

## Supporting Information Section for “Multi-component Peptide Hydrogels – A Systematic Study Incorporating Biomolecules for the Exploration of Diverse, Tuneable Biomaterials”

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**Scheme S1:** Reaction scheme of the synthesis of C14-FF.

**Figure S1:** <sup>1</sup>HNMR spectrum of C14-FF.

**Figure S2:** <sup>13</sup>CNMR spectrum of C14-FF.

**Figure S3:** FT-IR spectrum of C14-FF.

**Figure S4:** ESI-MS spectrum of C14-FF.

**Figure S5:** Graph showing the increase in stiffness (G'; storage modulus) of the hydrogel as the concentration of C14-FF increases.

**Figure S6:** Histogram of fiber thicknesses for C14-FF from TEM method.

**Figure S7:** Histogram of fiber thicknesses for C14-FF from AFM method.

**Figure S8:** CD spectra of ThT assay of C14-FF compound in non-gelling solvents; a) MeOH, b) ACN.

**Figure S9:** Frequency sweep rheology experiments on multi-component hydrogels. a) C14-FF + collagen, b) C14-FF + glucose, c) C14-FF+ glutamine, d) C14-FF + mix 1, e) C14-FF + mix 2, f) C14-FF + mix 3, g) C14-FF + NAG+GA, h) C14-FF + polylysine, i) C14-FF + starch, j) C14-FF + vitamins.

**Figure S10:** Step-strain rheology experiments on multi-component hydrogels. a) C14-FF + collagen, b) C14-FF + glucose, c) C14-FF+ glutamine, d) C14-FF + mix 1, e) C14-FF + mix 2, f) C14-FF + mix 3, g) C14-FF + NAG+GA, h) C14-FF + polylysine, i) C14-FF + starch, j) C14-FF + vitamins.

**Figure S11:** Histograms of fiber widths for multi-component hydrogels using AFM. a) C14-FF + collagen, b) C14-FF + glucose, c) C14-FF+ glutamine, d) C14-FF + mix 1, e) C14-FF + mix 2, f) C14-FF + mix 3, g) C14-FF + NAG+GA, h) C14-FF + polylysine, i) C14-FF + starch, j) C14-FF + vitamins.

**Figure S12:** Additional TEM images of multi-component hydrogels. a) C14-FF + collagen, b) C14-FF + glucose, c) C14-FF+ glutamine, d) C14-FF + mix 1, e) C14-FF + mix 2, f) C14-FF + mix 3, g) C14-FF + NAG+GA, h) C14-FF + polylysine, i) C14-FF + starch, j) C14-FF + vitamins.

**Figure S13:** Additional AFM images of multi-component hydrogels. a) C14-FF + collagen, b) C14-FF + glucose, c) C14-FF+ glutamine, d) C14-FF + mix 1, e) C14-FF + mix 2, f) C14-FF + mix 3, g) C14-FF + NAG+GA, h) C14-FF + polylysine, i) C14-FF + starch, j) C14-FF + vitamins.

**Figure S14:** Fluorescence spectra of multi-component hydrogels with ThT. a) ThT only, b) C14-FF + Collagen, c) C14-FF + Glucose, d) C14-FF + Glutamine, e) C14-FF + Mix 1, f) C14-FF + Mix 2, g) C14-FF + Mix 3, h) C14-FF + NAG+GA, i) C14-FF + Polylysine, j) C14-FF + Starch, k) C14-FF + Vitamins.

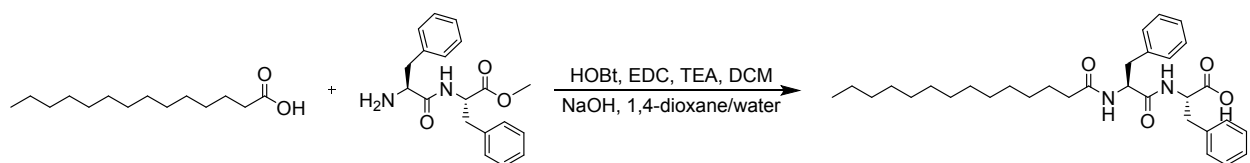
**Figure S15:** CD data of multi-component hydrogels with and without ThT a) C14-FF + collagen, b) C14-FF + glucose, c) C14-FF+ glutamine, d) C14-FF + mix 1, e) C14-FF + mix 2, f) C14-FF + mix 3, g) C14-FF + NAG+GA, h) C14-FF + polylysine, i) C14-FF + starch, j) C14-FF + vitamins.

