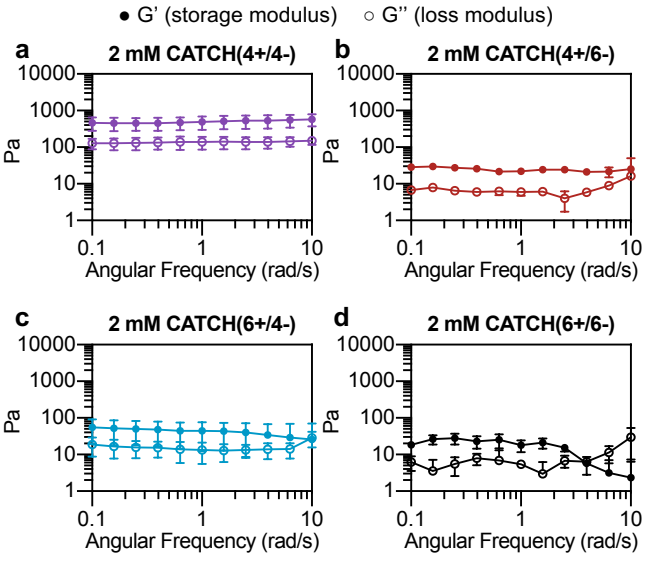
Electronic Supplementary Material (ESI) for Biomaterials Science.



e

Peptide pair	Gel-sol transition point
CATCH(4+/4-)	N/A
CATCH(4+/6-)	10 rad/s
CATCH(6+/4-)	10 rad/s
CATCH(6+/6-)	4 rad/s

Figure S1. Oscillating rheology of 2 mM CATCH(+/-) hydrogels. (a-d) Storage and loss modulus of 2 mM CATCH(4+/4-), CATCH(4+/6-), CATCH(6+/4-), and CATCH (6+/6-) at different angular frequencies. The filled circles correspond to the storage modulus and the open circles to the loss modulus. **(e)** Frequency at which the gel stops behaving as a viscoelastic solid and behaves as a liquid ("gel-sol transition point"). Data are presented as the mean ± standard deviation (n = 3).

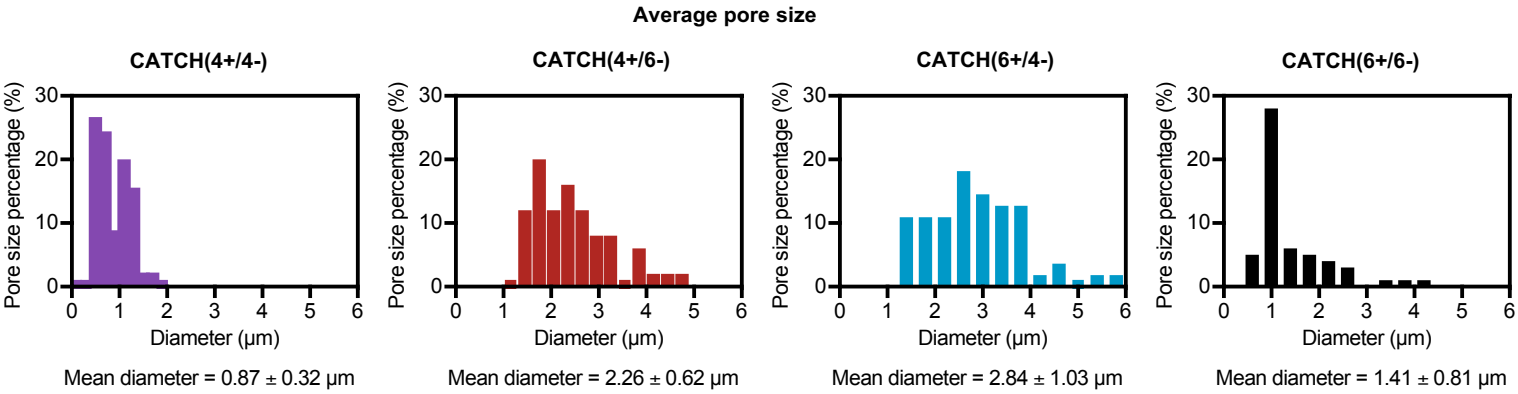


Figure S2. Average pore size analysis of CATCH(+/-) hydrogels. **H**istograms of the pore diameter measured from micrographs in (Figure 4) (n = 30 measurements).

