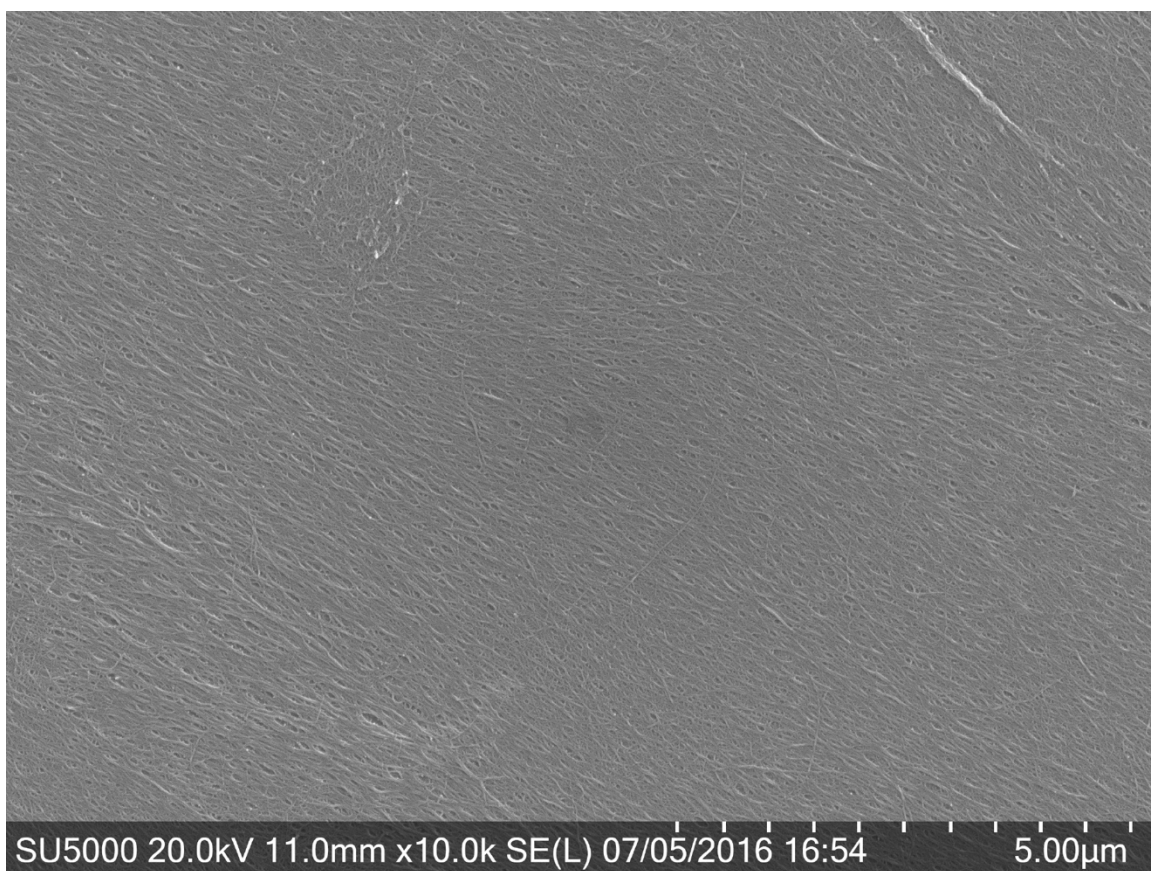
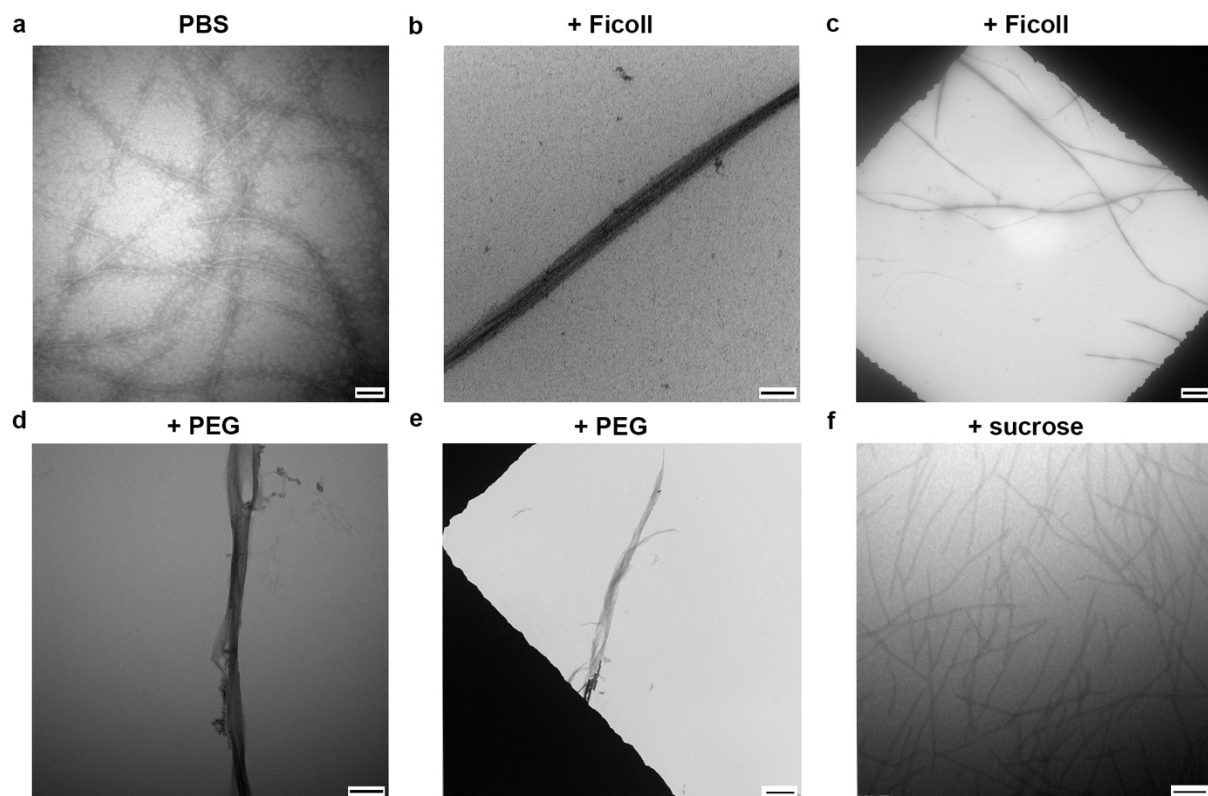


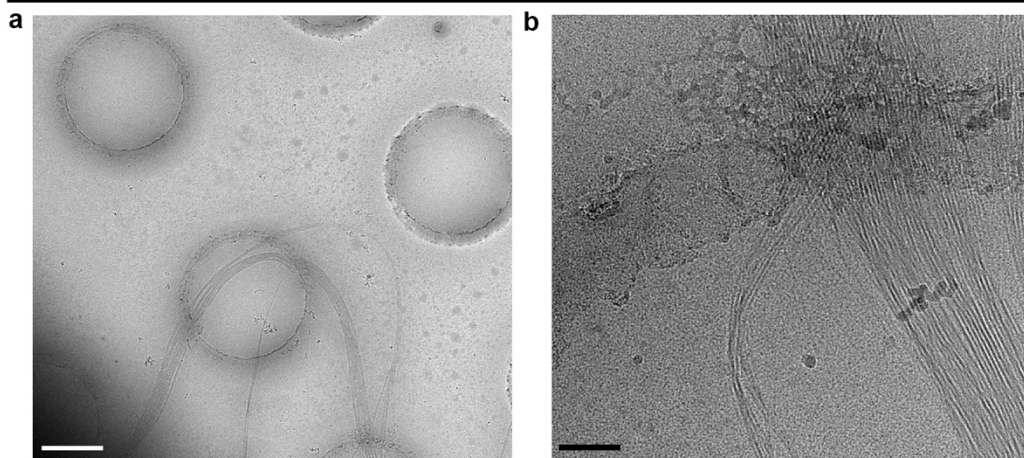


Supplementary Figure 1: FE-SEM of GQ11 hydrogel demonstrating long-range alignment of nanofibers.

