

## ***Supporting Information***

### The preparation of a novel regenerated silk fibroin-based hydrogel for extrusion bioprinting

Ni Chen<sup>a</sup>, Xinbo Zhang<sup>b</sup>, Jinyang Lyu<sup>c</sup>, Guanglei Zhao<sup>c</sup>, Kai Gu<sup>a</sup>, Jun Xia<sup>c</sup>, Zhongchun  
Chen<sup>d</sup>, Zhengzhong Shao<sup>a\*</sup>

\*: Corresponding author. E-mail: zzshao@fudan.edu.cn

a: State Key Laboratory of Molecular Engineering of Polymers, Laboratory of  
Advanced Materials and Department of Macromolecular Science, Fudan  
University, Shanghai 200433, P.R.China.

b: Department of Materials Science and Engineering, Southern University of  
Science and Technology, Shenzhen, Guangdong 518055, P.R.China.

c: Department of Orthopedic Surgery, Huashan Hospital, Fudan University,  
Shanghai 200040, P.R.China.

d: Department of Otorhinolaryngology-Head and Neck Surgery, Huashan  
Hospital, Fudan University, Shanghai 200040, P.R.China.

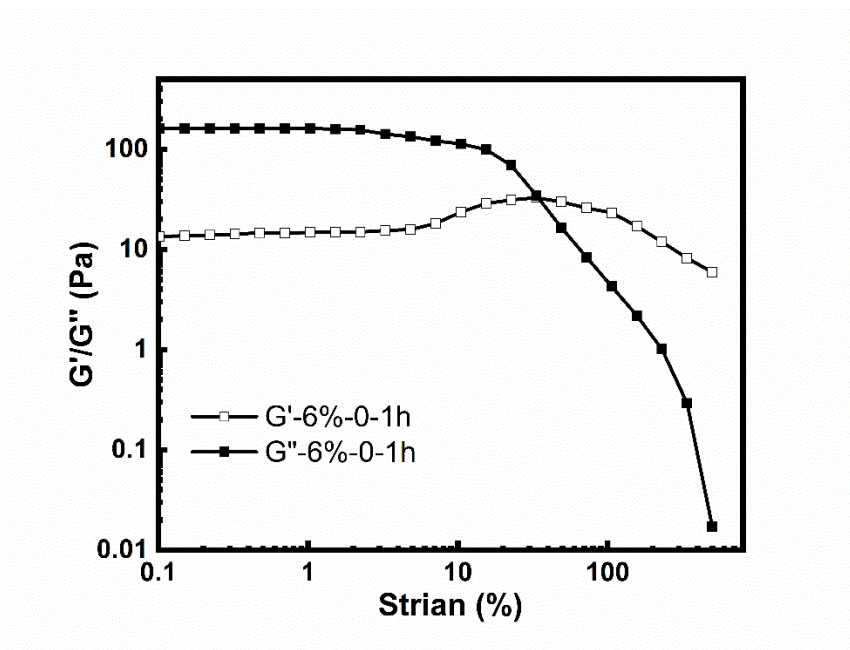


Figure S1. Strain sweep of the 6%-0-1h HMWRSF-based precure hydrogel with the lowest G' modulus value among all precure hydrogels.

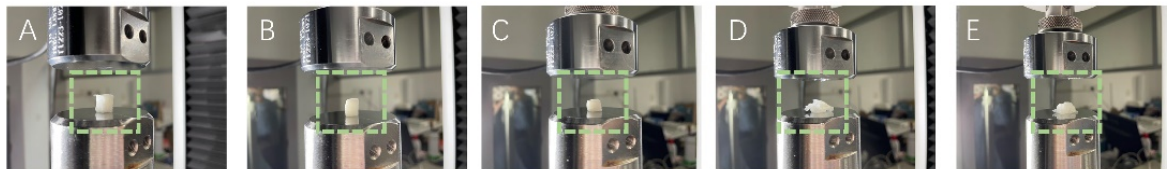


Figure S2. Digital images of 3D-printed RSF/HPMC scaffolds after experiencing 50% compressive strain. A.r-6%-0-2h, B.r-8%-0.5-1h, C.r-10%-1.0-1h, D.r-L-8%-0-1h, E. r-L-10%-0-1h.

