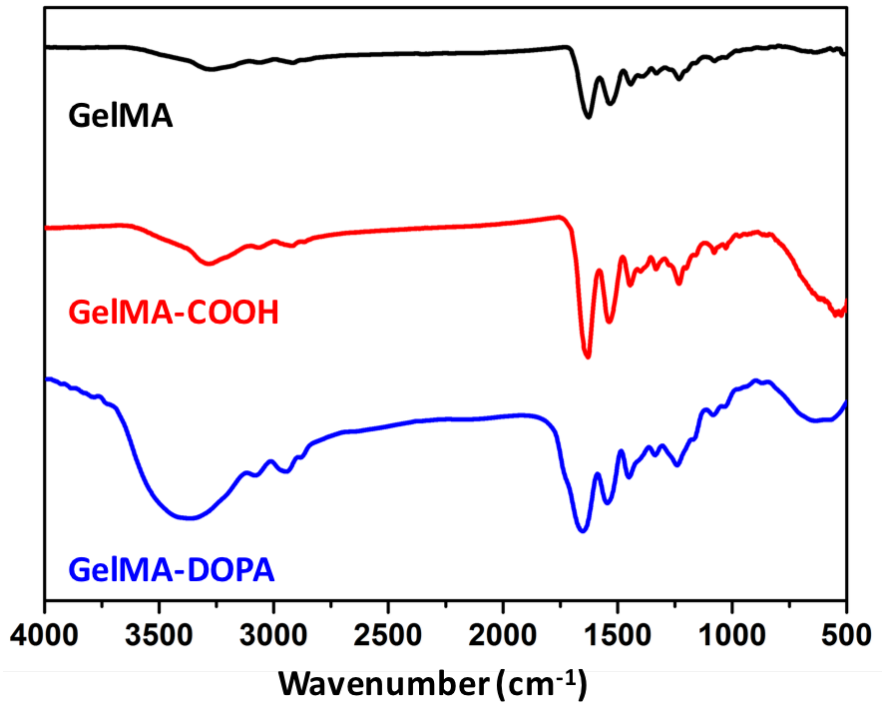

Figure S1. FT-IR spectra of GelMA (black curve), GelMA-COOH (red curve), and GelMA-DOPA (blue curve).

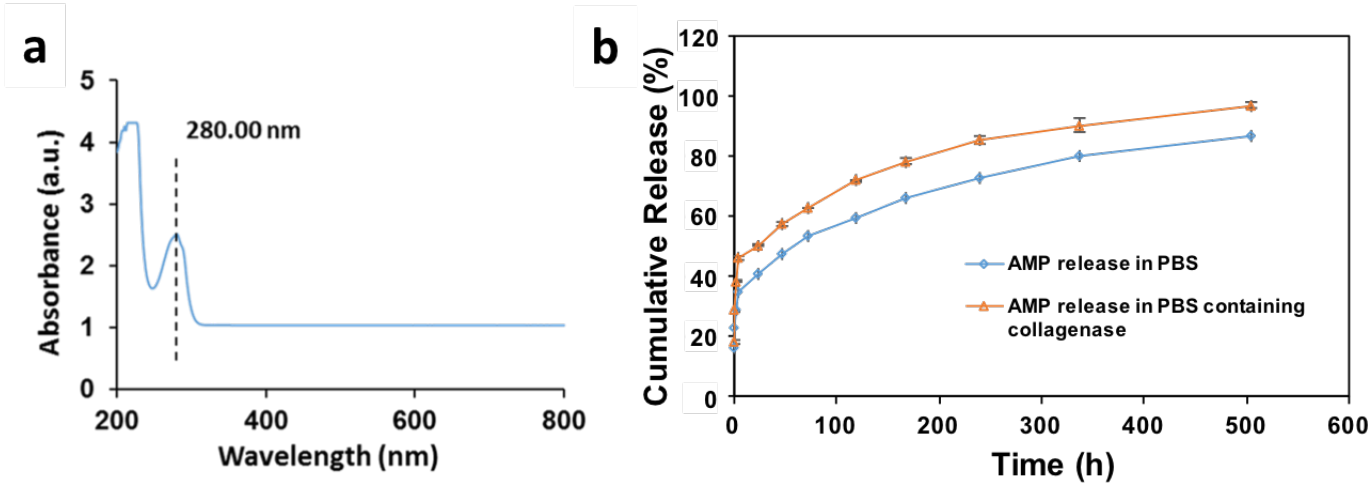

Figure S2. *In vitro* AMP release from 20% (w/v) GelMA-DOPA-AMP-SN hydrogels. **(a)** UV-Vis spectrum of AMP in PBS. Absorbance at 280 nm was monitored to determine the released amount of AMP from hydrogel matrix. **(b)** Cumulative release kinetics of AMP released from the GelMA-DOPA AMP-SN composite hydrogels coated on Ti substrates immersed in PBS or PBS containing 2U/mL collagenase in 21 days.