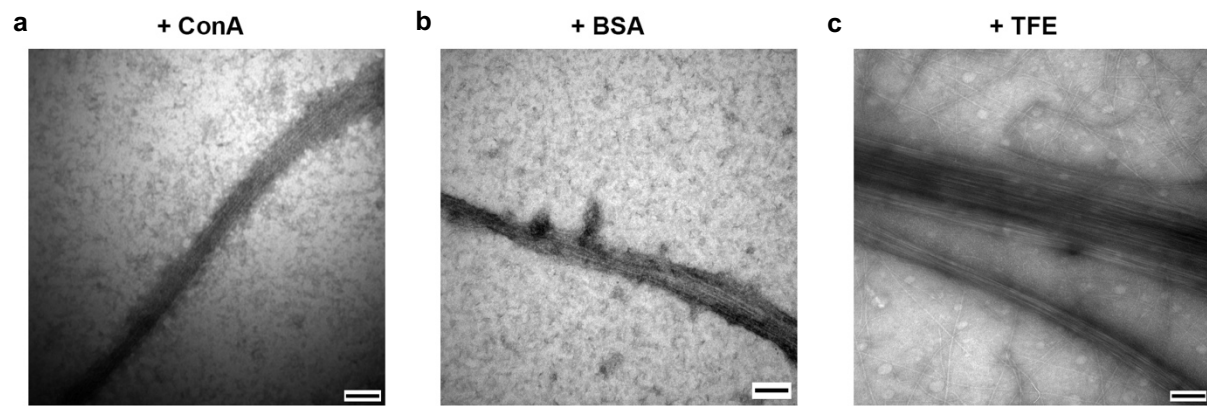


Supplementary Figure 2: TEM images of GQ11 in **a**, PBS, or in the presence of **b-c**, Ficoll, **d-e**, PEG2000, and **f**, sucrose. Scale bar = 50 nm in **a**. Scale bar = 100 nm in **b**, **d**, and **f**. Scale bar = 2 μm in **c**. Scale bar = 1 μm in **e**.

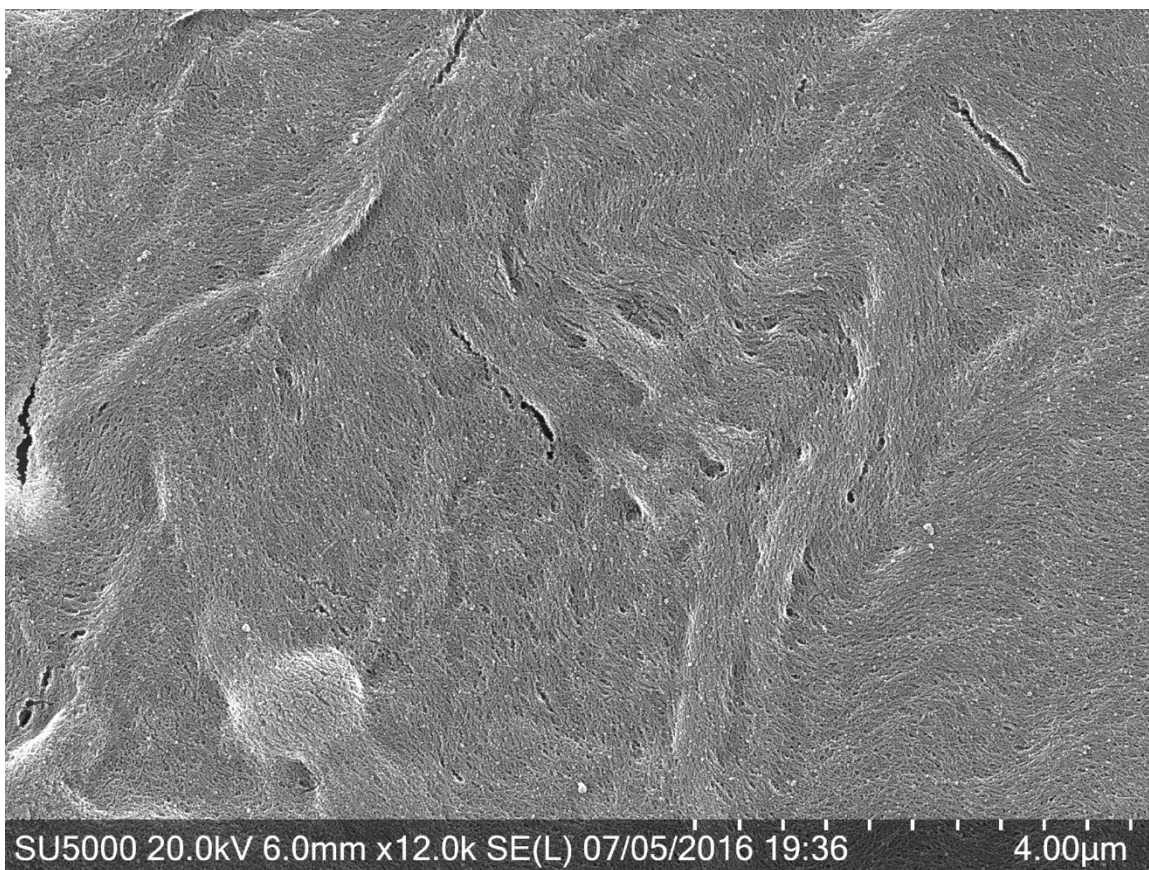
GQ11 + Ficoll



Supplementary Figure 3: Cryo-TEM of GQ11 nanofibers in the presence of Ficoll. Scale bar = 1 μm in a and 100 nm in b.

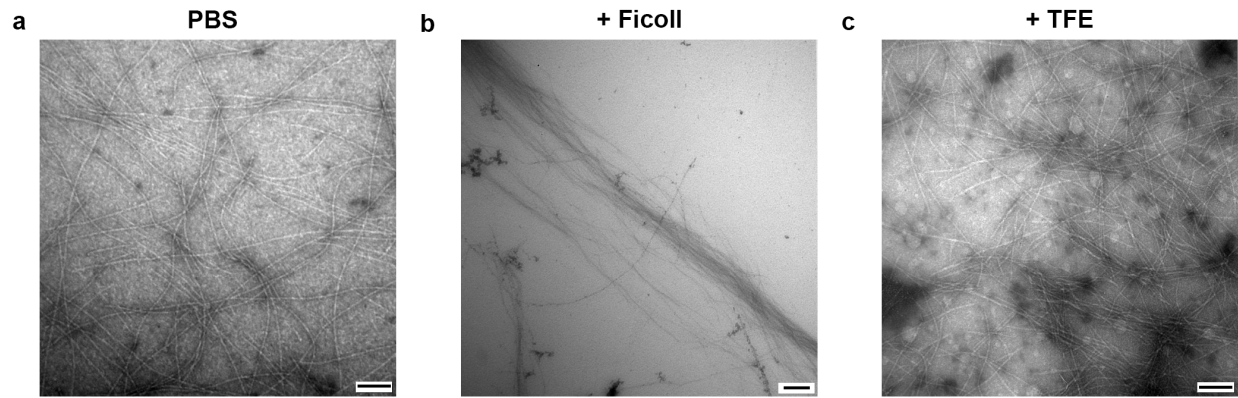


Supplementary Figure 4: TEM images of GQ11 in **a**, ConA, **b**, BSA, and **c**, TFE. Scale bar = 100 nm in a, b, and c.

