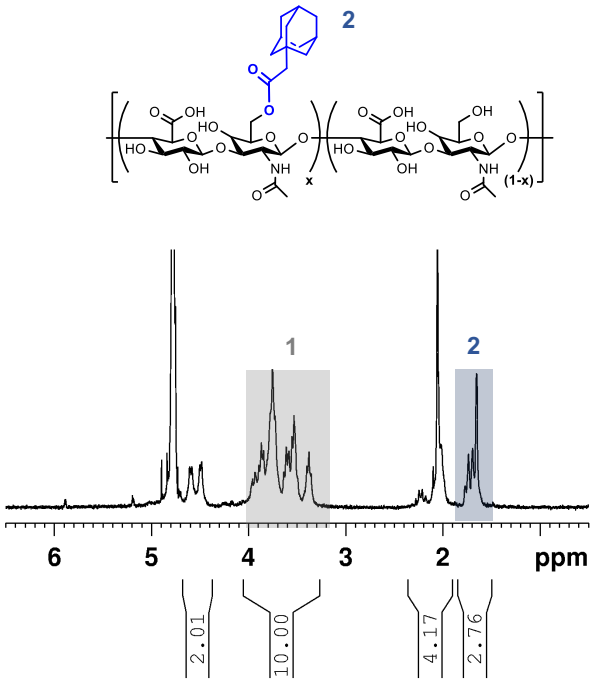
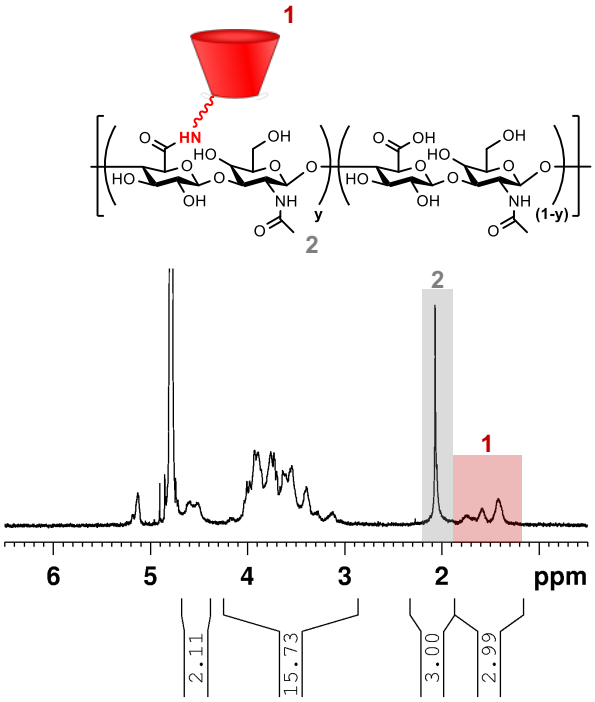
Electronic Supplementary Material (ESI) for Journal of Materials Chemistry B.

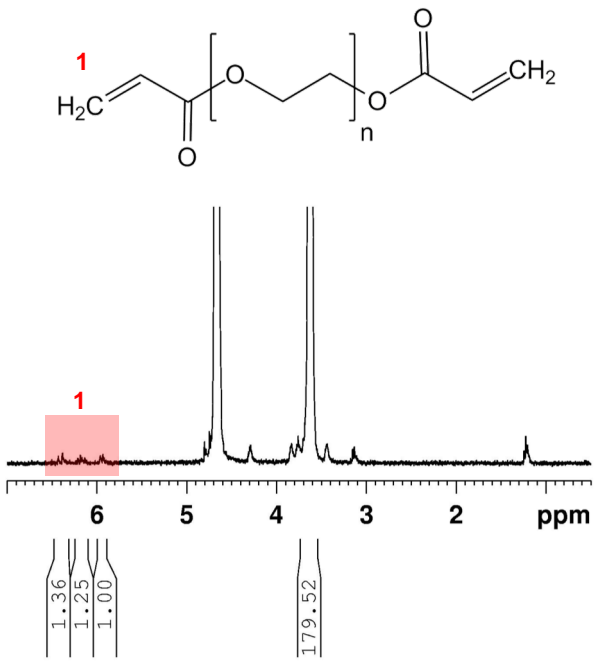

**Supplementary Figure 1 $^{1}$H NMR spectrum of adamantane-functionalized hyaluronic acid (Ad-HA) in $D_2O$.**

Modification of HA with pendant Ad (23%) determined by integration of the ethyl multiplet (12H, shaded blue) relative to the sugar ring of HA (10H, shaded grey).

