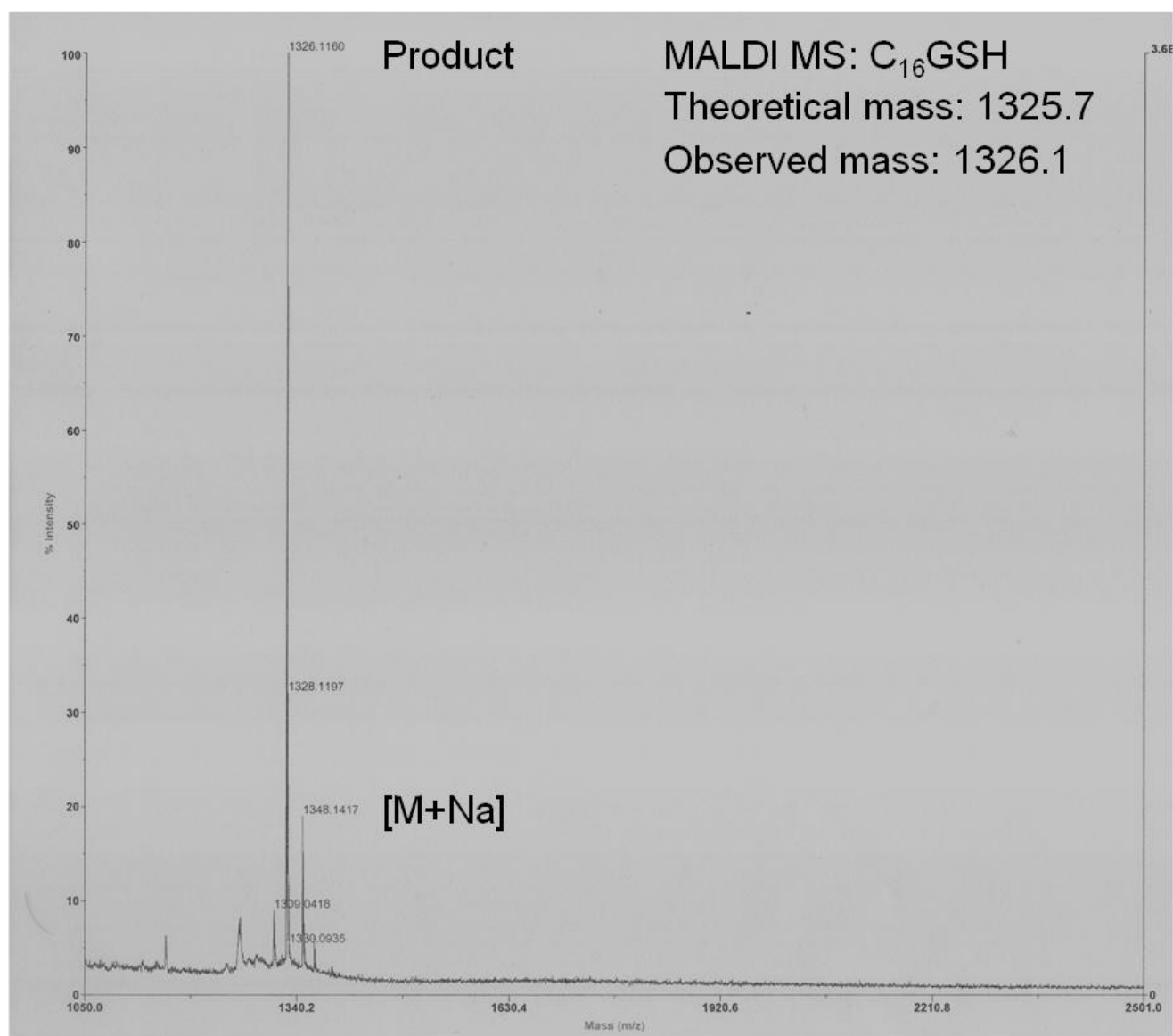
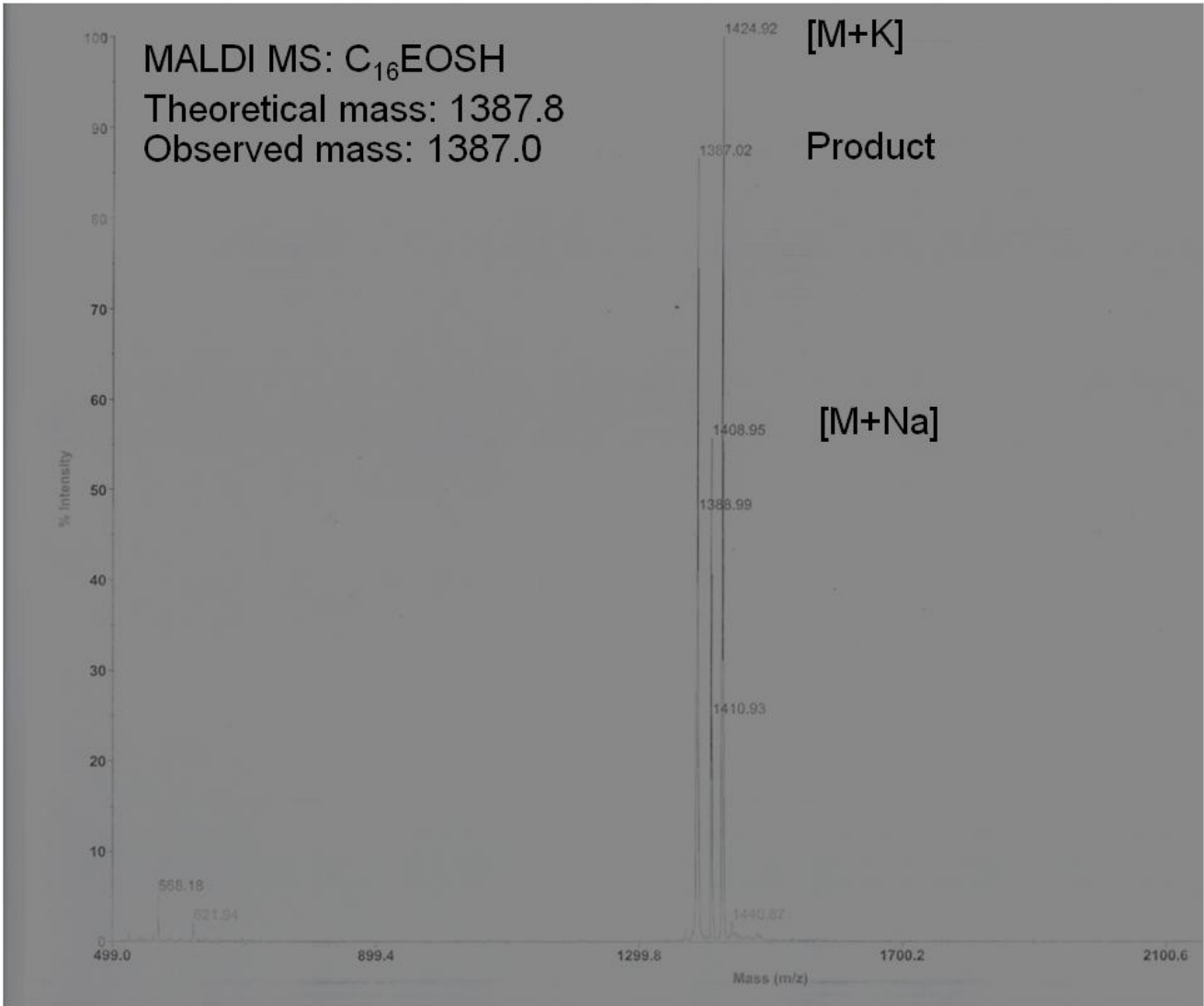