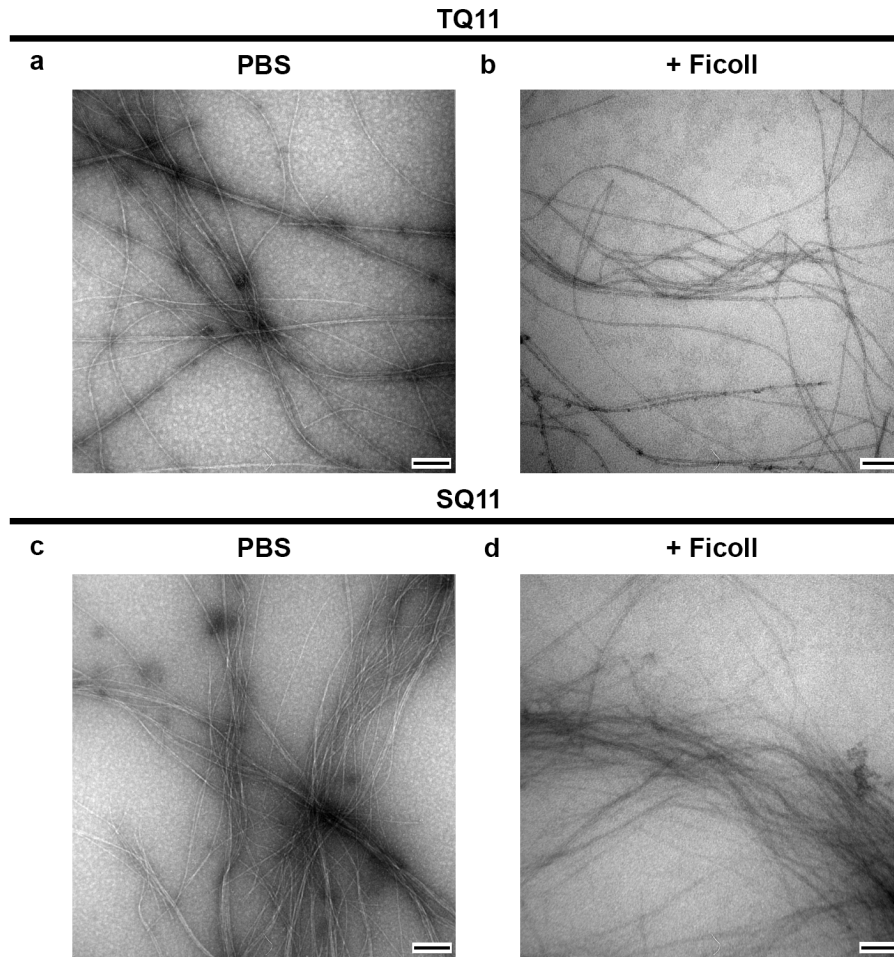


Supplementary Figure 5: FE-SEM of AcGQ11 hydrogel.