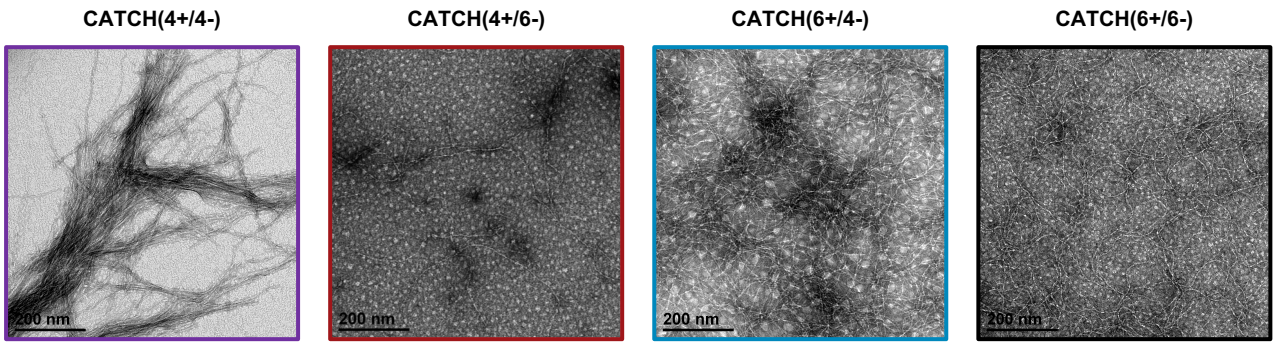


Figure S3. Electron microscopy analysis of CATCH(+/-) nanofibers morphology. TEM micrographs of 1 mM CATCH(4+/4-), CATCH(4+/6-), CATCH(6+/4-), and CATCH(6+/6-) nanofibers.