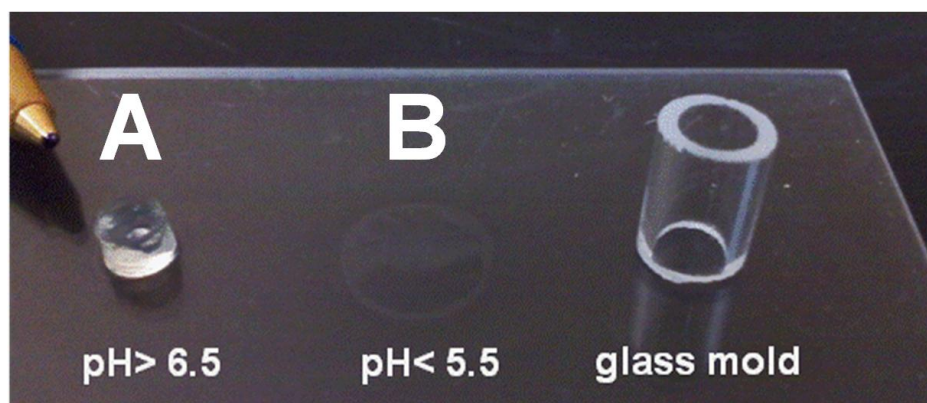
**Supplemental Information 1A.** Maldi MS of C<sub>16</sub>GSH.



