Supporting Information

Multicomponent Supramolecular Hydrogels Composed of Cationic Phenylalanine Derivatives and Anionic Amino Acids

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Table S1. Procedure for preparing assemblies of compounds **1**–**3** with monosodium salts of either glutamic acid or aspartic acid. All hydrogels have a final concentration of compounds **1**–**3** of 15 mM.

Equivalents of anionic amino acid with respect to Fmoc-Phe-DAP derivative (compounds **1**, **2**, or **3**)	Final concentration of anionic amino acid (mM)	Volume of stock solution of anionic amino acid (500 mM) added to solution of compound **1**, **2**, or **3** (μL)	Volume of water added (μL)
1	15	30	470
2	30	60	440
3	45	90	410
4	60	120	380
5	75	150	350
6	90	180	320
7	105	210	290
8	120	240	260
9	135	270	230
10	150	300	200

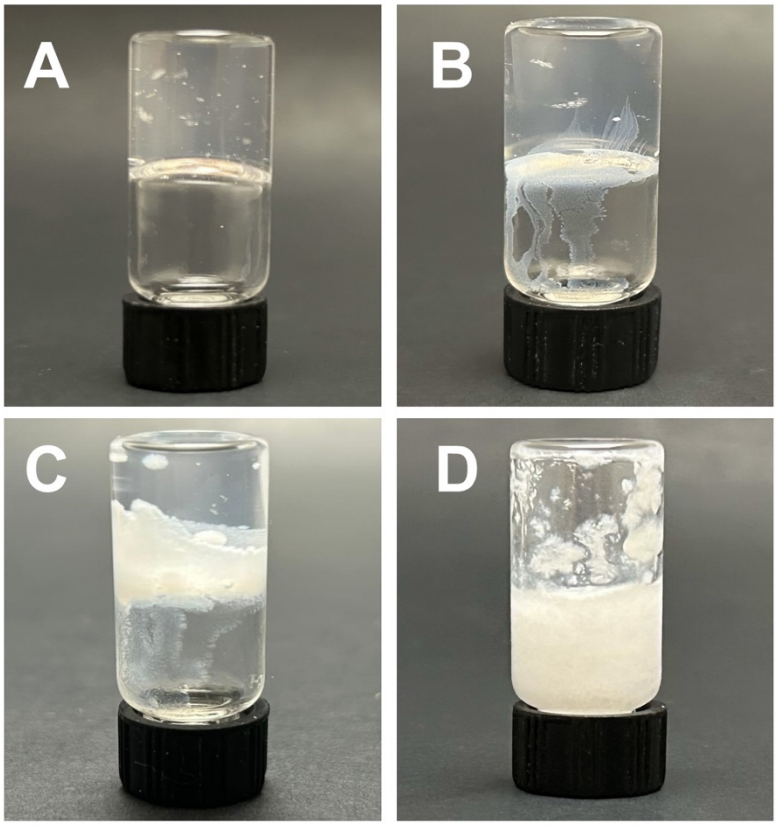

Figure S1. Digital images of assemblies of Fmoc-3F-Phe-DAP (**2**)/aspartate mixtures at different time intervals. These images show the progression from a hydrogel to a precipitate over 12 hours. **A.** 5 minutes after gelation, **B.** 30 minutes after gelation, **C.** 2 hours after gelation, and **D.** 12 hours after gelation.