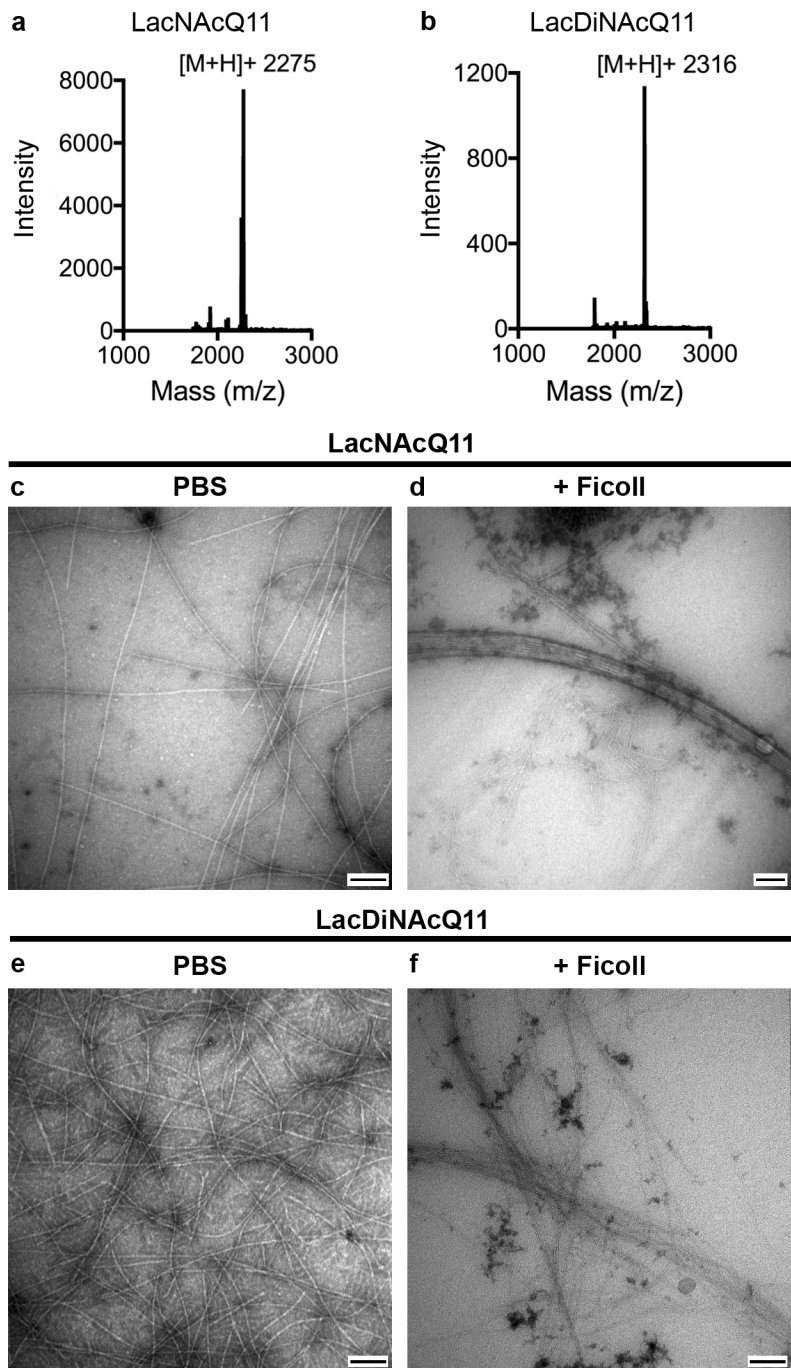
AcGQ11



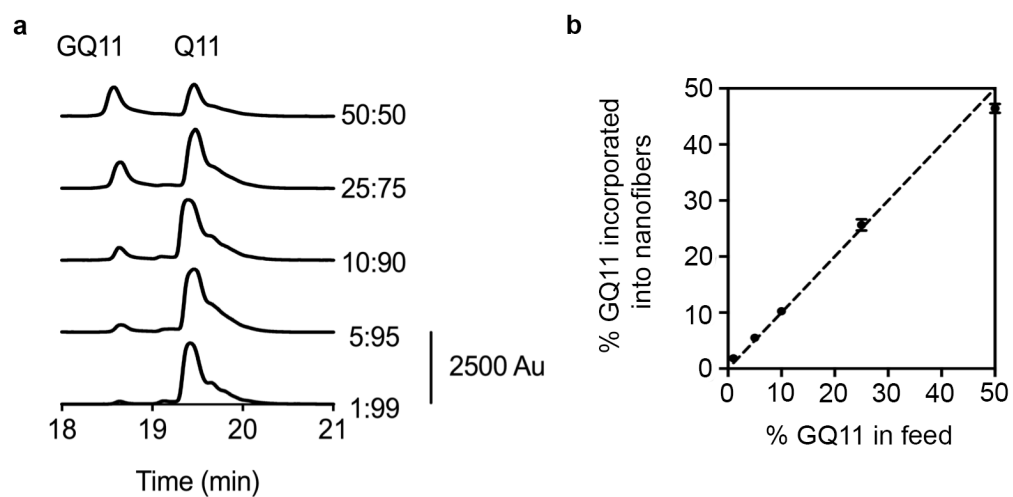
Supplementary Figure 6: TEM images of AcGQ11 in **a**, PBS, or in the presence of **b**, Ficoll, or **c**, TFE. Scale bar = 100 nm in **a** and **c**. Scale bar = 200 nm in **b**.



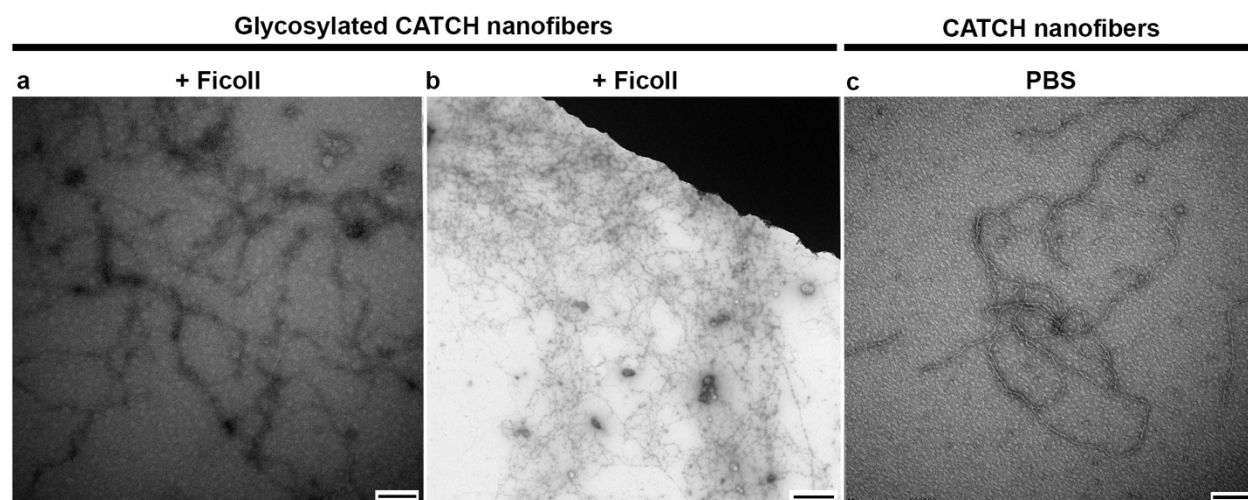
Supplementary Figure 7: TEM images of TQ11 in **a**, PBS and **b**, PBS + Ficoll. TEM images of SQ11 in **c**, PBS and **d**, PBS + Ficoll. Scale bar = 100 nm in a-d.



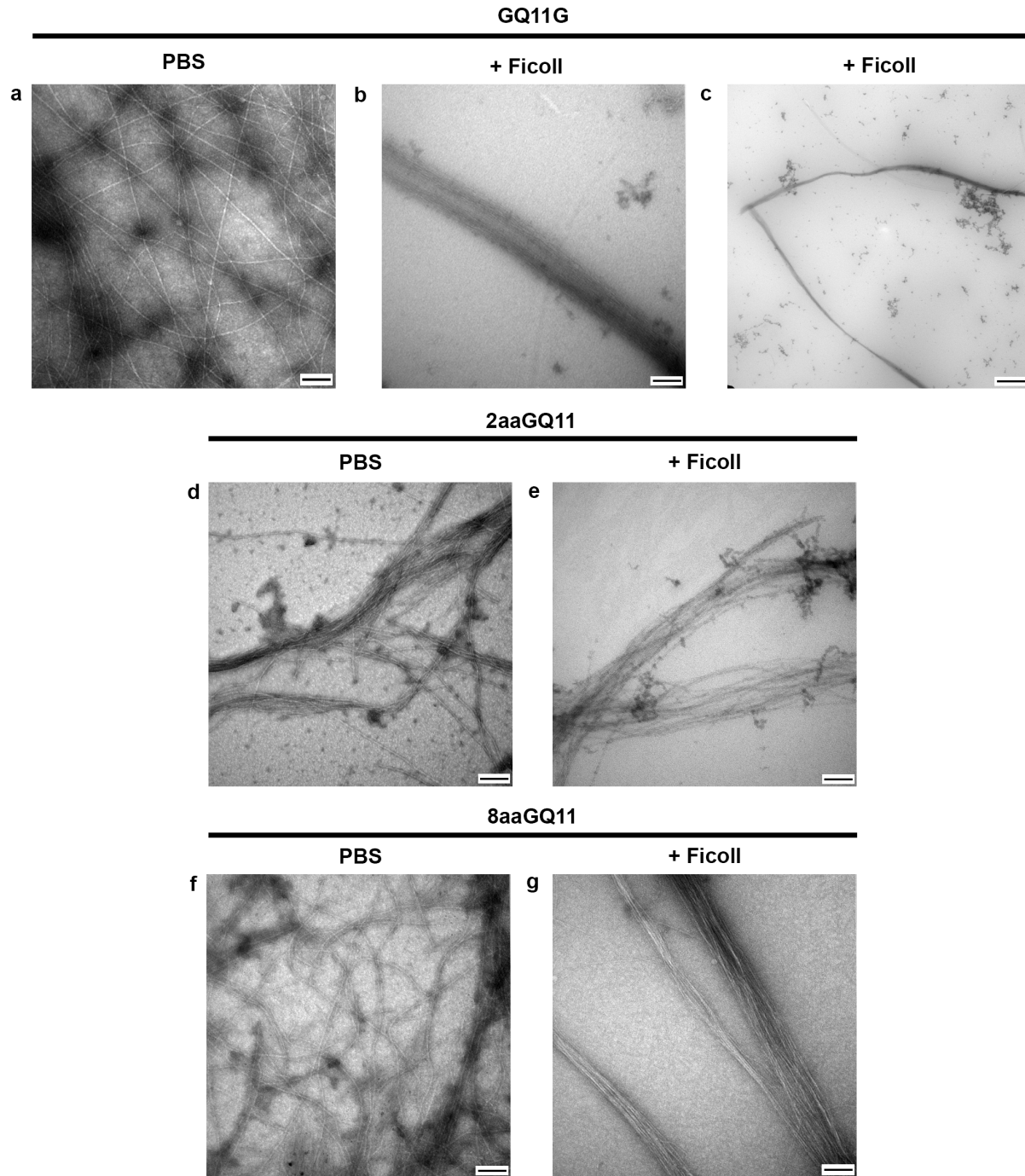
Supplementary Figure 8: MALDI-TOF spectra of **a**, LacNacQ11 and **b**, LacDiNacQ11 after enzymatic conversion. TEM images of LacNacQ11 in **c**, PBS and **d**, PBS + Ficoll. TEM images of LACDiNacQ11 in **e**, PBS and **f**, Ficoll. Scale bar = 100 nm in c, e, and f. Scale bar = 50 nm in d.



