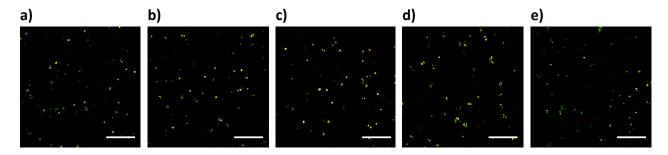


**Supplementary Figure 1.** a) Number of electrostatic contacts between positively charged groups of the peptide and the negatively charged groups of glycosaminoglycan. These contacts occur when the positively charged groups of the peptide (i.e., guanidinium group of arginine and primary amine of lysine) are within the Bjerrum length (7 Å) of the negatively charged groups of the glycosaminoglycan (i.e., carboxyl groups, COO<sup>-</sup>, and sulfate groups, SO<sub>3</sub><sup>-</sup>). The total number of electrostatic contacts with both carboxyl and sulfate groups is also shown. b) Number of hydrogen bonds (H-bonds) between the peptide and the glycosaminoglycan. As described in the main manuscript, hydrogen bonds are defined by geometric criteria involving the donor, hydrogen, and acceptor atoms. We quantify hydrogen bonds forming between the peptide and several groups of atoms in the glycosaminoglycan: atoms in carboxyl groups (COO<sup>-</sup>), atoms in sulfate groups (SO<sub>3</sub><sup>-</sup>), and all non-carbon atoms in the glycosaminoglycan (All). "Total" in the legend refers to hydrogen bonds formed between all hydrogen bond donors/acceptors. Error bars are (SD) of three independent simulation trials.

## Supplementary Figure 2.



**Supplementary Figure 2.** Representative confocal microscopy images of chondrocyte viability 28 days post encapsulation in PEG hydrogels with 1 mg/g HA encapsulated and covalently tethered peptide at concentrations of a) 0 mM, b) 0.1 mM native peptide, c) 1 mM native peptide, d) 5 mM native peptide, or e) 5 mM non-native peptide. Live cells fluoresce green and dead cells fluoresce red. Scale bars represent 200 µm.