**Figure S16:** TEM images of the 0.05% w/v C14-FF hydrogel after immediately cooling the hot solution in an ice bath to obtain hydrogel state in less time demonstrating shorter fibre lengths compared to the gradual cooling at room temperature.

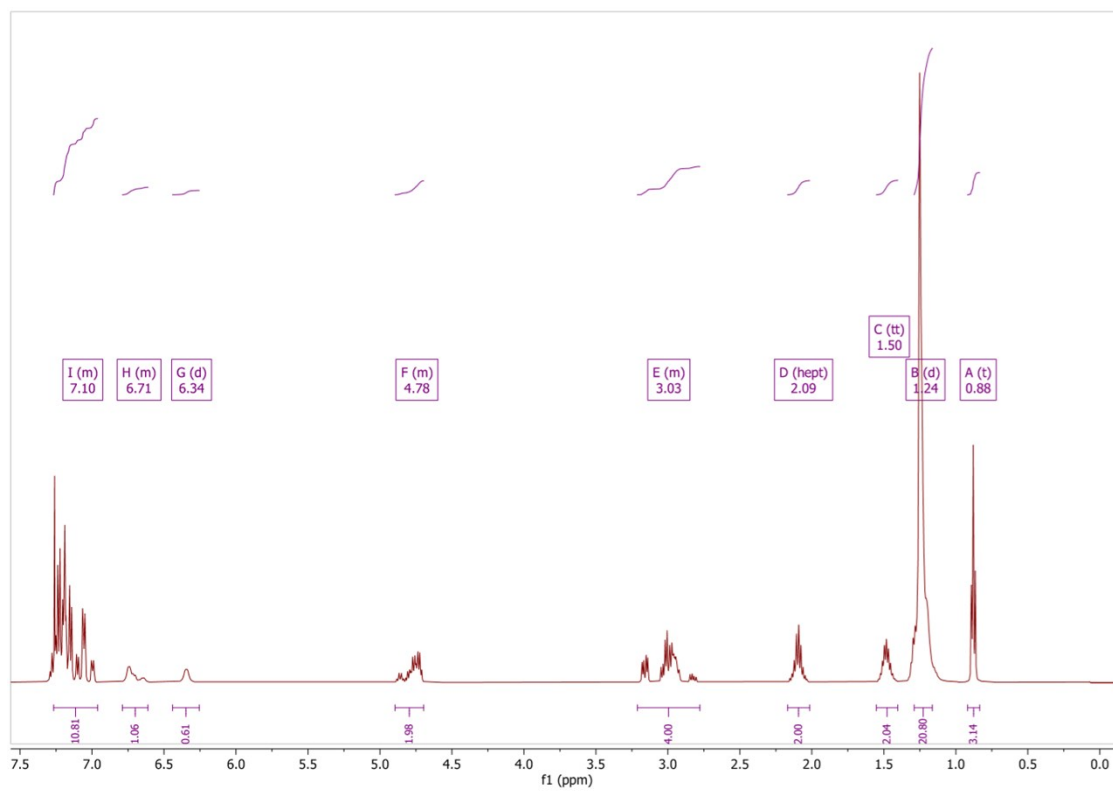
**Figure S17:** TEM images of multi-component gels after preparing them through an alternate method where the both gelator and component powders are placed together, buffer is added and then the solution was heated until everything dissolved and slowly cooled until hydrogel forms. a-c) C14-FF + starch d-f) C14-FF + collagen.

**Figure S18:** Frequency sweep rheology of multi-component gels using alternate method of mixing. a) C14-FF + starch, b) C14-FF + collagen.