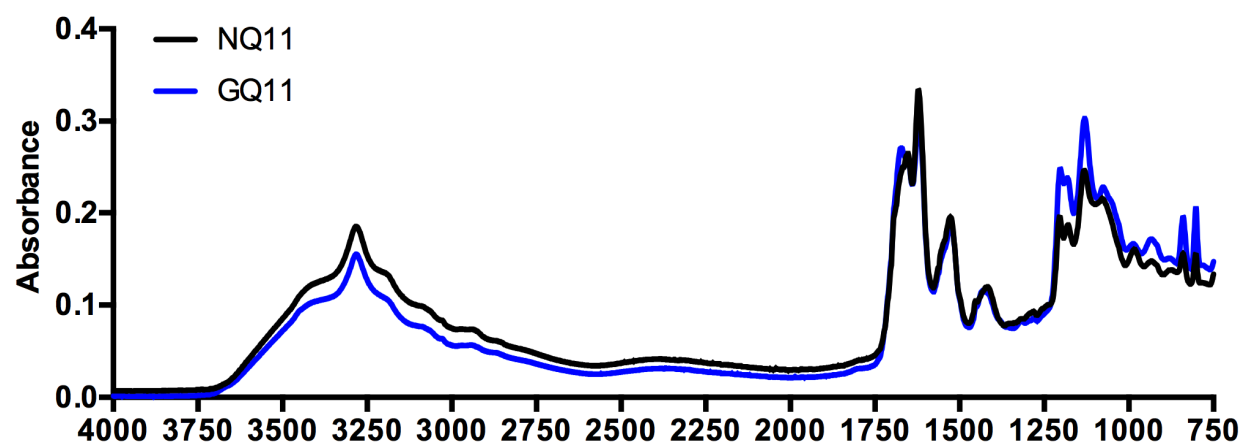
Supplementary Figure 9: Co-assembly of GQ11 and Q11 in nanofibers with tunable carbohydrate content. (a) HPLC traces of co-assembled GQ11:Q11 nanofibers. (b) Quantification of area under the peaks from HPLC traces demonstrates a linear relationship between peptide in the feed and peptide integrated into the nanofiber. Data reprinted from Restuccia, A., Tian, Y.F., Collier, J.H., Hudalla, G.A., Cellular and Molecular Bioengineering 8(3): 471-487, 2015.



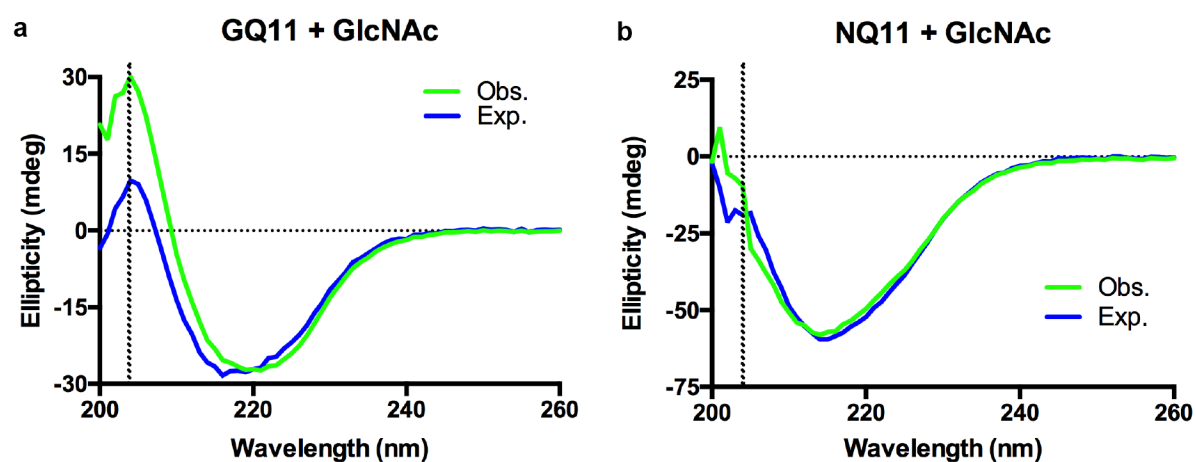
Supplementary Figure 10: **a, b** TEM micrographs of glycosylated CATCH nanofibers in the presence of Ficoll. **c**, non-glycosylated CATCH nanofibers in PBS. Scale bar = 100 nm in a and c. Scale bar = 1 μm in b.



Supplementary Figure 11: TEM images of GQ11G in **a**, PBS and **b-c**, PBS + Ficoll. TEM images of 2aaGQ11 in **d**, PBS and **e**, PBS plus Ficoll. TEM images of 8aaGQ11 in **f**, PBS and **g**, PBS + Ficoll. Scale bar = 100 nm in **a**, **b**, **d**, **e**, **f**, and **g**. Scale bar = 1 μ m in **c**.

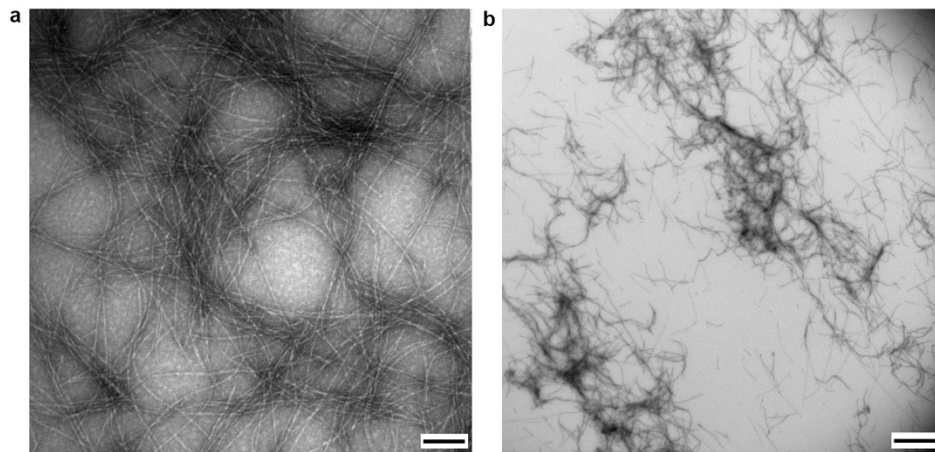


Supplementary Figure 12: FTIR spectra of GQ11 and NQ11 in PBS over the range of 4000-750 cm⁻¹.