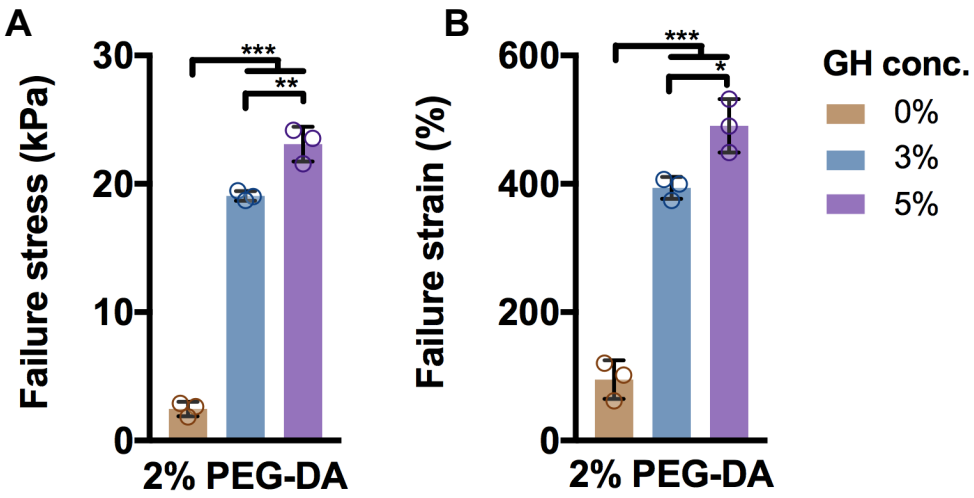



**Supplementary Figure 2 $^{1}$H NMR spectrum of β-cyclodextrin-functionalized hyaluronic acid (CD-HA) in $D_2O$.**

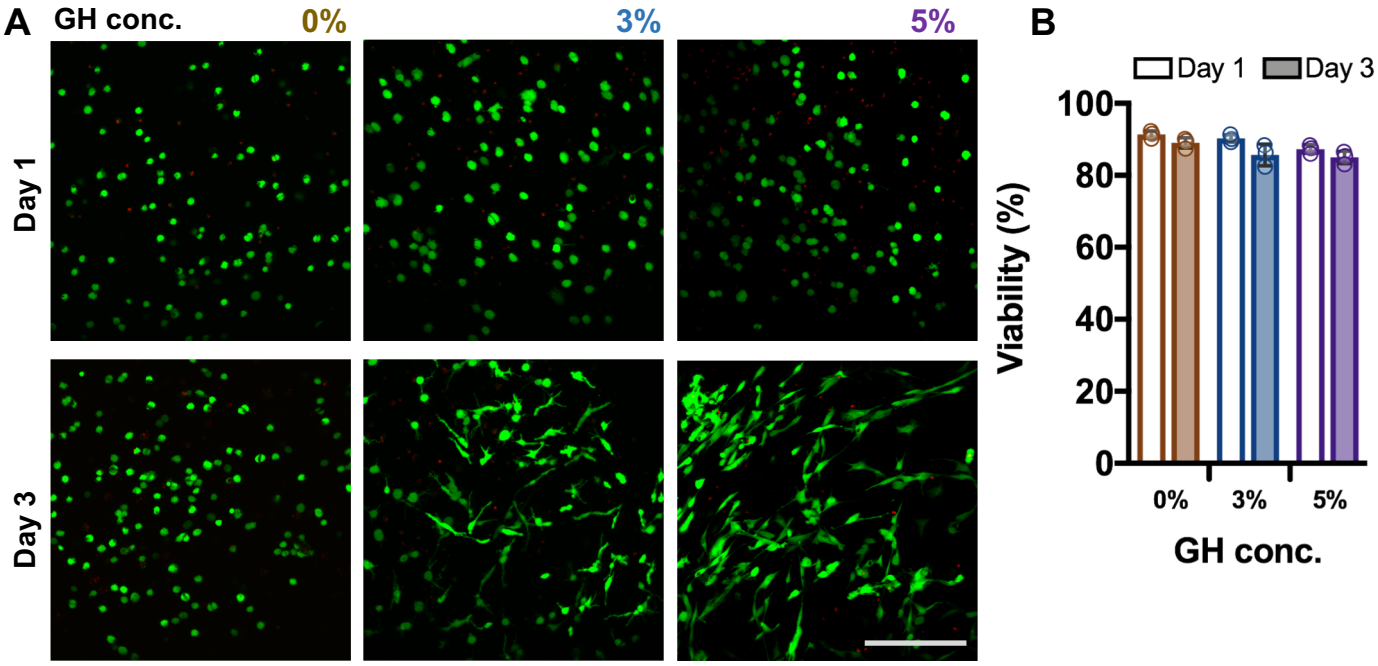
Modification of HA with pendant CD (24.9%) determined by integration of the hexane linkers (12H, shaded red) relative to the N-acetyl singlet of HA (3H, shaded grey).



