Supporting information

Self-recovering dual cross-linked hydrogels based on bioorthogonal

click chemistry and ionic interactions

Henan Zhan, Shanshan Jiang, Anika M. Jonker, Imke A. B. Pijpers, Dennis W. P. M. Löwik^{*}



Fig. S1 Oscillatory strain sweep measurements of different dual cross-linked hydrogel compositions (normalization to a strain of 0.1%).



Fig. S2 Storage moduli (G' value) of CC (100c), PC (100p) and DC (xxp/yyc) hydrogels after two times of mechanical rupture. (n = 3; * p<0.033, ** p<0.002, ***p<0.001, one-way Anova)



Fig. S3 Swelling ratios of pure chemical, physical and DC hydrogels at 1 h, 3 h, 7 h and 24 h. (n = 3; * p<0.033, ** p<0.002, one-way Anova)



Fig.S4 Dry mass of PC and DC hydrogels at 1 h and 24 h.



Fig.S5 Cryo-SEM images of star-PEG-DBCO/star-PEG-N₃ (20 mg/mL, left) and star-PEG-AA/Ca²⁺ (30 mg/mL, right) cross-linked hydrogel systems. The scale bars represent 10 μ m.



Fig. S6 Mesh size distribution of hydrogels.



Fig. S7 Cryo-SEM images of the 100% physically cross-linked hydrogels at different polymer concentration. The scale bars represent 10 μ m.



Fig. S8 Cryo-SEM images of the 100% physically cross-linked hydrogel systems at different Ca^{2+} concentration (1 eq polymer combined with 10 eq, 15 eq, 20 eq, 30 eq or 40 eq Ca^{2+}). The scale bars represent 10 μ m.



Fig. S9 Confocal microscopy images of cell morphology after 5-day culture inside different hydrogel systems.



Fig. S10 MALDI-TOF spectra of star-PEG-alendronic acid.



Fig. S11 ³¹P NMR spectra of star-PEG-alendronic acid.



Figure S12. MALDI-TOF spectra of star-PEG-DBCO.



Fig. S13 ¹H NMR spectra of star-PEG-DBCO.



Fig. S14 Mass spectrometry spectra of azido-RGDS.



Fig. S15 MALDI-TOF spectras of (A) star-PEG-N₃, (B) star-PEG-RGDS-N₃ and (C) star-PEG-CF-N₃.



Fig. S16 1 H NMR spectras of (A) star-PEG-N₃, (B) star-PEG-RGDS-N₃ and (C) star-PEG-CF-N₃.