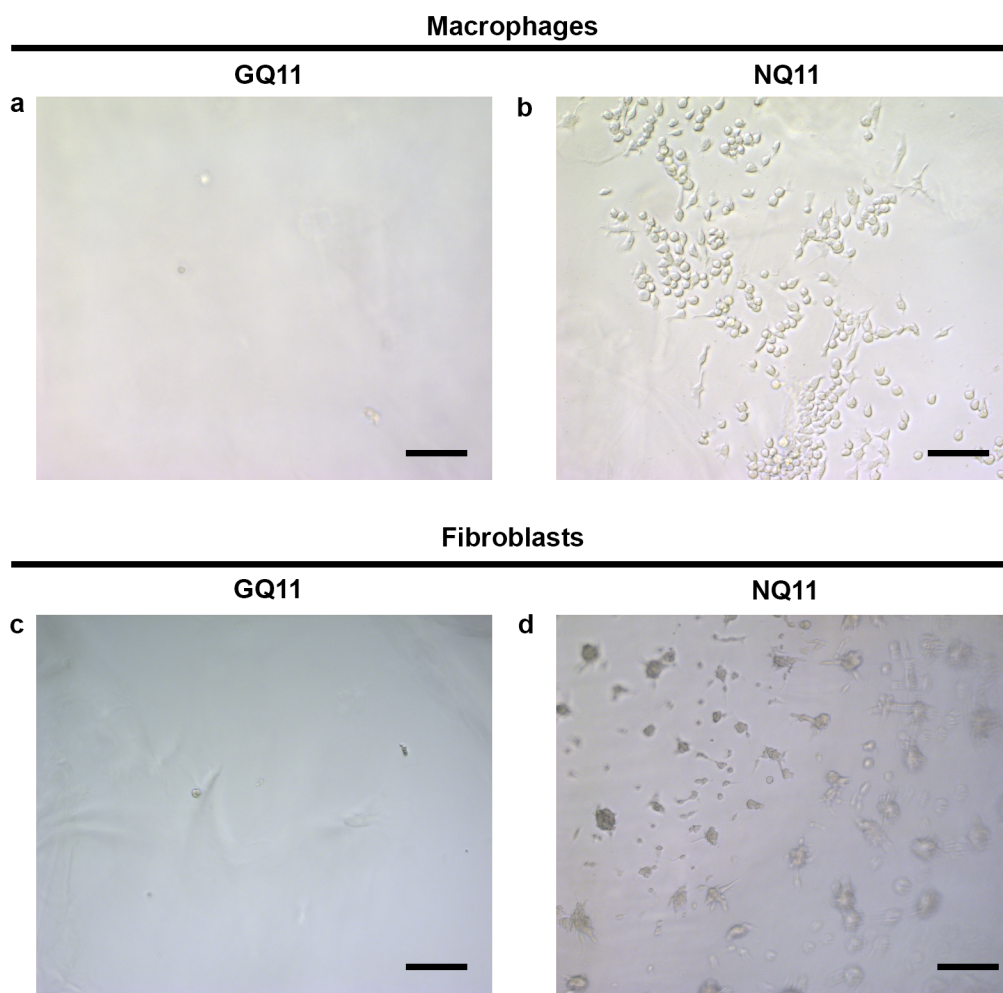


Supplementary Figure 13: Independent biological replicates of samples of **a**, GQ11 + GlcNAc and **b**, NQ11 + GlcNAc analyzed with CD. “Obs” denotes the observed spectrum for a sample of nanofiber + GlcNAc, whereas “Exp” denotes the expected spectrum based on the sum of the nanofiber and GlcNAc spectra collected independently. Dashed line indicates 203 nm.