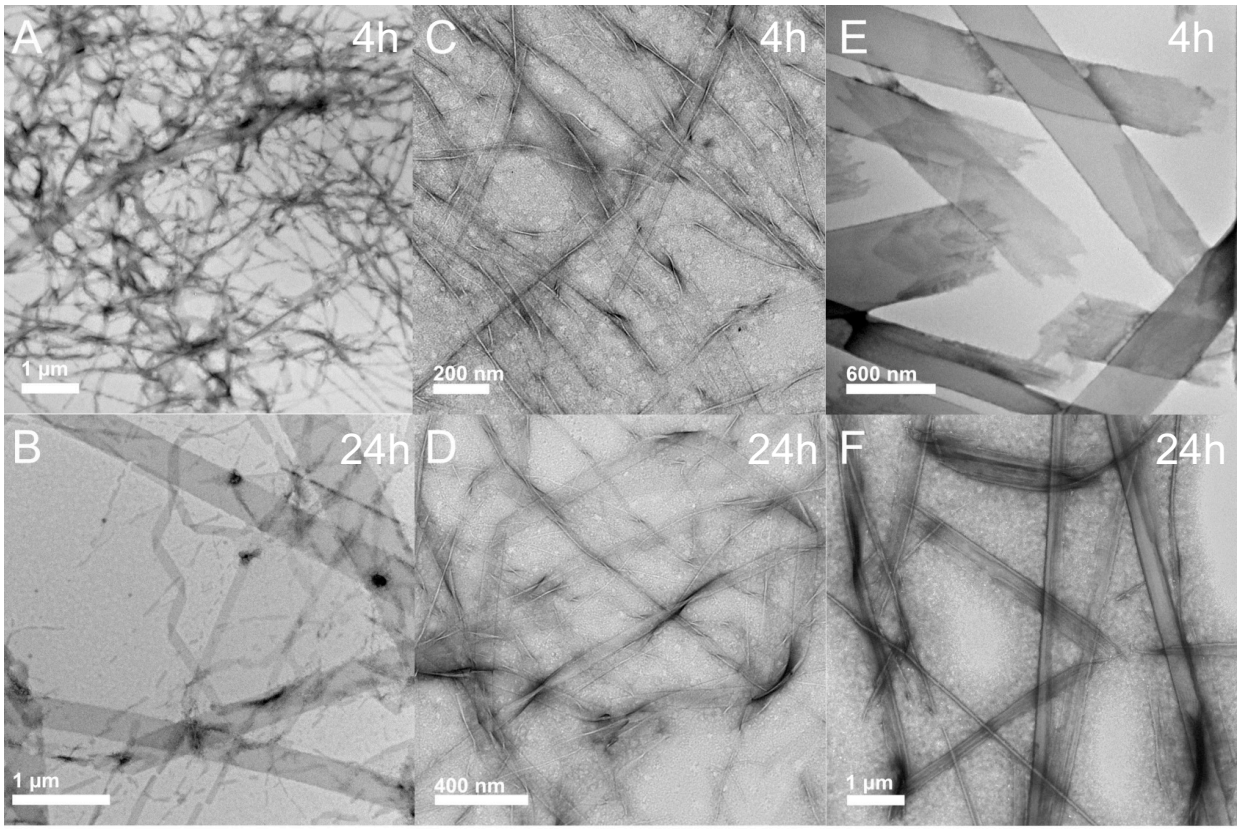

Figure S2. TEM images of Fmoc-Phe-DAP (**1**) assembled with NaCl (114 mM), glutamate (7 equivalents), or aspartate (7 equivalents) ions after 4 hours and 24 hours. **A.** Fmoc-Phe-DAP (**1**)/NaCl after 4 hours, **B.** Fmoc-Phe-DAP (**1**)/NaCl after 24 hours, **C.** Fmoc-Phe-DAP (**1**)/glutamate after 4 hours, **D.** Fmoc-Phe-DAP (**1**)/glutamate after 24 hours, **E.** Fmoc-Phe-DAP (**1**)/aspartate after 4 hours, **F.** Fmoc-Phe-DAP (**1**)/aspartate after 24 hours.