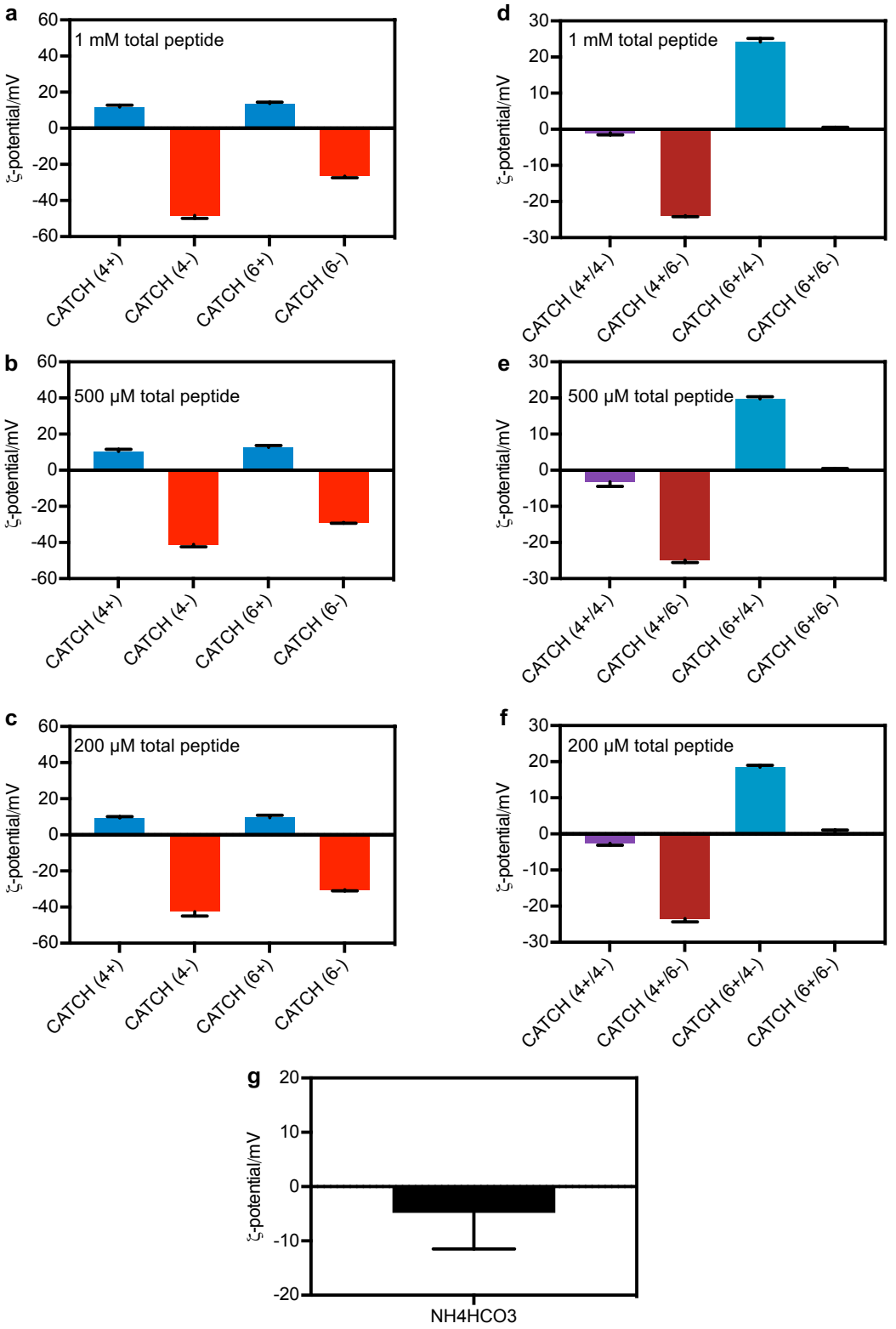


Figure S4. Zeta potential of CATCH(+/-) peptides alone and in combination. (a-c) Zeta potential measurements of CATCH(4+), CATCH(4-), CATCH(6+), and CATCH(6-) at 1 mM, 500 μM, and 200 μM total peptide. **(d-f)** Zeta potential measurements of equimolar mixtures of CATCH(4+/4-), CATCH(4+/6-), CATCH(6+/4-), and CATCH(6+/6-) at 1 mM, 500 μM, and 200 μM total peptide. **(g)** Zeta potential measurement of NH4HCO3 buffer. Data are presented as the mean ± standard deviation (n = 3).