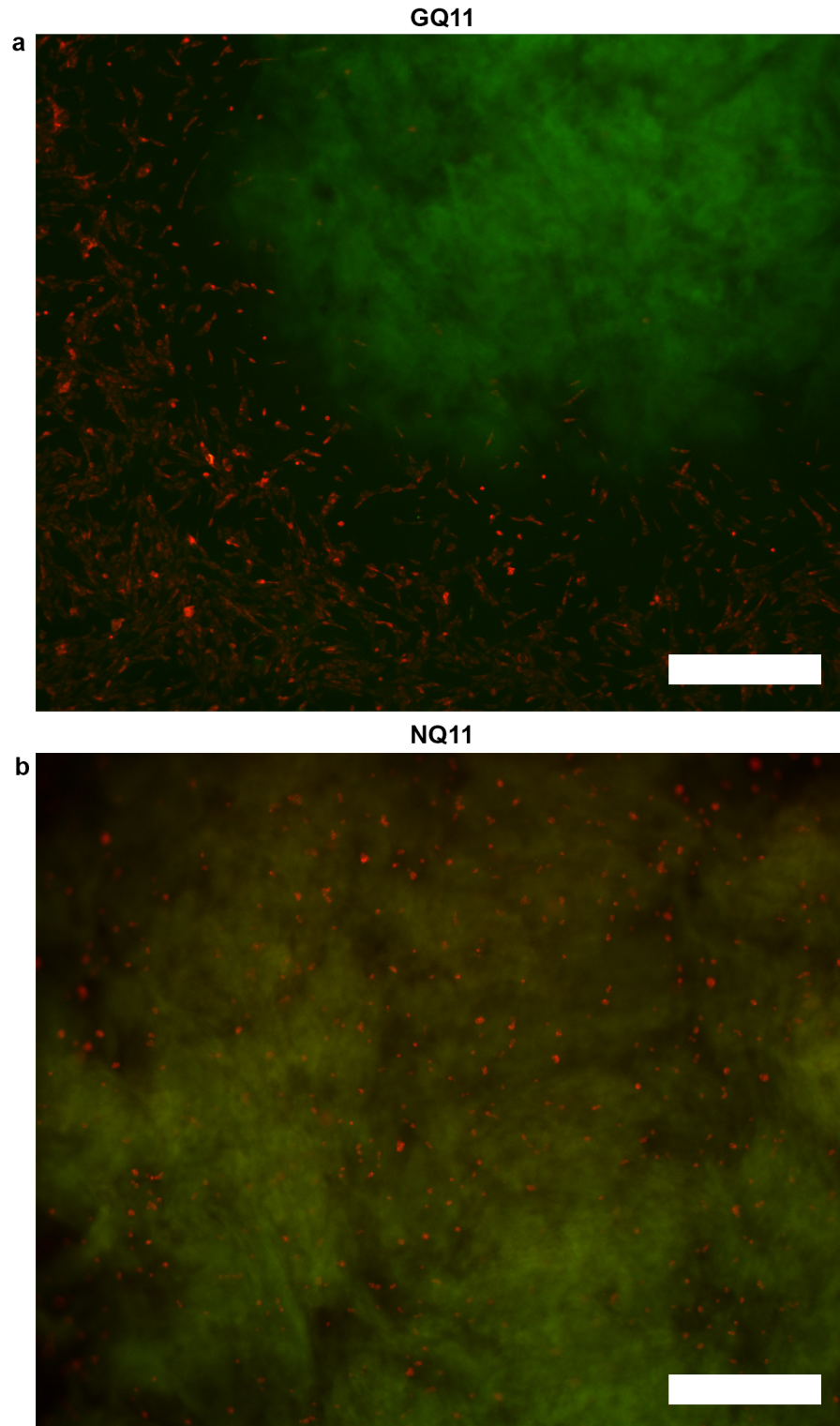
GQ11 + N,N',N triacetylchitotriose



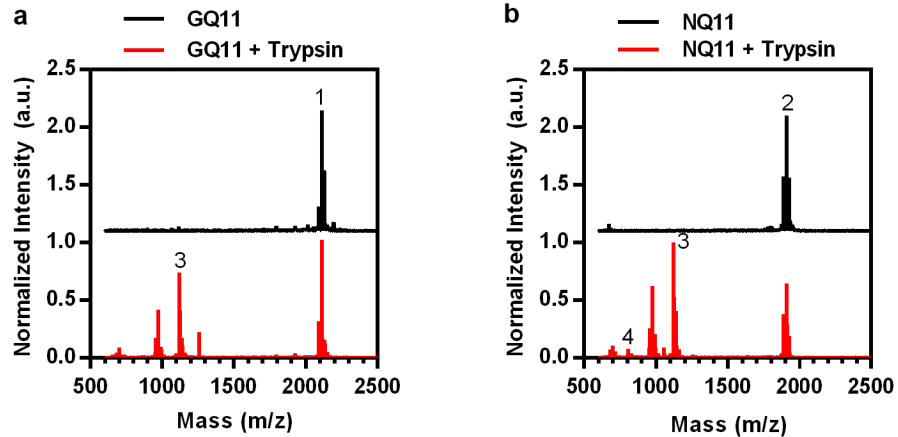
Supplementary Figure 14: a, b TEM micrographs of GQ11 nanofibers in 1x PBS plus N,N',N triacetylchitotriose. Scale bar = 100 nm in a and 1 μm in b.



Supplementary Figure 15: Bright-field images of RAW264.7 macrophage adhesion on **a**, GQ11 and **b**, NQ11 hydrogels. Bright-field images NIH3T3 fibroblast adhesion on **c**, GQ11 and **d**, NQ11 hydrogels. Scale bar = 100 μm .

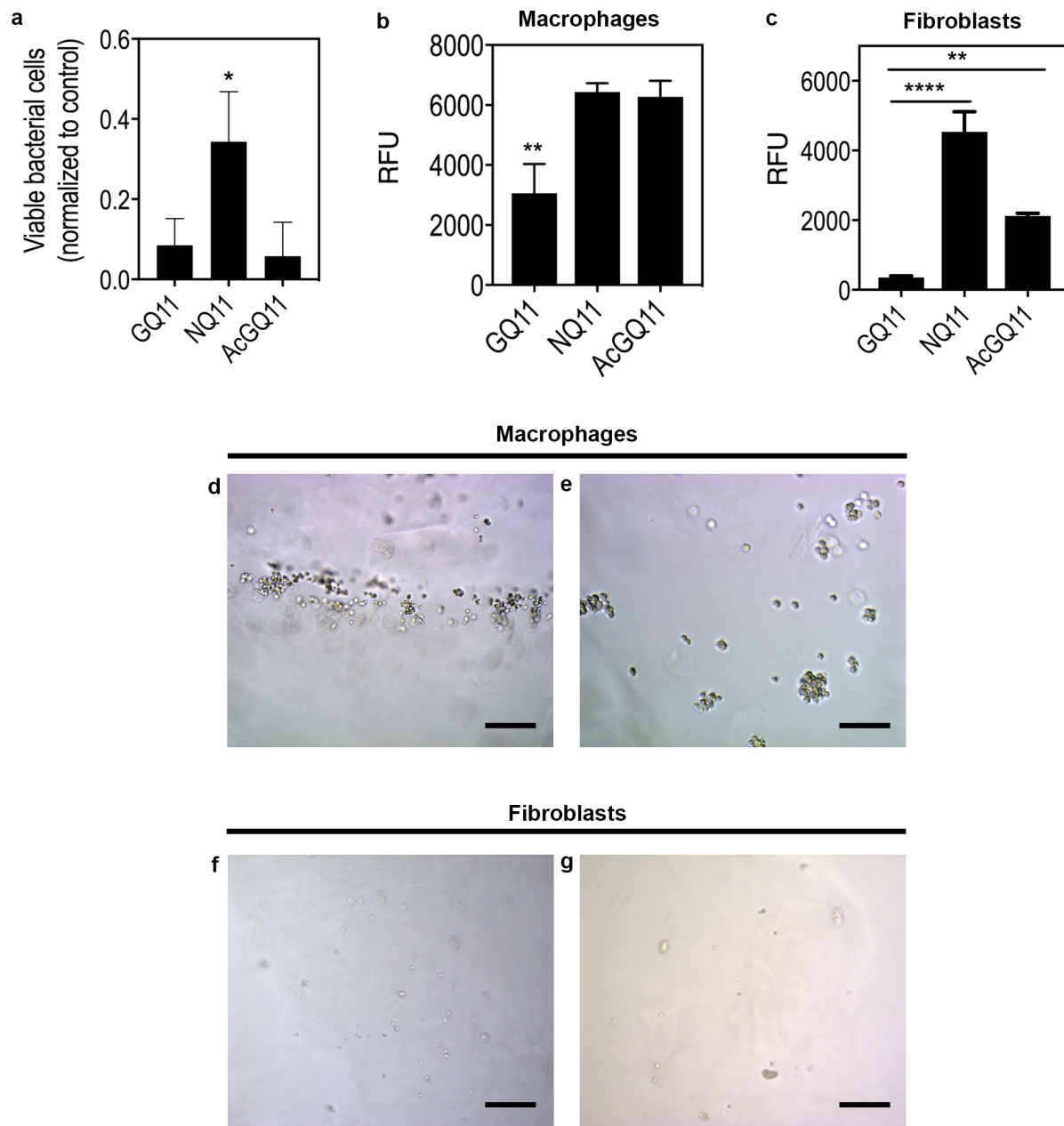


Supplementary Figure 16: Fibroblast adhesion to hydrogel-coated glass surfaces. NIH3T3 fibroblasts **a**, did not adhere to GQ11-coated glass, but **b**, did adhere to NQ11-coated glass. Nanofibers were stained green with Thioflavin T dye (green) and cells were labeled with CellTracker (red). Scale bar 500 μm .