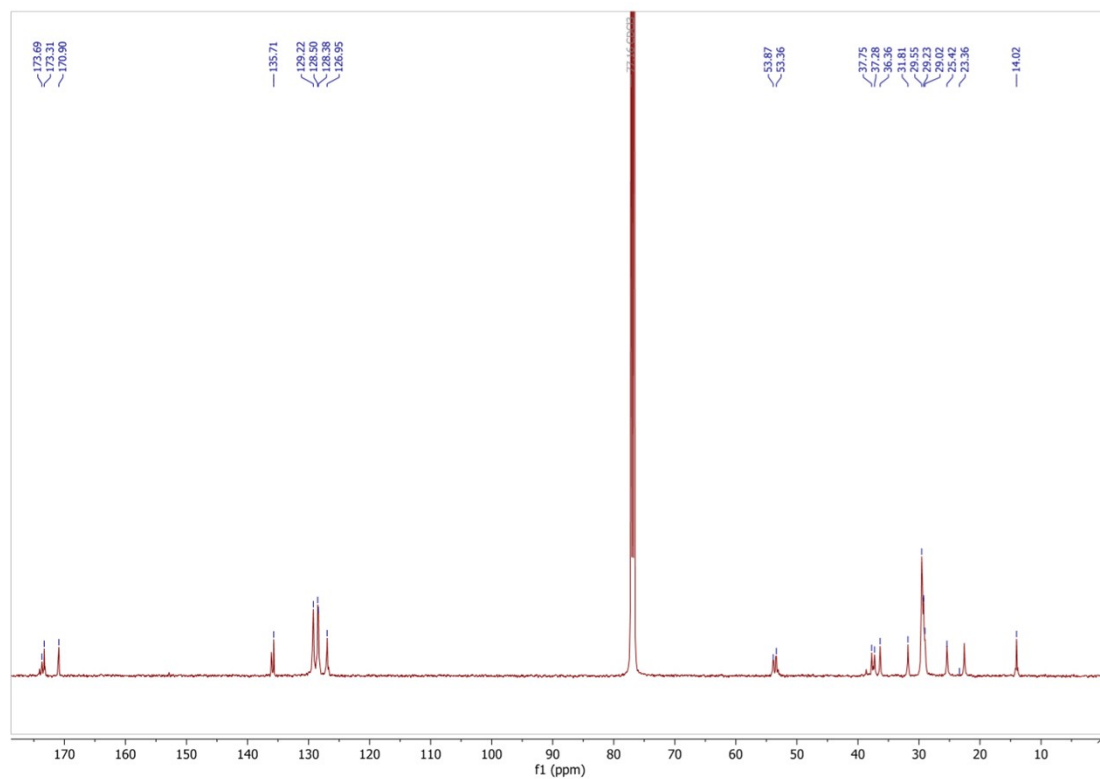
**Table S1:** Change in matrix properties upon addition of components.



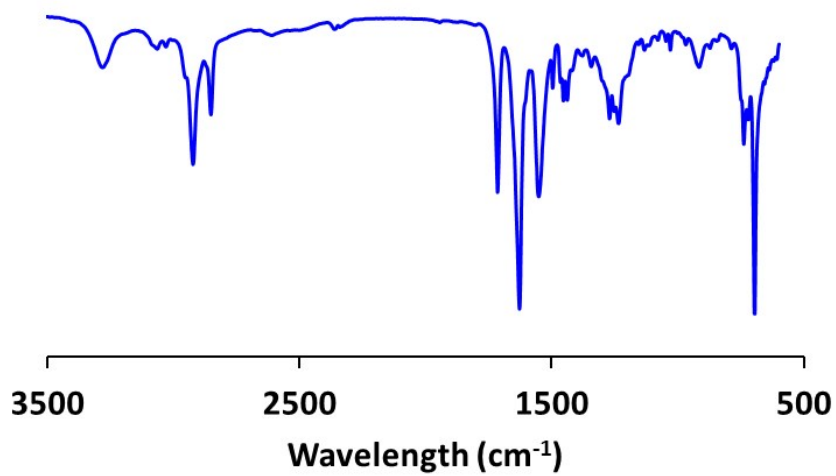
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