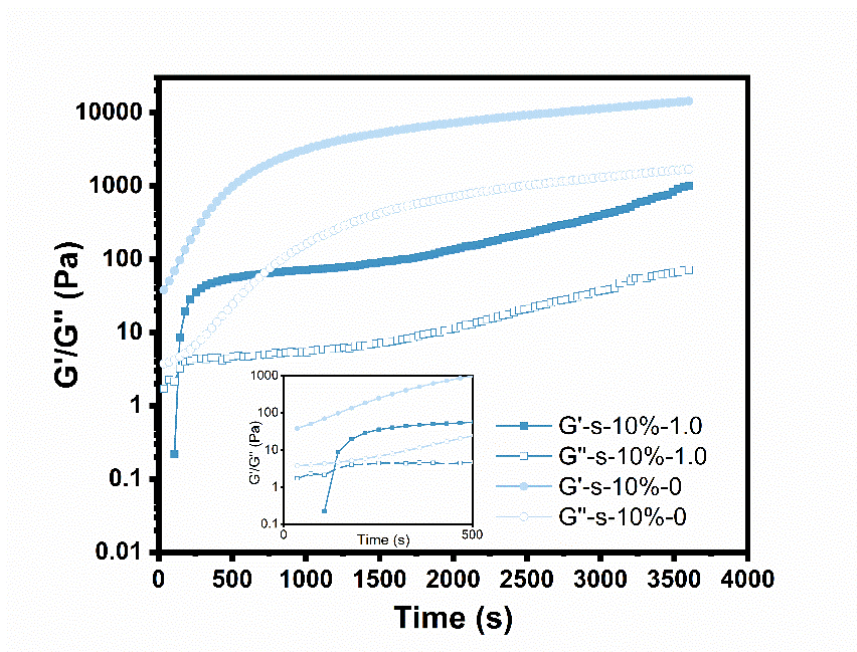


Figure S3. Evaluation of  $G'$  and  $G''$  in gelation process of the s-10%-1.0 and s-10%-0 upon heating at 70°C. inset: first 500s of gelation process.

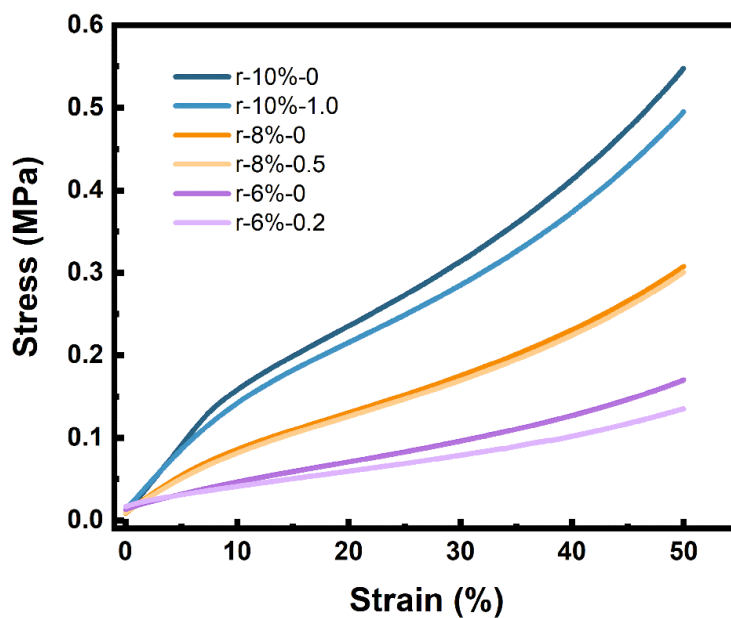


Figure S4. Stress-strain curves of RSF/HPMC hydrogels of different solid content and urea content.

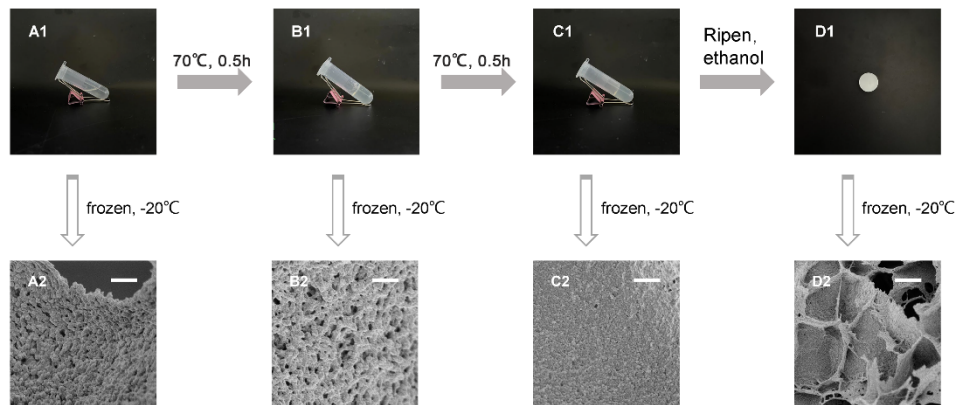


Figure S5. Digital images of 8% -0.5 HMWRSF/HPMC-urea aqueous solution and its produced hydrogels at different stages (A1-D1). SEM images of the cross section of those corresponded samples after lyophilization (A2-D2). 8% means the solid content in the solution and 0.5 means the weight ratio of urea to HMWRSF. The weight ratio of HMWRSF to HPMC is 9:1. Scale bar is 500 nm.