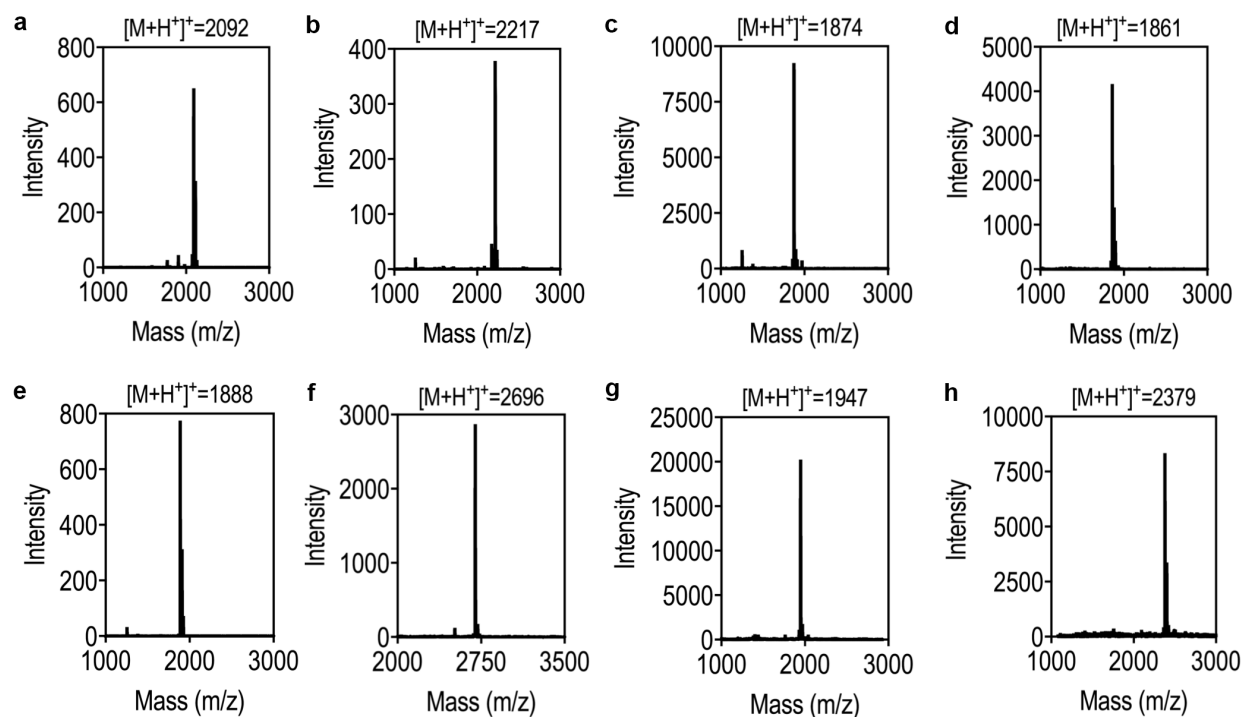


Fragment	Peptide/Fragment	Molecular Weight
1	N(GlcNAc)SGSGQQKFQFQFEQQ	2090+Na
2	NSGSGQQKFQFQFEQQ	1186+Na
3	FQFQFEQQ	1101+Na
4	NSGSGQQK	805

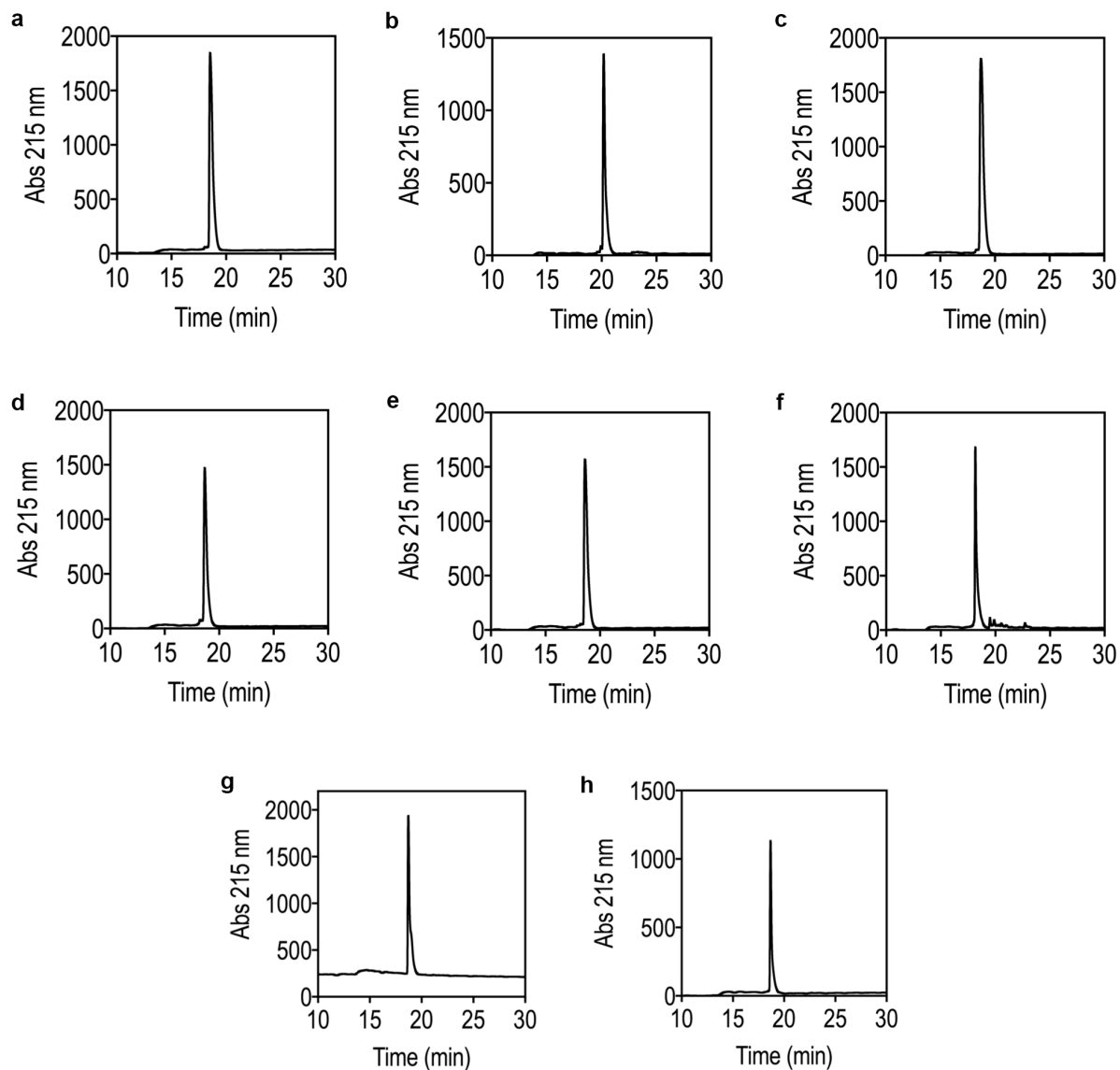
Supplementary Figure 17: MALDI-TOF mass spectra of **a**, GQ11 and **b**, NQ11 before (black) and after (red) digestion with trypsin. Table assigning peptide fragments to observed MALDI-TOF peaks.



Supplementary Figure 18: Cell adhesion to AcGQ11 hydrogels. Quantitative comparison of **a**, Bacteria, **b**, RAW264.7 macrophages, and **c**, NIH3T3 fibroblast adhesion to AcGQ11, GQ11, and NQ11 hydrogels. Micrographs of **d-e**, macrophage and **f-g**, fibroblast adhesion onto AcGQ11 hydrogels. Scale bar = 100 μ m in **d-g**. In **a-c**, data presented as mean $\pm$ standard deviation (n = 3). For **a**, * represents $p < 0.05$. For **b**, ** represents $p < 0.01$. For **c**, ** represents $p < 0.002$ and **** $p < 0.0001$.



Supplementary Figure 19: Peptide molecular weights determined using MALDI-TOF. **a**, GQ11. **b**, AcGQ11. **c**, TQ11. **d**, SQ11. **e**, NQ11. **f**, GQ11G. **g**, 2aaGQ11. **h**, 8aaGQ11.



Supplementary Figure 20: HPLC traces of Q11 peptide variants. **a**, GQ11. **b**, AcGQ11. **c**, TQ11. **d**, SQ11. **e**, NQ11. **f**, GQ11G. **g**, 2aaGQ11. **h**, 8aaGQ11. Peptide purity was greater than 90% for all experiments.