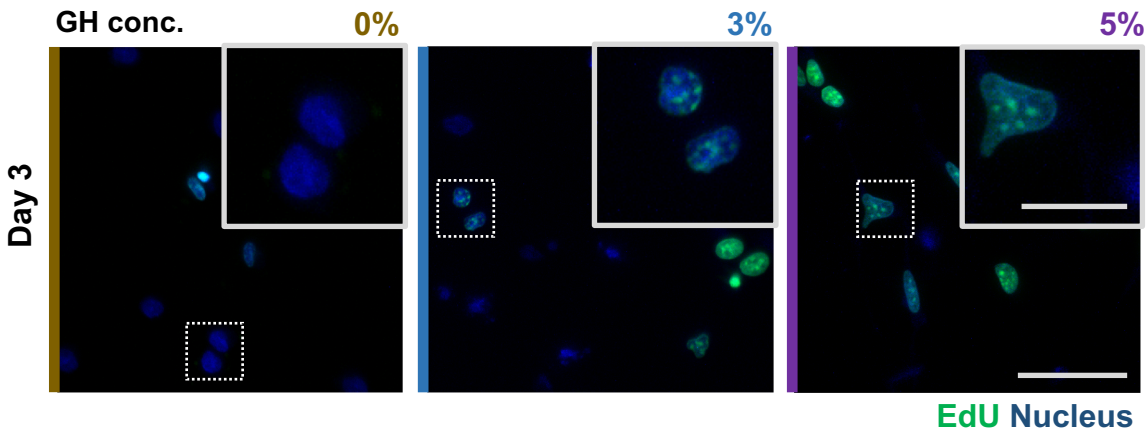

**Supplementary Figure 3 $^{1}$H NMR spectrum of poly(ethylene glycol) (PEG-DA) in $D_2O$.**
Modification of PEG (10 kDa) with diacrylate determined by integration of the acrylate groups (each 1H, shaded red).



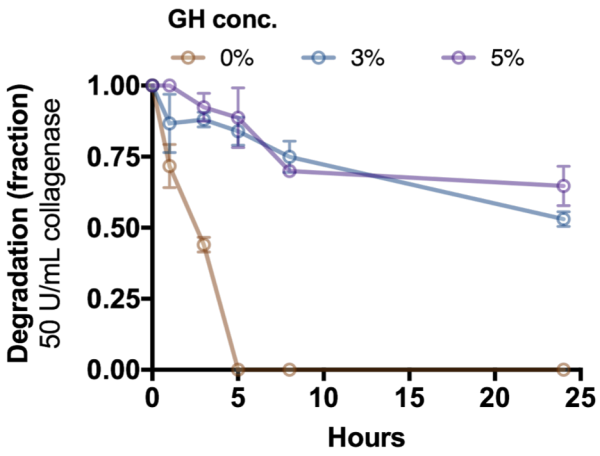

**Supplementary Figure 4 Characterization of tensile hydrogel properties.**
Influence of GH concentration on tensile (5.0 mm $sec^{-1}$ elongation) **A** failure stress and **B** failure strain of GH-DN hydrogels including PEG-fibrinogen (8 mg/mL plus 2% PEG-DA) and varying GH concentrations (conc.) of 0%, 3% and 5% (n = 3 replicates per group, mean ± SD, *p ≤ 0.05, ***p ≤ 0.001 by one-way ANOVA with Bonferroni *post hoc*).



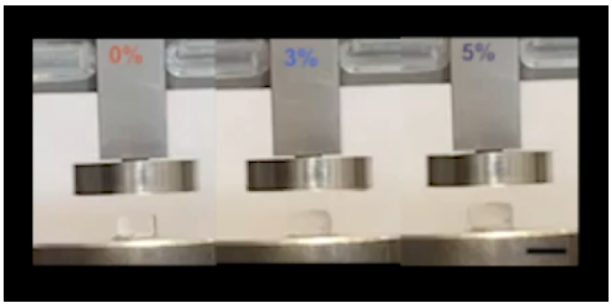

**Supplementary Figure 5 Cell viability in GH-DN hydrogels.**
A Representative images of MSCs stained with Calcein-AM (green, live) and Ethidium homodimer-1 (red, dead) after one and three days in GH-DN hydrogels including PEG-fibrinogen (8 mg/mL plus 2% PEG-DA) and varying GH concentrations (conc.) of 0%, 3% and 5% (scale bar = 200 µm). B Quantification of cell viability after one and three days of culture (n = 3 replicates per group, mean ± SD).