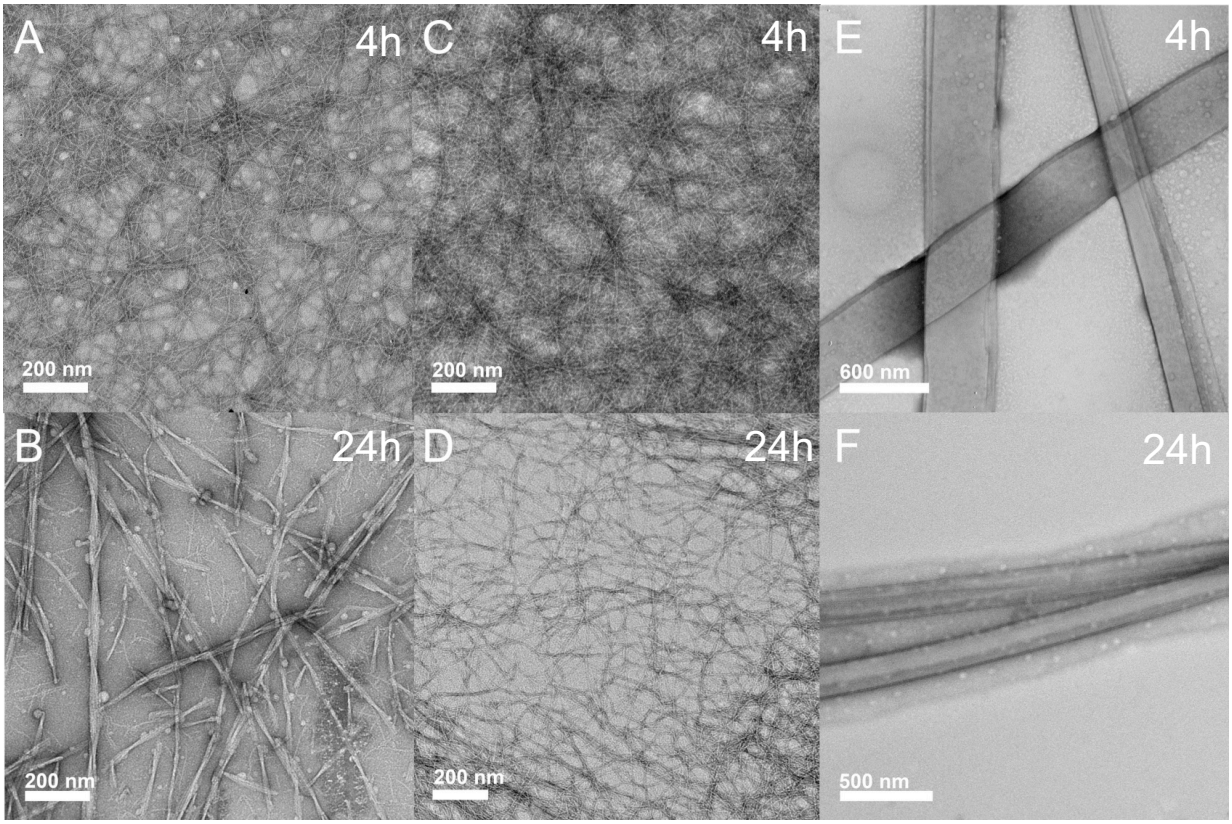

Figure S3. TEM images of Fmoc-3F-Phe-DAP (**2**) assembled with NaCl (114 mM), glutamate (7 equivalents), or aspartate (7 equivalents) ions after 4 hours and 24 hours. **A.** Fmoc-3F-Phe-DAP (**2**)/NaCl after 4 hours, **B.** Fmoc- 3F-Phe-DAP (**2**)/NaCl after 24 hours, **C.** Fmoc-3F-Phe-DAP (**2**)/glutamate after 4 hours, **D.** Fmoc-3F-Phe-DAP (**2**)/glutamate after 24 hours, **E.** Fmoc-3F-Phe-DAP (**2**)/aspartate after 4 hours, **F.** Fmoc-3F-Phe-DAP (**2**)/aspartate after 24 hours.