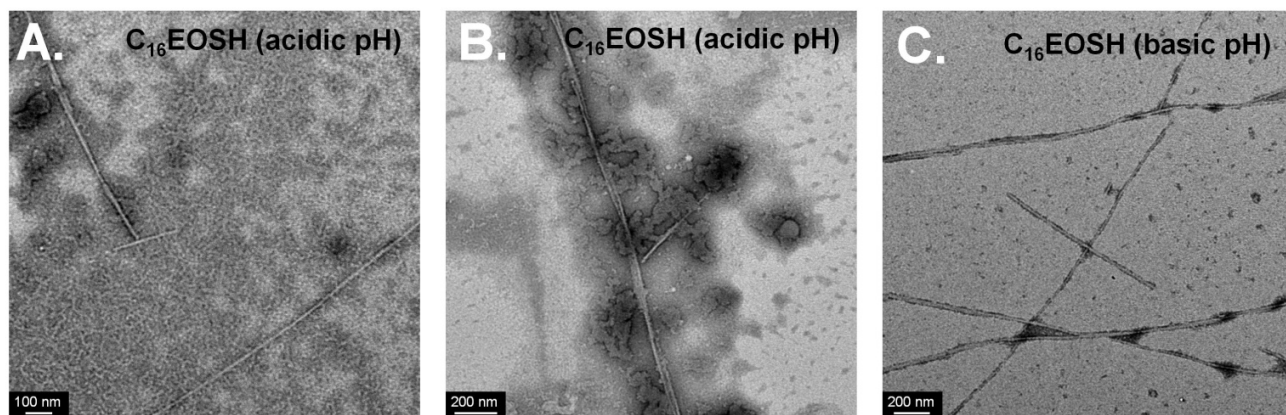
**Supplemental Information 1B.** MALDI MS C<sub>16</sub>EOSH.

The initial concept was to develop a hydrogel system by combining two dissimilar PAs. One PA was palmitoyl-trihistidine and the other palmitoyl-triserine. After the PAs were synthesized employing SPPS, it was realized that the palmitoyl-triserine counterpart formed a strong gel in aqueous solvents across a wide pH and temperature range, aqueous/ organic mixtures, and organic solvents. The gelation prohibited proper purification and characterization (i.e. mass spectrometry) of the material. The palmitoyl-trihistidine counterpart was easily soluble in several solvent conditions. This led to the hypothesis that if both peptides were conjugated onto the same molecule, the resulting peptide amphiphile would have enhanced solubility over the palmitoyl-triserine molecule. We later learned that the branched peptide amphiphile was indeed easily soluble.

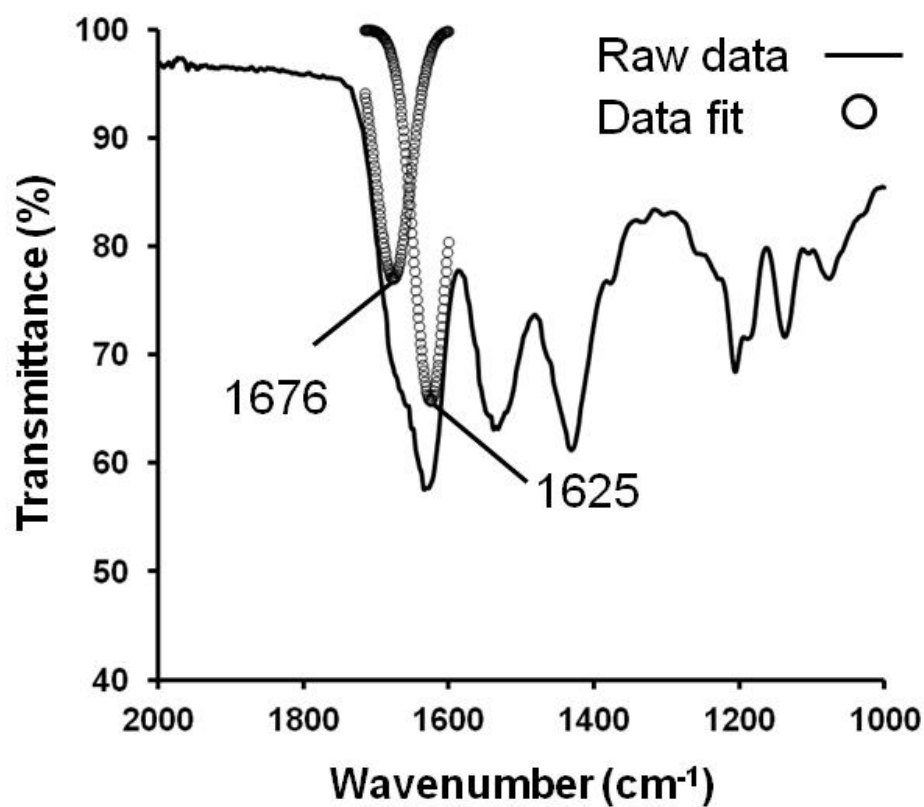
**Supplemental Information 2.** Description of two-component PA hydrogel.