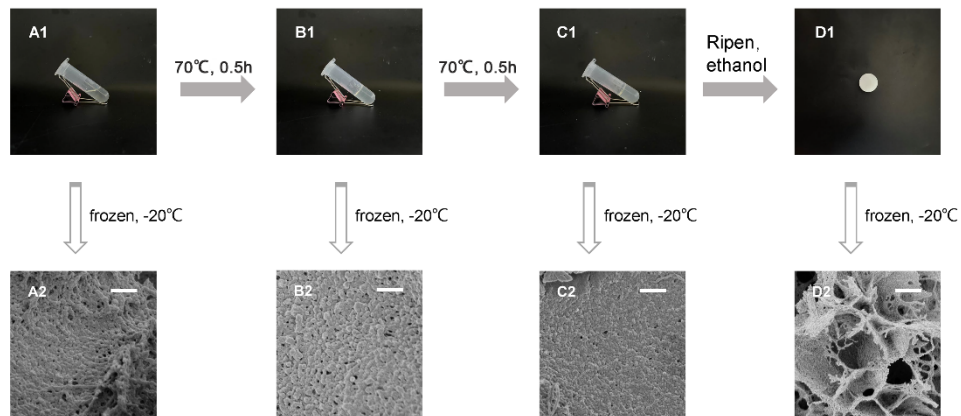


Figure S6. Digital images of 10% -1.5 HMWRSF/HPMC-urea aqueous solution and its produced hydrogels at different stages (A1-D1). SEM images of the cross section of those corresponded samples after lyophilization (A2-D2). 10 % means the solid content in the solution and 1.0 means the weight ratio of urea to HMWRSF. The weight ratio of HMWRSF to HPMC is 9:1. Scale bar is 500 nm.

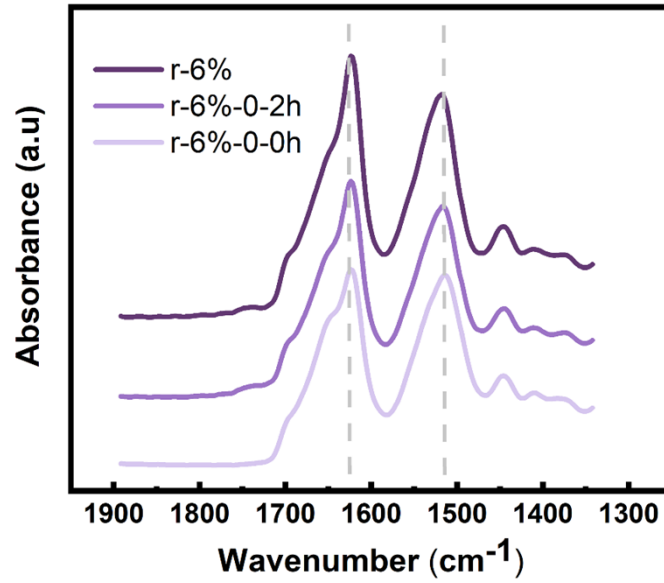


Figure S7. ATR-FTIR characterization of the samples lyophilized from HMWRSF/HPMC solution (r-6%-0-0h), precure HMWRSF/HPMC hydrogel (r-6%-0-2h), and ripened HMWRSF/HPMC hydrogel (r-6%).

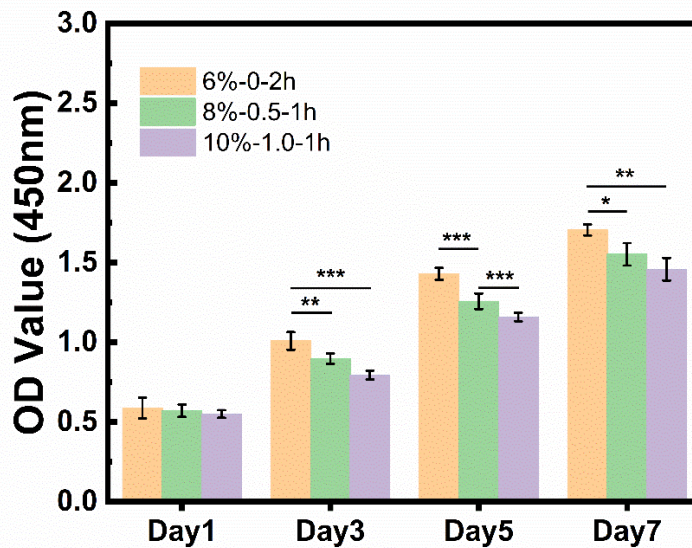


Figure S8. The proliferation of dental pulp mesenchymal stem cells (DPSCs) on 3D-biprinted HMWRSF-based scaffolds evaluated by cell counting kit-8 (CCK-8).

Table S1. Solid content and compression modulus of different RSF/HPMC hydrogels (n=5).  $W_a$ : the actual solid content.

Samples	$W_a$ (wt%)	Compressive modulus (MPa)
r-10%-0	12.7±0.2	1.6±0.2
r-10%-1.0	11.9±0.3	1.5±0.2
r-8%-0	9.7±0.2	0.95±0.02
r-8%-0.5	9.3±0.3	0.84±0.08
r-6%-0	7.2±0.2	0.35±0.03
r-6%-0.2	6.8±0.2	0.28±0.05