Supporting Information for

3D Bioprinting of carbohydrazide-modified gelatin into microparticle-suspended oxidized alginate for fabrication of complex-shaped tissue constructs

Dong Nyoung Heo,^{a,b,c} Mecit Altan Alioglu,^{a,b} Yang Wu,^{a,b,d} Veli

Ozbolat,^e Bugra Ayan,^{a,b} Madhuri Dey,^{b,f} Youngnam Kang,^{a,b} Ibrahim

T. Ozbolat*^{a,b,g,h,i}

^a Department of Engineering Science and Mechanics Department, The Pennsylvania State University, University Park, PA 16802, USA

^b The Huck Institutes of the Life Sciences, The Pennsylvania State University, University Park, PA 16802, USA

^c Department of Dental Materials, School of Dentistry, Kyung Hee University, 26 Kyungheedae-ro, Dongdaemun-gu, Seoul 02447, Republic of Korea

^d School of Mechanical Engineering and Automation, Harbin Institute of Technology, Shenzhen 518055, China

^e Department of Mechanical Engineering, Ceyhan Engineering Faculty, Cukurova University, Adana 01950, Turkey

 $^{\rm f}$ Chemistry Department, Penn State University, University Park, PA 16802, USA

^g Biomedical Engineering Department, Pennsylvania State University, University Park, PA, 16802, USA

^h Materials Research Institute, Pennsylvania State University, University Park, PA, 16802, USA

ⁱ Neurosurgery Department, College of Medicine, Pennsylvania State University, Hershey, PA, 17033, USA

* Correspondence author

E-mail: itol@psu.edu (Ibrahim T. Ozbolat). Tel.: 1-814-863-5819

Video Captions:

Video S1: Mixing and casting of OAlg and Gel-CDH hydrogels into 3D printed PDMS molds for fabrication of samples for the compression test. A5G5, A4G6, A3G7 and A2G8 solutions were loaded into mixer nozzles (McMaster-Carr, GA) and extruded into PDMS molds with an inner diameter of 7.5 mm and a height of 15 mm which were 3D printed using an INKREDIBLE bioprinter (Cellink, Sweden).

Video S2: Compression testing of samples made of different OAlg and Gel-CDH combinations. Video shows the mechanical properties of composite hydrogels which were determined at room temperature using a compression tester (Instron 5966, Instron, MA) equipped with a 1-kN load-cell at a compression speed of 50 mm/min. The specimens were compressed to 30% strain. Young's modulus was calculated form the stress-strain curve. Five samples (n=5) were used for each group.

Video S3: Crosslinking of OAlg and Gel-CDH via Schiff's base reaction. OAlg and Gel-CDH were crosslinked via Schiff base reaction to form the imine bond between aldehyde groups of OAlg and amino groups of Gel-CDH.

Video S4: 3D Bioprinting of a meshed tube construct with preheated Gel-CDH Ink and slurry bath consisting of OAlg and crosslinked gelation microparticles. Gel-CDH was preheated to 37 °C and extruded into the slurry bath composed of gelatin microparticles and OAlg solution. Gel-CDH and OAlg were then rapidly crosslinked to form filaments.

Video S5: A 3D bioprinted meshed tube construct removed from the slurry bath. Meshed tube construct maintained its structural integrity without any collapse or filament decomposition after pipetting of PBS indicating the remarkable mechanical stability of the printed constructs.

Video S6: A 3D bioprinted "ball-in-a-cage" construct removed from the slurry bath. "ball-in-a-cage" construct was 3D printed, and the ball (red color) was trapped and able to freely rotate within the enclosing mesh cage (green color).