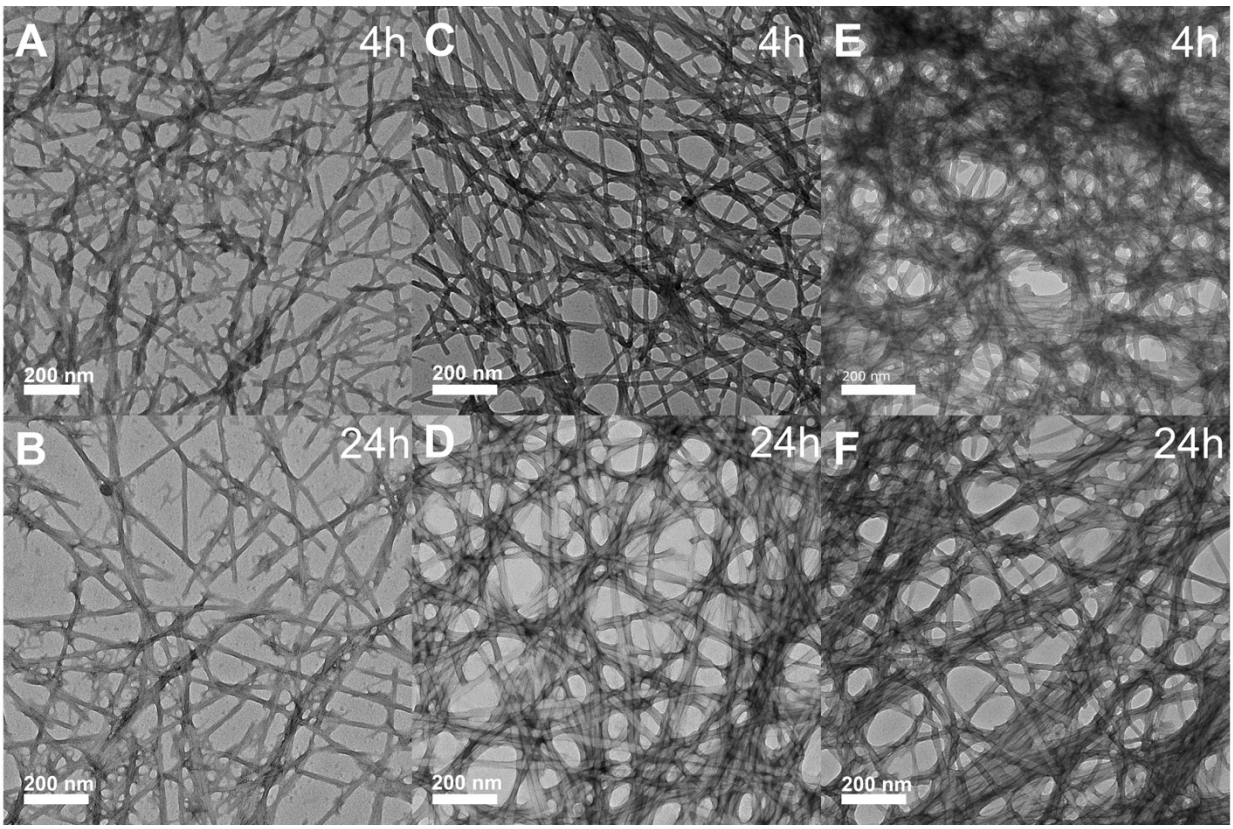

Figure S4. TEM images of Fmoc-F_5-Phe-DAP (**3**) assembled with NaCl (114 mM), glutamate (7 equivalents), or aspartate (7 equivalents) ions after 4 hours and 24 hours. **A.** Fmoc-F_5-Phe-DAP (**3**)/NaCl after 4 hours, **B.** Fmoc-F_5-Phe-DAP (**3**)/NaCl after 24 hours, **C.** Fmoc-F_5-Phe-DAP (**3**)/glutamate after 4 hours, **D.** Fmoc-F_5-Phe-DAP (**3**)/glutamate after 24 hours, **E.** Fmoc-F_5-Phe-DAP (**3**)/aspartate after 4 hours, **F.** Fmoc-F_5-Phe-DAP (**3**)/aspartate after 24 hours.