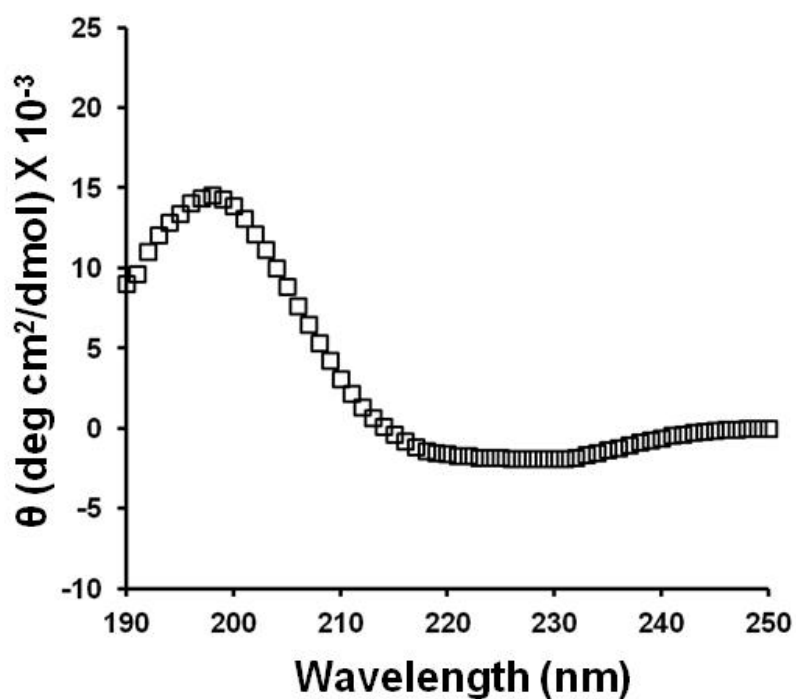
**Supplemental Information 3.** 2 mg/ml  $C_{16}$ GSH were cast in glass molds. The samples were raised above pH 6.5 using a minimal amount of NaOH. (A) The resulting self-supporting hydrogel after removal of glass mold above pH 6.5. (B) A minimal amount of HCl was used to reduce the pH of a self-supporting hydrogel transitioning it back to a liquid. Phase changes occur within seconds to minutes after the addition of acid/base.



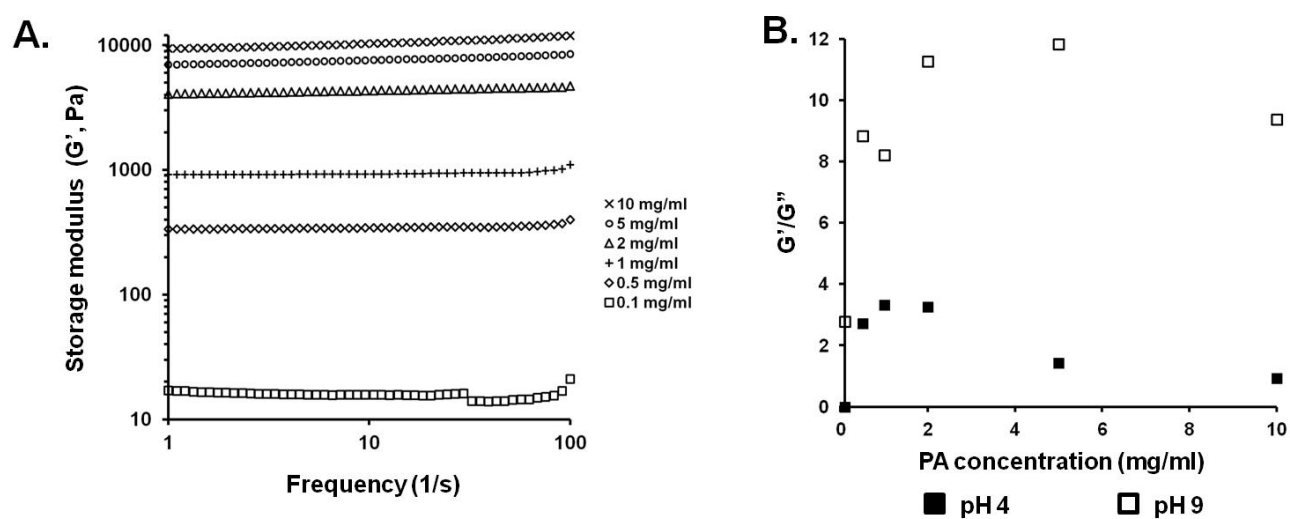
**Supplemental Information 4.** Self-assemblies of C<sub>16</sub>EOSH. (A-B) Negative stain of self-assemblies in acidic pH. (C) Negative stain of self-assemblies in basic pH.