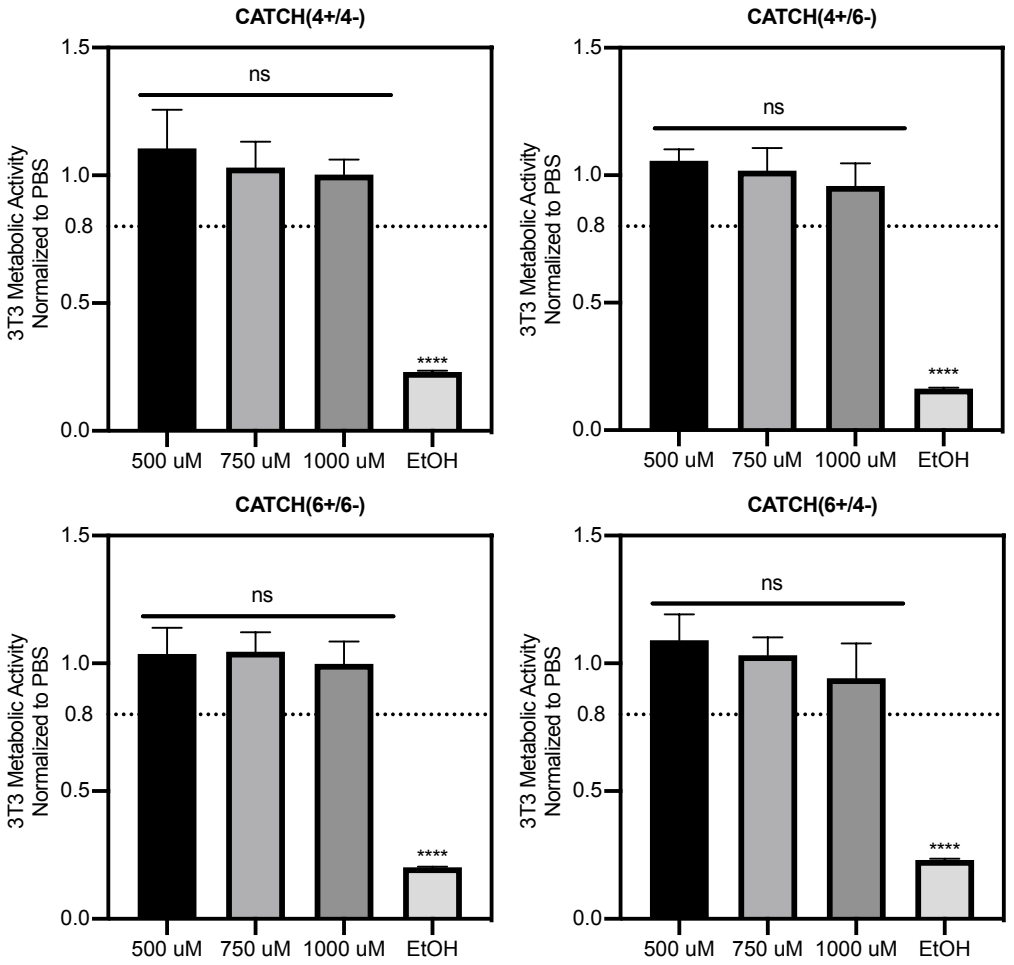


Figure S5. Metabolic activity of NIH 3T3 fibroblast cells after treatment with CATCH(+/-) nanofibers. Data are presented as mean ± standard deviation (n = 5). **** represents $p < 0.0001$, ANOVA with Tukey's post-hoc.

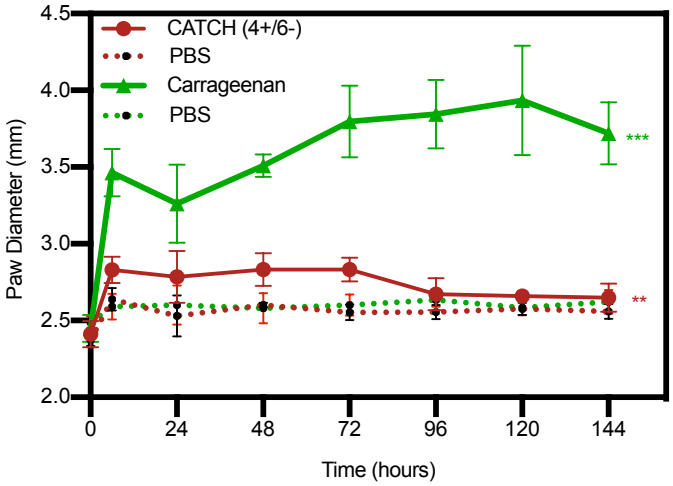


Figure S6. Caliper measurements of paw thickness after injection of CATCH(4+/6-) hydrogel, λ-Carrageenan, or PBS vehicle control into male C57BL/6 mice. Data presented as mean ± standard deviation (n = 5). ** represents $p < 0.01$, *** represents $p < 0.001$, repeated measures ANOVA with Dunnett's multiple comparison.

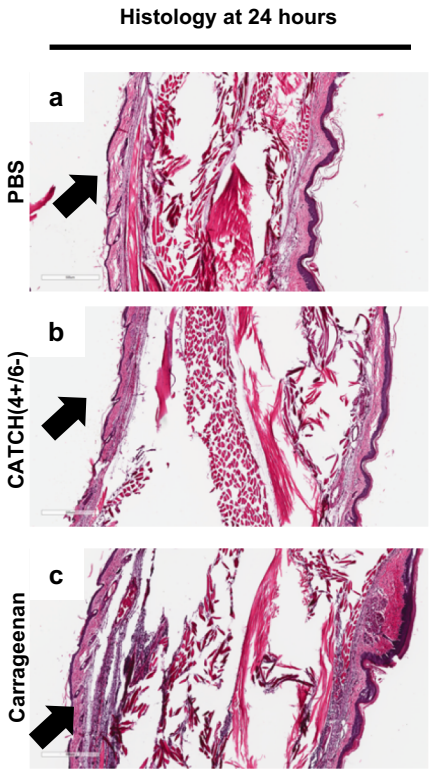


Figure S7. Representative histology sections of tissues collected 24 h after injection of CATCH(4+/6-), λ-Carrageenan, or PBS into the subcutaneous space of female C57BL/6 mice. Arrows show the site of injection.