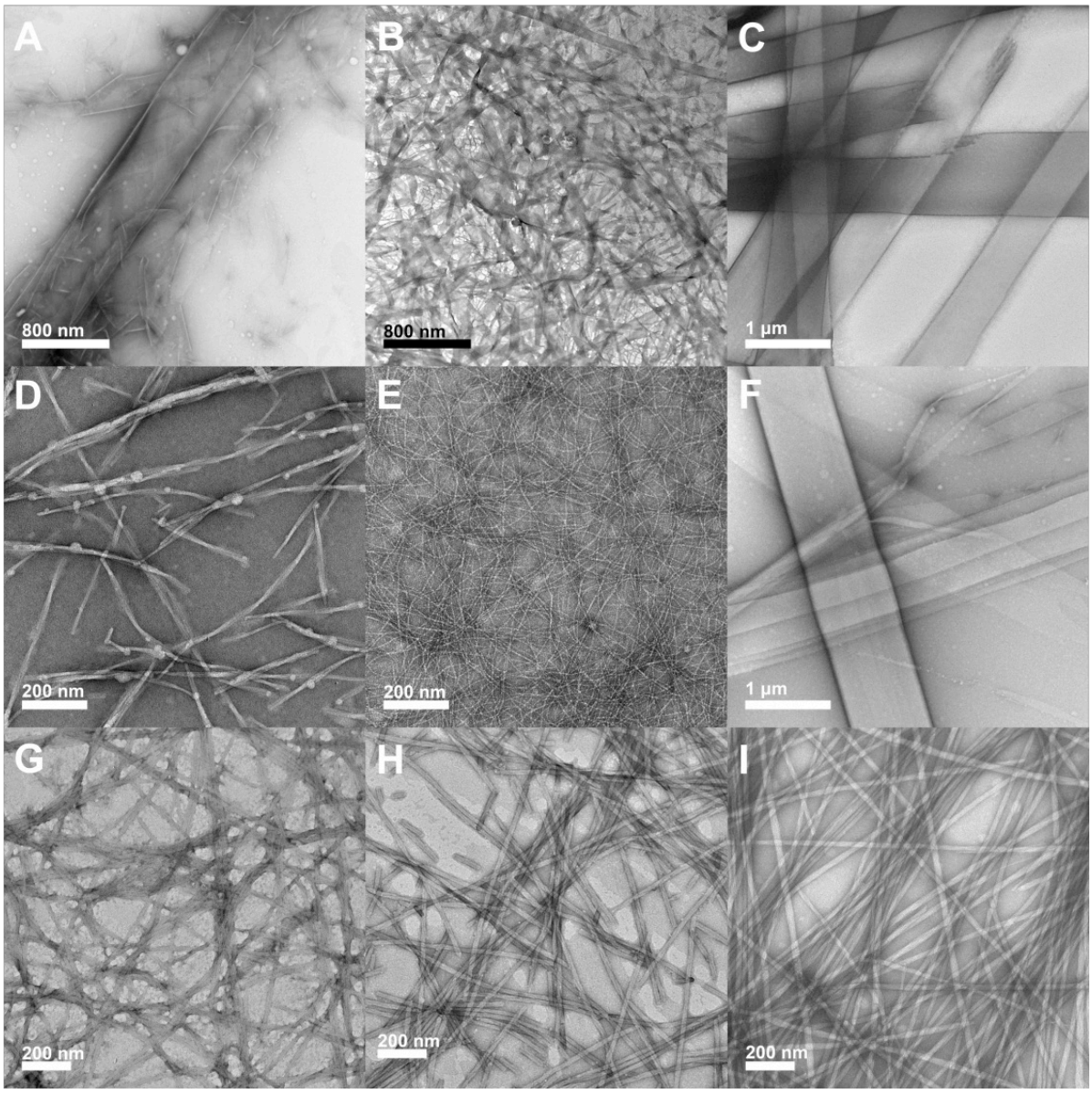

Figure S5. TEM images of assemblies of Fmoc-Phe-DAP derivatives after 7 days. All assemblies are mixtures of compounds **1**, **2**, or **3** (15 mM) and either 114 mM NaCl or 7 equivalents of monosodium glutamate or monosodium aspartate. **A.** Fmoc-Phe-DAP (**1**)/NaCl, **B.** Fmoc-Phe-DAP (**1**)/glutamate **C.** Fmoc-Phe-DAP (**1**)/aspartate, **D.** Fmoc-3F-Phe-DAP (**2**)/NaCl, **E.** Fmoc-3F-Phe-DAP (**2**)/glutamate, **F.** Fmoc-3F-Phe-DAP (**2**)/aspartate, **G.** Fmoc-F_5-Phe-DAP (**3**)/NaCl, **H.** Fmoc-F_5-Phe-DAP (**3**)/glutamate, **I.** Fmoc-F_5-Phe-DAP (**3**)/aspartate.