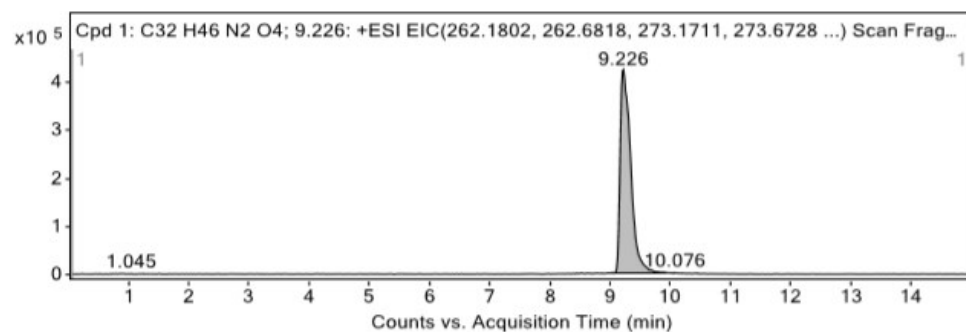
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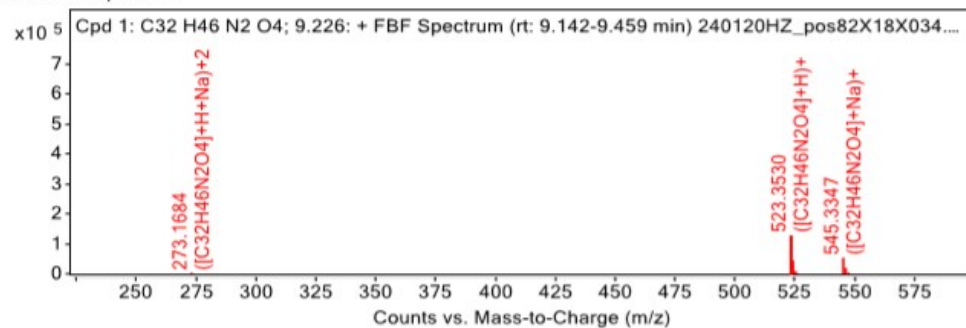
**Figure S2:** <sup>13</sup>CNMR spectrum of C14-FF.