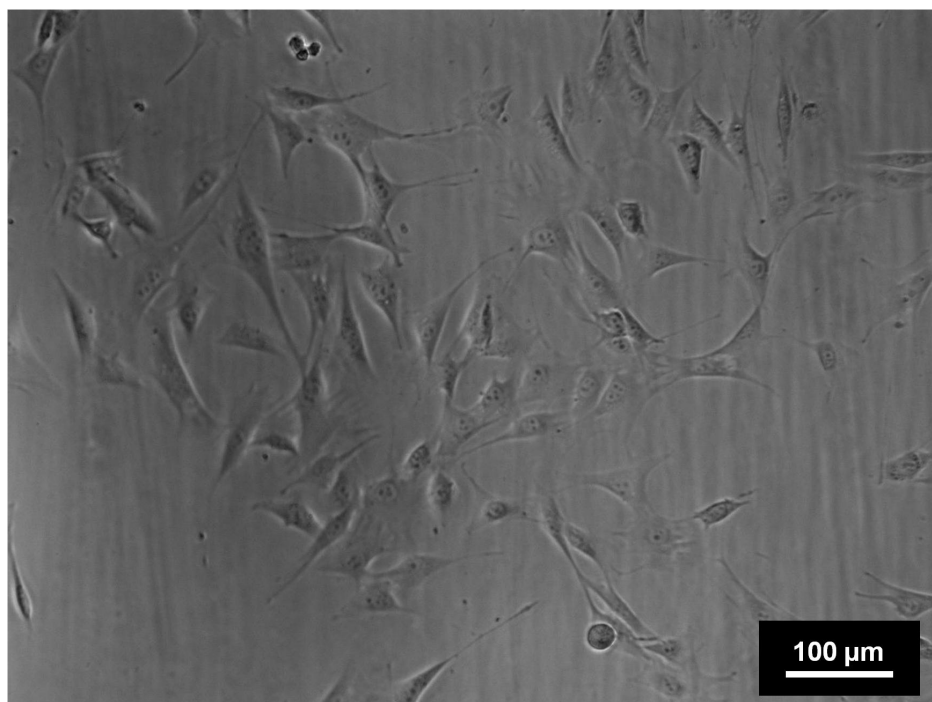
**Supplemental Information 5.** IR spectrum of C<sub>16</sub>GSH hydrogel. IR confirmed the formation of  $\beta$ -sheets with amide I frequencies at 1676 cm<sup>-1</sup> (parallel band) and 1625 cm<sup>-1</sup> (perpendicular band). Peak deconvolution performed with Fityk.