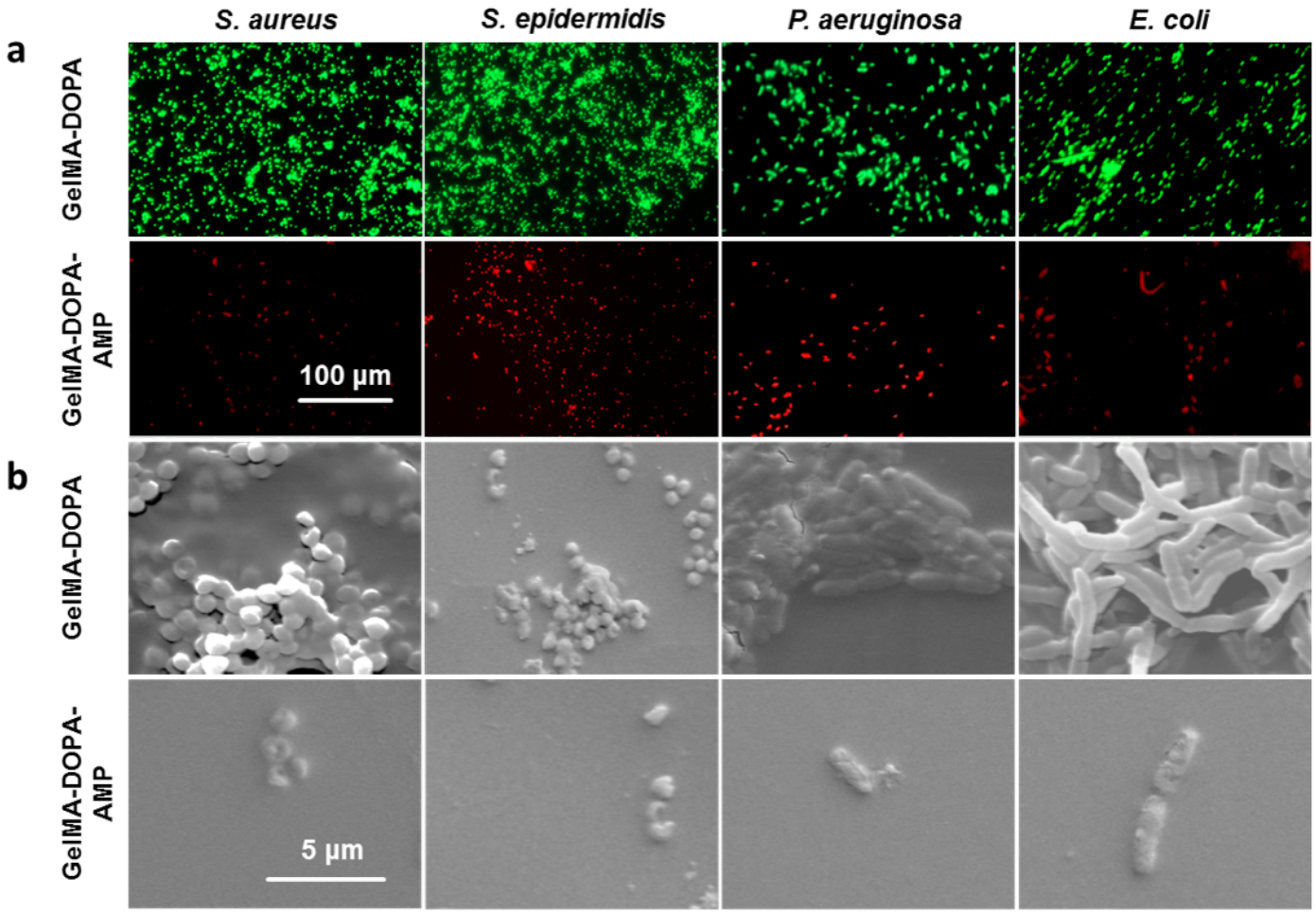

Figure S3. Antimicrobial activity of GelMA-DOPA hydrogels at 20% (w/v) prepolymer concentration with different additives. **(a)** AMP released from GelMA-DOPA-AMP samples demonstrated efficient antimicrobial ability to kill the tested bacteria, *S. aureus*, *S. epidermidis*, *P. aeruginosa*, and *E. coli*. Compared to GelMA-DOPA samples without AMP loading, no live bacteria were observed on the GelMA-DOPA-AMP samples after 24 h culture (green: live bacteria; red: dead bacteria; scale bar: 100 μm). **(b)** SEM images of surfaces of GelMA-DOPA and GelMA-DOPA-AMP hydrogels incubated overnight with *S. aureus*, *S. epidermidis*, *P. aeruginosa*, and *E. coli*. Very few bacteria were observed on the AMP-loaded samples (scale bar: 5 μm).