**Figure S3:** FT-IR spectrum of C14-FF.



MS Zoomed Spectrum



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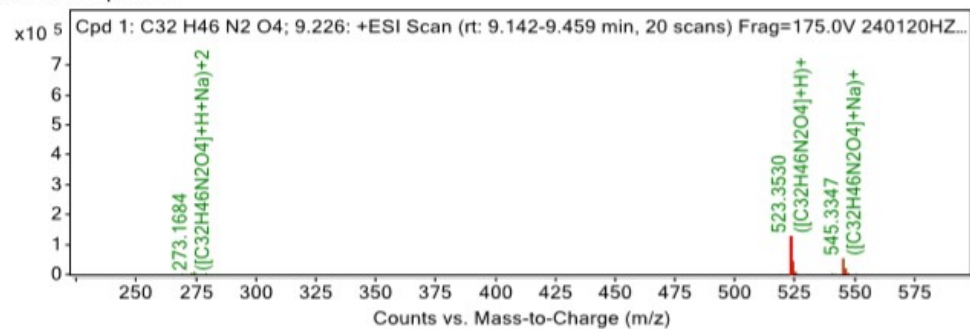
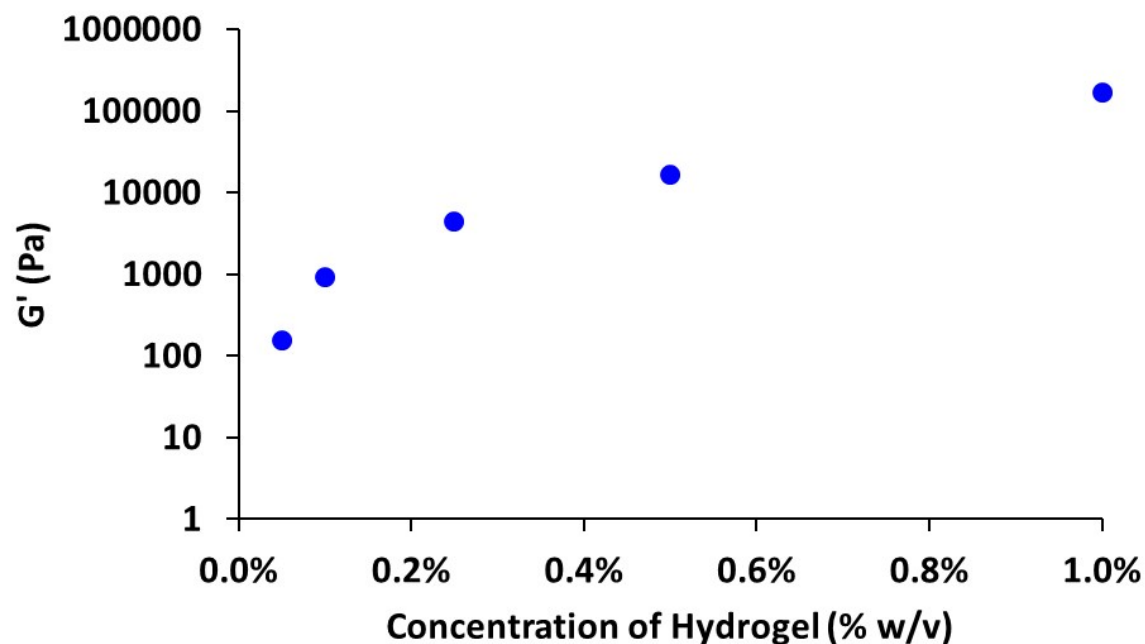
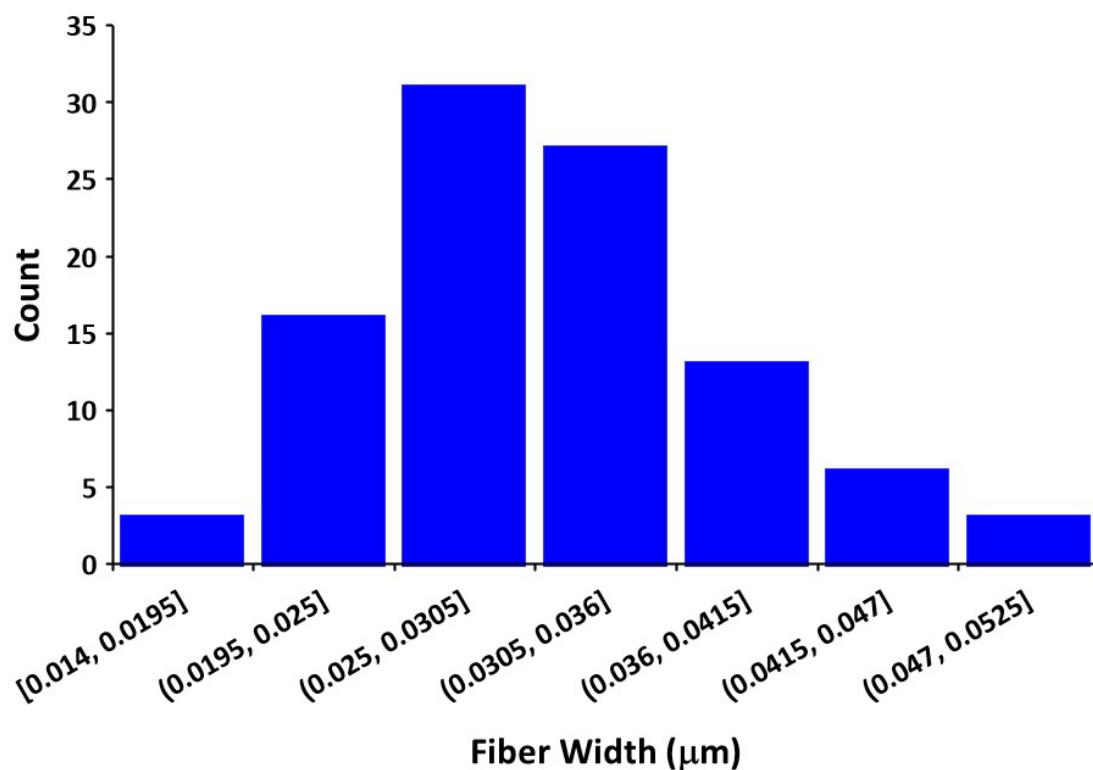