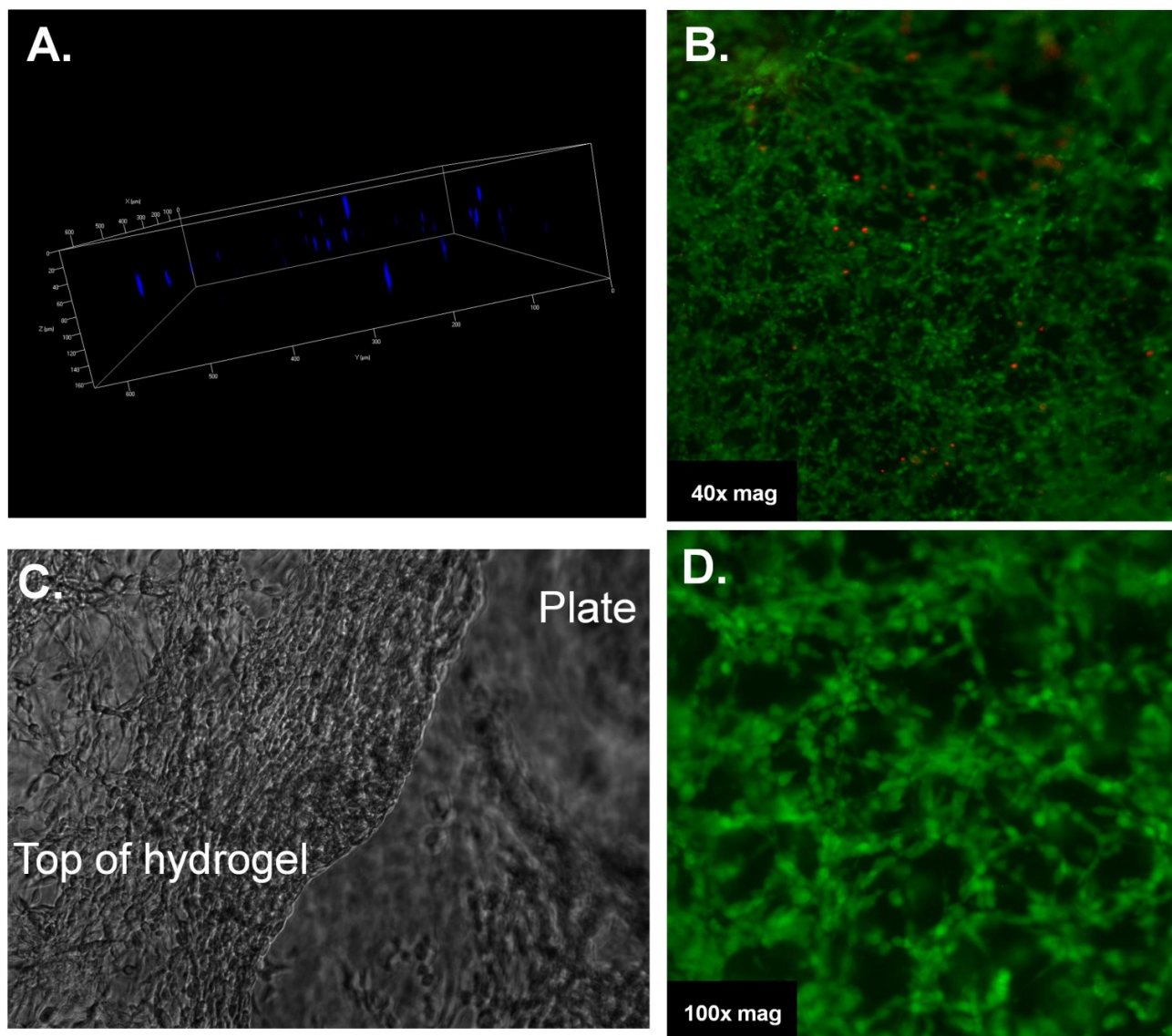
**Supplemental Information 6.** CD determined that C<sub>16</sub>GSH formed  $\beta$ -sheets at pH 7.0. This experiment was performed by carefully raising the pH of an acidic PA solution to pH 7.0 without overshooting. Sample prepared at 150  $\mu$ M.



**Supplemental Information 7.** C16GSH frequency sweep data and the effect of  $G'/G''$  ratio with increased pH. (A) Complete frequency sweep data of C<sub>16</sub>GSH at pH 9, corresponds to data in Figure 5. (B) The hydrogel became more solid-like as the pH increased determined by the increased ratio of  $G'/G''$ .



**Supplemental Information 8.** Typical morphology of non-confluent layer of NIH 3T3 fibroblasts after 24 hours when cultured on polystyrene plates. 100x magnification.



**Supplemental Information 9.** Hydrogels support cell growth in three-dimensions.

Images are cells cultured for 96 hours. (A) Z-stack image of Hoechst 33342 stained cell nuclei (blue). The elongated fluorescence in the Z-direction was due to scattering. (B) Live/Dead assay of cells. Live cells are in green and punctate red spots are dead cells. (C) Cells grown near an edge of the hydrogel, 100x magnification. (D) Calcein AM stained fibroblasts grown on a hydrogel. Note cells appear to grow along fibers of hydrogels and not on a flat surface.