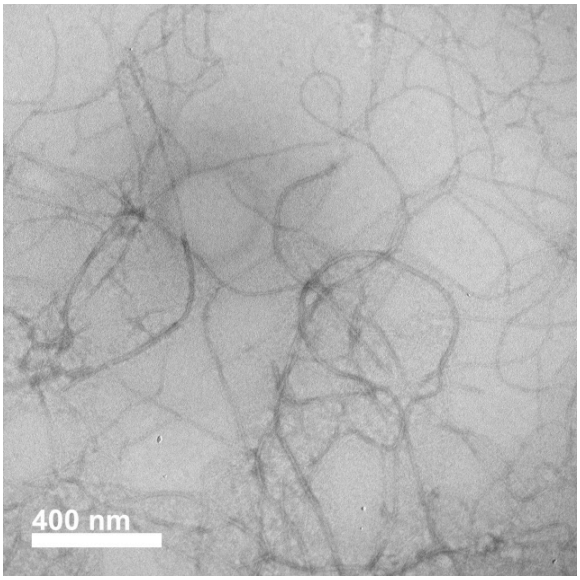

Figure S6. TEM image of Fmoc-3F-Phe DAP (**2**) with 7 equivalents of aspartate after 4 hours that shows the minor fibrous/wormlike micelle constituents of these mixtures.

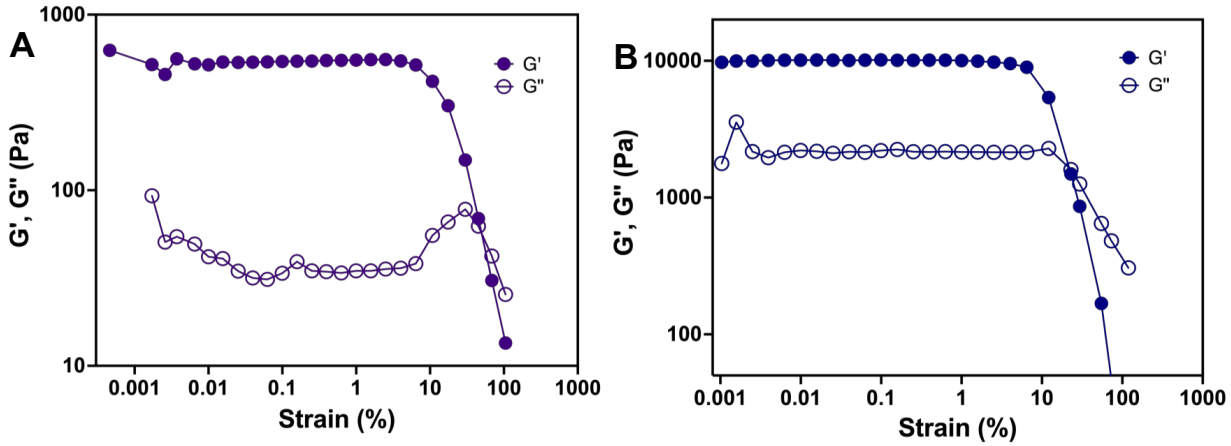

Figure S7. Oscillatory rheology amplitude sweep data for **A.** hydrogels of Fmoc-3F-Phe-DAP (**2**) (15 mM) with 7 equivalents of glutamate, and **B.** hydrogels of Fmoc-3F-Phe-DAP (**2**) (15 mM) with NaCl (114 mM).