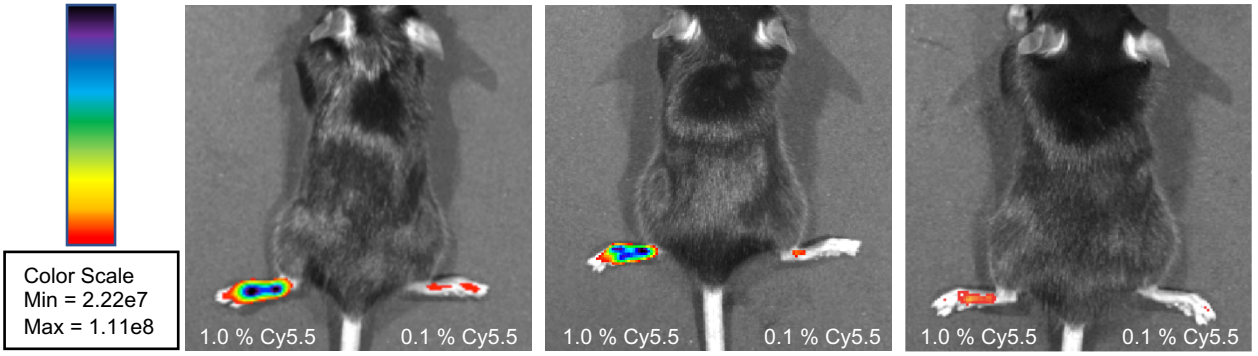


Figure S8. In vivo fluorescence imaging of CATCH(4+/6-) hydrogels. Hydrogels with Cy5.5-labeled CATCH(4+) peptide (1.0 or 0.1% of total peptide), unlabeled CATCH(4+) peptide (49 or 49.9% of total peptide) and CATCH(6-) (50% of total peptide) were injected subcutaneously and imaged using an IVIS system. Color scale represents radiant efficiency ([p/sec/cm^2/sr] / [μW/cm^2]).

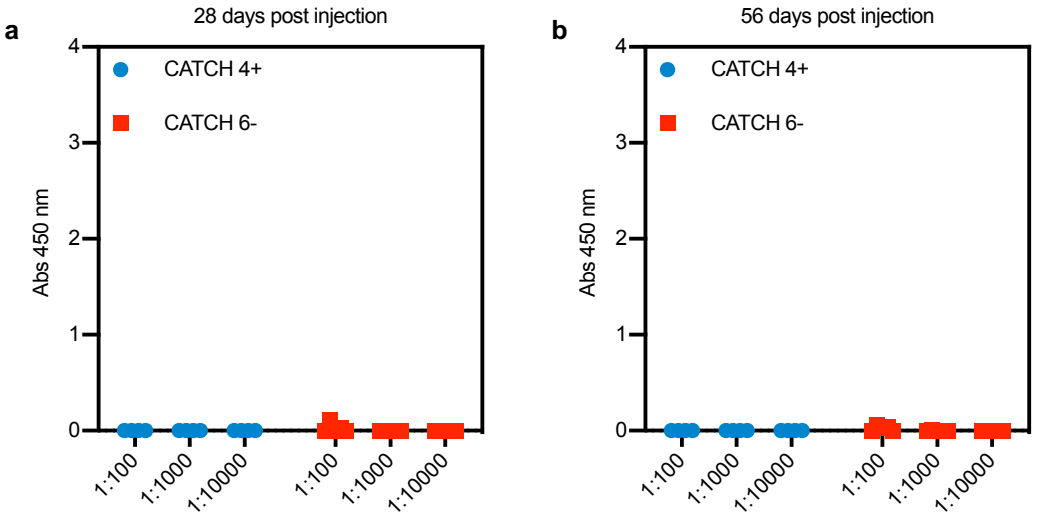


Figure S9. Total IgG ELISA absorbance readings of dilutions of sera collected on day 28 and day 56 from female C57BL/6 mice that received a subcutaneous injection of 12 mM CATCH(4+/6-) hydrogel on day 0, followed by CATCH(4+) or CATCH(6-) in PBS on days 14 and 42. Data presented as a single point for each animal (n = 5).

Peptide	Sequence	Adjusted Rank
CATCH(4+)	QQKFKFKFKQQ	322.81
CATCH(6-)	EQEFEFEFEQE	417.08
Q11	QQKFQFQFEQQ	331.38
OVA	ISQAVHAAHAEINEAGR	4.4

Figure S10. Immune Epitope Database (IEDB) predictions for binding of CATCH(4+), CATCH(6-), Q11, and OVA to C57BL/6 MHC II (H2-IAb background).