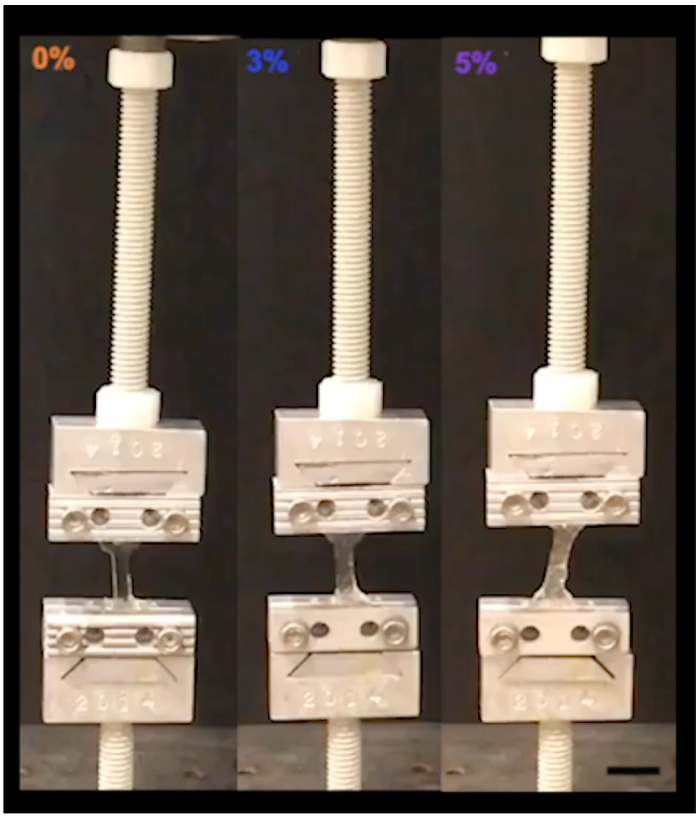

**Supplementary Figure 6 Detection of cell proliferation in GH-DN hydrogels.**
Representative images of 5-ethynyl-2'-deoxyuridine (EdU) incorporation into newly synthesized DNA over three days of MSCs in GH-DN hydrogels including PEG-fibrinogen (8 mg/mL plus 2% PEG-DA) and varying GH concentrations (conc.) of 0%, 3% and 5%, visualized with a fluorescent azide using click chemistry (scale bar 50 µm, inset 20 µm).



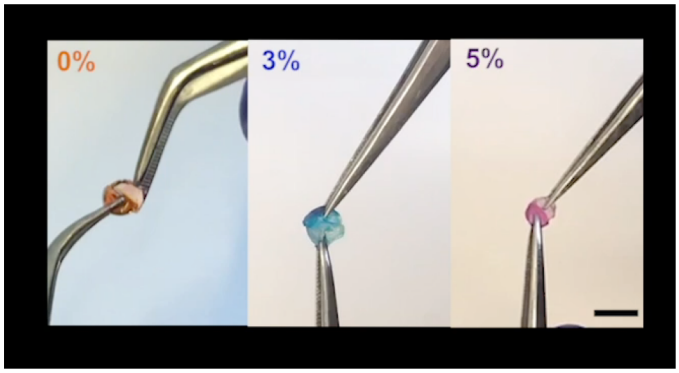

**Supplementary Figure 7 Enzymatic degradation of GH-DN hydrogel properties.**
**A** Proteolytic degradation over the time course of 24 hours in 50 Units/mL collagenase in PBS at 37° C of hydrogels including PEG-fibrinogen (8 mg/mL plus 2% PEG-DA) and varying GH concentrations (conc.) of 0%, 3% and 5% (n = 3 replicates per group, mean ± SD).




**Supplementary Video 1 Compressive failure of GH-DN hydrogels.**
GH-DN hydrogels including PEG-fibrinogen (8 mg/mL plus 2% PEG-DA) and varying GH concentrations of 0% (left), 3% (middle) and 5% (right) were compressed to 90% strain (strain rate 0.5 $N/min^{-1}$, scale bar 5 mm).




**Supplementary Video 2 Tensile failure of GH-DN hydrogels.**
GH-DN hydrogels including PEG-fibrinogen (8 mg/mL plus 2% PEG-DA) and varying GH concentrations of 0% (left), 3% (middle) and 5% (right) were elongated (5 $mm/sec^{-1}$) until failure was observed (scale bar 5 mm).




**Supplementary Video 3 Self-healing and mechanical loading of GH-DN hydrogels.**
GH-DN hydrogel fragments including PEG-fibrinogen (8 mg/mL plus 2% PEG-DA) and varying GH concentrations of 0% (left), 3% (middle) and 5% (right) were subjected to repeated application of tension upon contact (scale bar 5 mm).