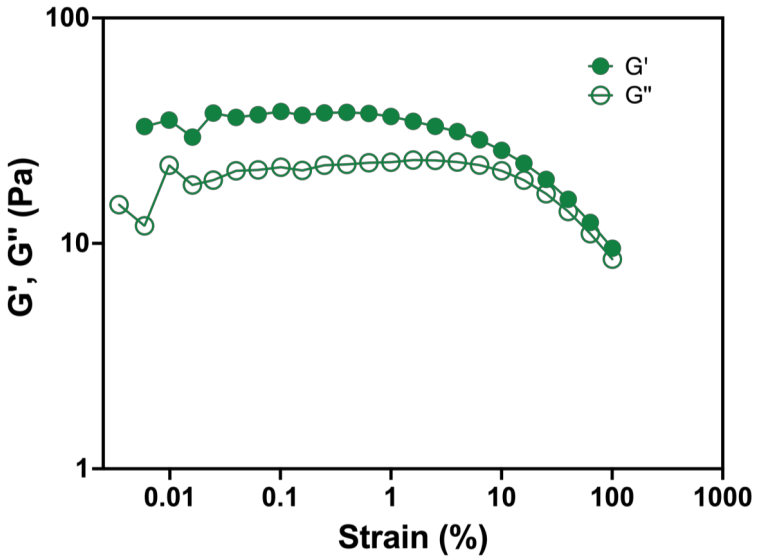

Figure S8. Oscillatory rheology amplitude sweep data for a hydrogel of Fmoc-Phe-DAP (**1**) (15 mM) with 7 equivalents of glutamate.[#]

It should be noted that while we performed this amplitude sweep experiment for hydrogels of Fmoc-Phe-DAP (**1**) (15 mM) with 7 equivalents of glutamate, these hydrogels were exceptionally weak. The raw phase angle for these hydrogels in this data is greater than 175° for several data points, indicative of instrumental inertia dominating the signal. These data should thus be viewed with caution since they are not strictly reliable indicators of the properties of the material only.

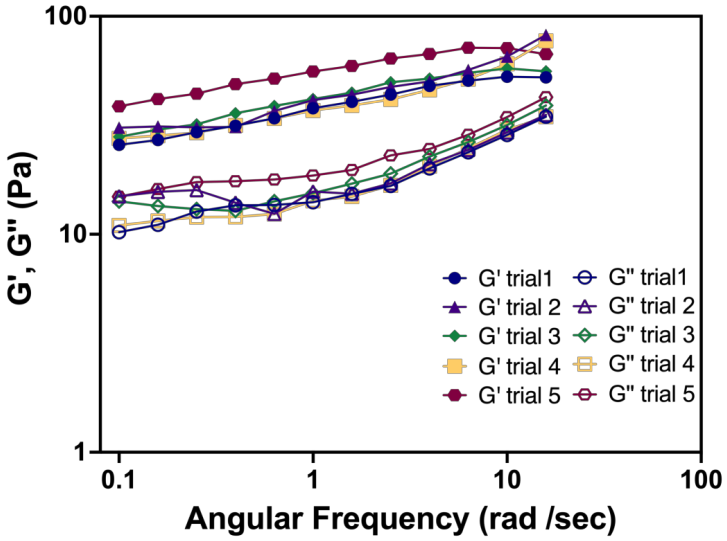

Figure S9. Oscillatory rheology frequency sweep plot for hydrogels of Fmoc-Phe-DAP (**1**) with 7 equivalents of monosodium glutamate.

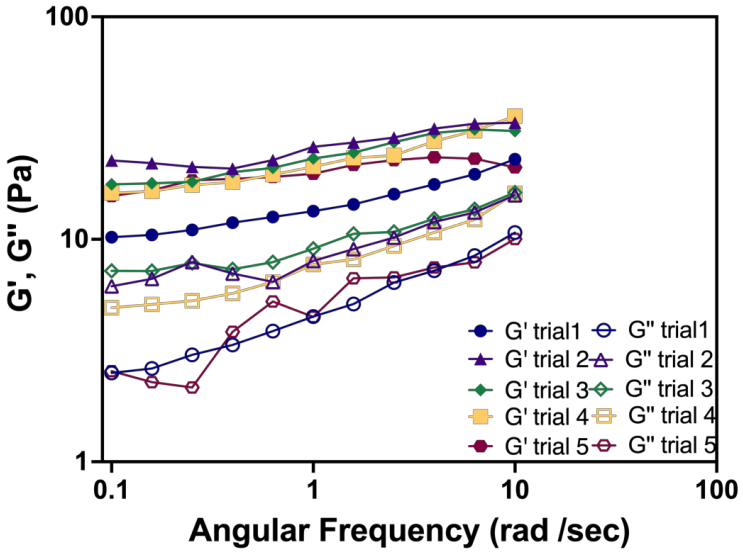

Figure S10. Oscillatory rheology frequency sweep plot for hydrogels of Fmoc-3F-Phe-DAP (**2**) with 3 equivalents of monosodium glutamate.