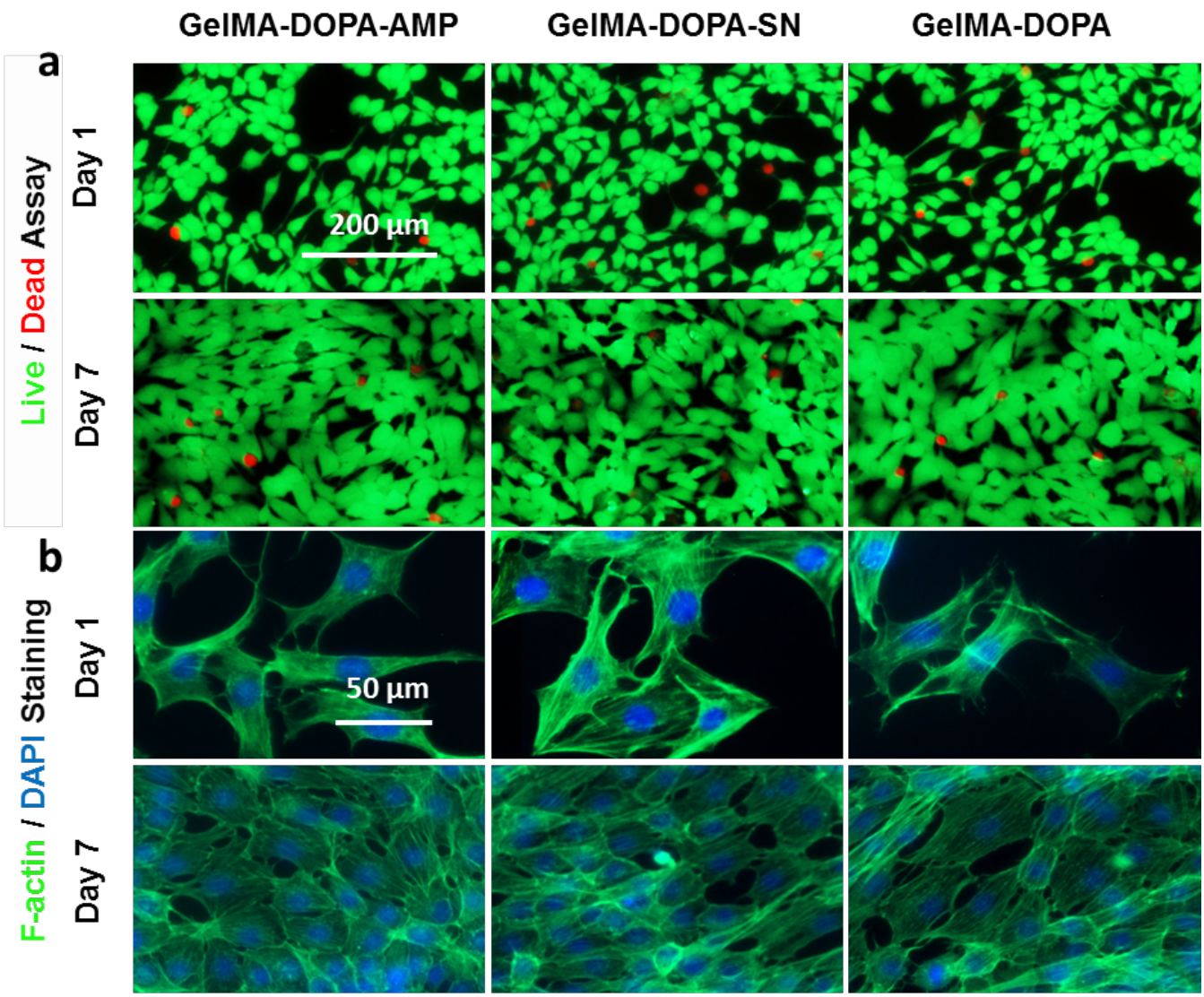

Figure S4. Cytotoxicity and cellular evaluation of different hydrogel formulations at 20% (w/v) prepolymer concentration. **(a)** Images of hMSCs seeded on the surface of GelMA-DOPA-AMP, GelMA-DOPA-SN and GelMA-DOPA hydrogels with Live/Dead staining on days 1 and 7 (scale bar: 200 µm). **(b)** Images of hMSCs seeded on the surface of GelMA-DOPA-AMP, GelMA-DOPA-SN, and GelMA-DOPA hydrogels with staining for f-actin/cell nuclei on days 1 and 7 (scale bar: 50 µm).

Table S1. Primer sequences for qPCR experiments.

Genes	***Forward***	***Reverse***
Osteocalcin	ATGAGAGCCCTCACACTCCTCG	GTCAGCCAACTCGTCACAGTCC
Osteopontin	TTCCAAGTAAGTCCAACGAAAG	GTGACCAGTTCATCAGATTCAT
ALP	GAGTATGAGAGTGACGAGAA	AGTGGGAGTGCTTGTATC
RUNX2	CGGAATGCCTCTGCTGTTAT	ACTCTTGCCTCGTCCACTCC