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**Fig. S1. Cell vitality in bPU, tPU3, and tPU4 dispersion.** The bPU, tPU3, and tPU4 dispersion were diluted to 25 wt% by culture medium. The number of healthy/unhealthy/dead cells in bPU, tPU3, and tPU4 dispersion was measured by staining with VB-48, AO, and PI after an hour. The number of healthy cells for the bPU, tPU3, and tPU4 were 62.0%, 85.7%, and 71.7%, respectively. The number of healthy cells in tPU3 and tPU4 was lower than that in tPU1 and tPU2 (Figure 1B) but was higher than bPU.



**Fig. S2. Cell vitality in tPU2/gelatin and tPU2/agarose.** The number of healthy/unhealthy/dead cells in tPU2/gelatin and tPU2/agarose was measured by staining with VB-48, AO, and PI after an hour. The number of healthy cells in tPU2/gelatin and tPU2/agarose was 97% and 95%, respectively.



**Fig. S3. The self-healing property of PUC1, PUC2, and PUC4 hydrogels.** (A) Gross observation of self-healing (fluidity) through sealing of the central holes in the PUC1, PUC2, and PUC4 hydrogels. The hydrogel was stained with trypan blue for easy observation. Hole sealing was expressed as the change of diameter of the hole vs. time (Fig. 6A). (B) The stretching experiment of PUC1, PUC2, and PUC4 hydrogels was conducted foe self-healed PUC1, PUC2, and PUC4 hydrogels Two semicircular pieces of the hydrogel with and without trypan blue were placed in contact for a period of time. Two sides of the healed PUC1, PUC2, and PUC4 hydrogels were stretched with tweezers and observed for the rupture site after stretching.



**Fig. S4. Rheological properties of PUC1, PUC2, and PUC4 hydrogels.** (A) The moduli (G' and G") of the PUC1, PUC2, and PUC4 hydrogels during gelling at 25°C, measured at 1 Hz and 1% strain. Heating to 37 °C (indicated by the long box) did not change the values of G' and G". (B) Strain sweep of the equilibrated PUC1, PUC2, and PUC4 hydrogels, measured at 1 Hz at 37°C. (C) The damage-healing cycles of the PUC1, PUC2, and PUC4 hydrogels were measured at 37°C (1 Hz) between the alternative strains of 1% and 230%. (D) Steady shear viscosities of PUC1, PUC2, and PUC4 hydrogels were measured at 37°C with a 0.1-100 shear rate.



Fig. S5. The image of 3D printed PUC3 filaments under the optical microscope. The PUC3 hydrogel was printed through the 160  $\mu$ m nozzle using the 3D printer. The width of one filament was 243 ± 47  $\mu$ m. The exudate swelling was observed.



Fig. S6. The degradation rates of tPU2 and PUC3 hydrogels. The degradation of hydrogels was measured in 37°C PBS. The tPU and PUC3 hydrogels lost ~6 wt% and ~23 wt% after 7 days, respectively. # denotes the p value < 0.05 among the indicated groups.



Fig. S7. The biodegradation rates of tPU2 and PUC3 constructs containing NSCs. Biodegradation of constructs was measured at 37°C in a period of 7 days. The tPU and PUC3 constructs lost ~10 wt% and ~35 wt% after 7 days, respectively. # denotes the *p* value < 0.05 among the indicated groups.



**Fig. S8. Rheological properties of tPU2-gelatin hydrogel.** (A) The moduli (G' and G") of the tPU2-gelatin hydrogel crosslinking with  $Ca^{2+}$ , measured at 37°C, 1 Hz, and 1% strain.