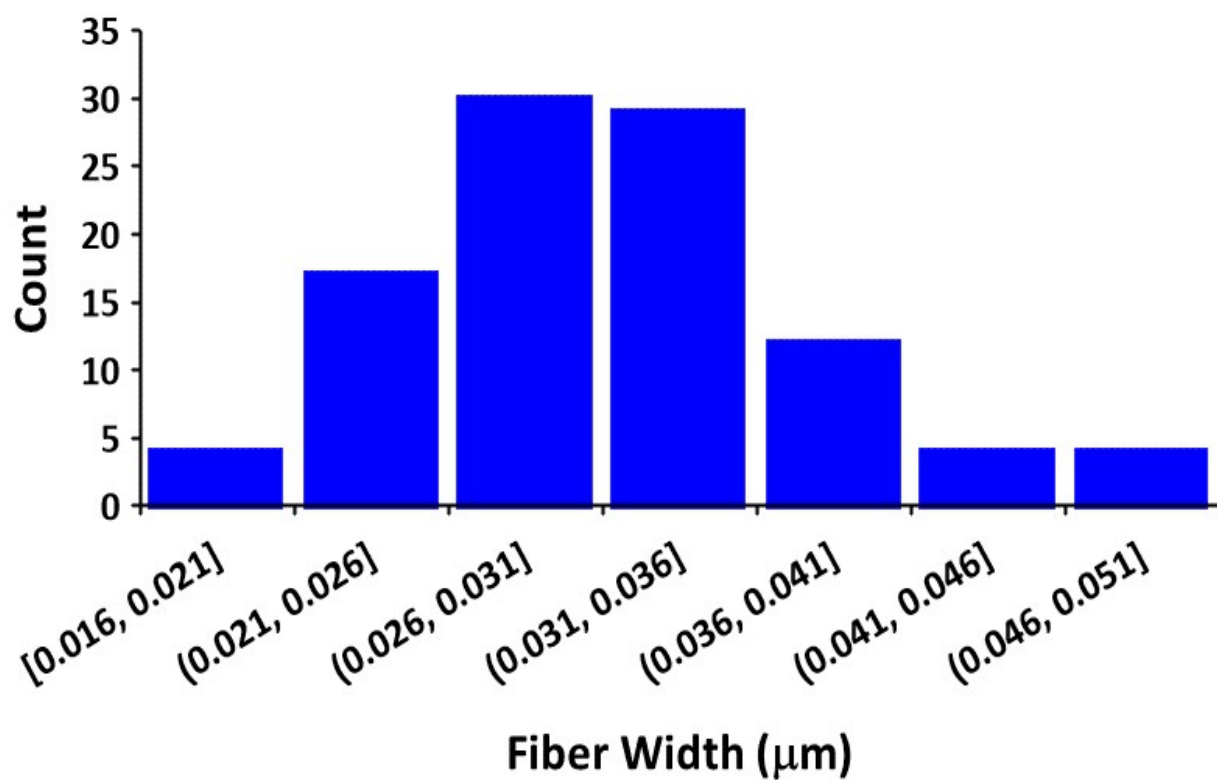
Figure S4: ESI-MS spectrum of C14-FF.



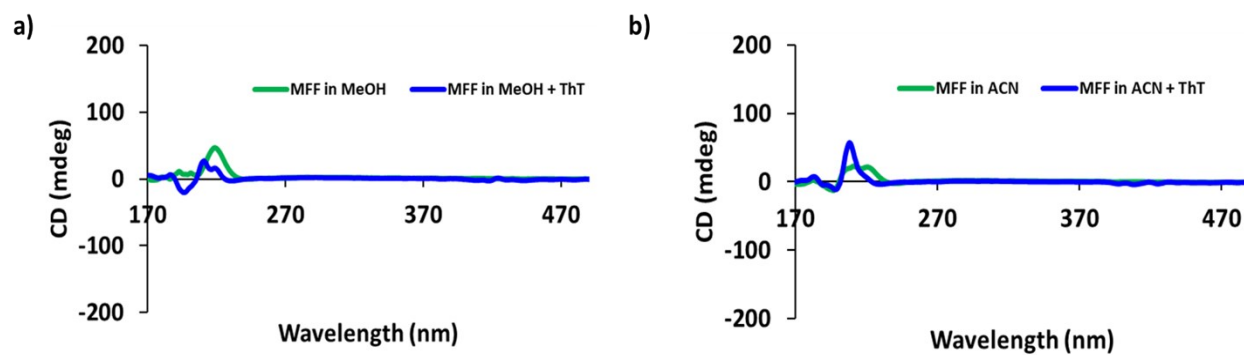
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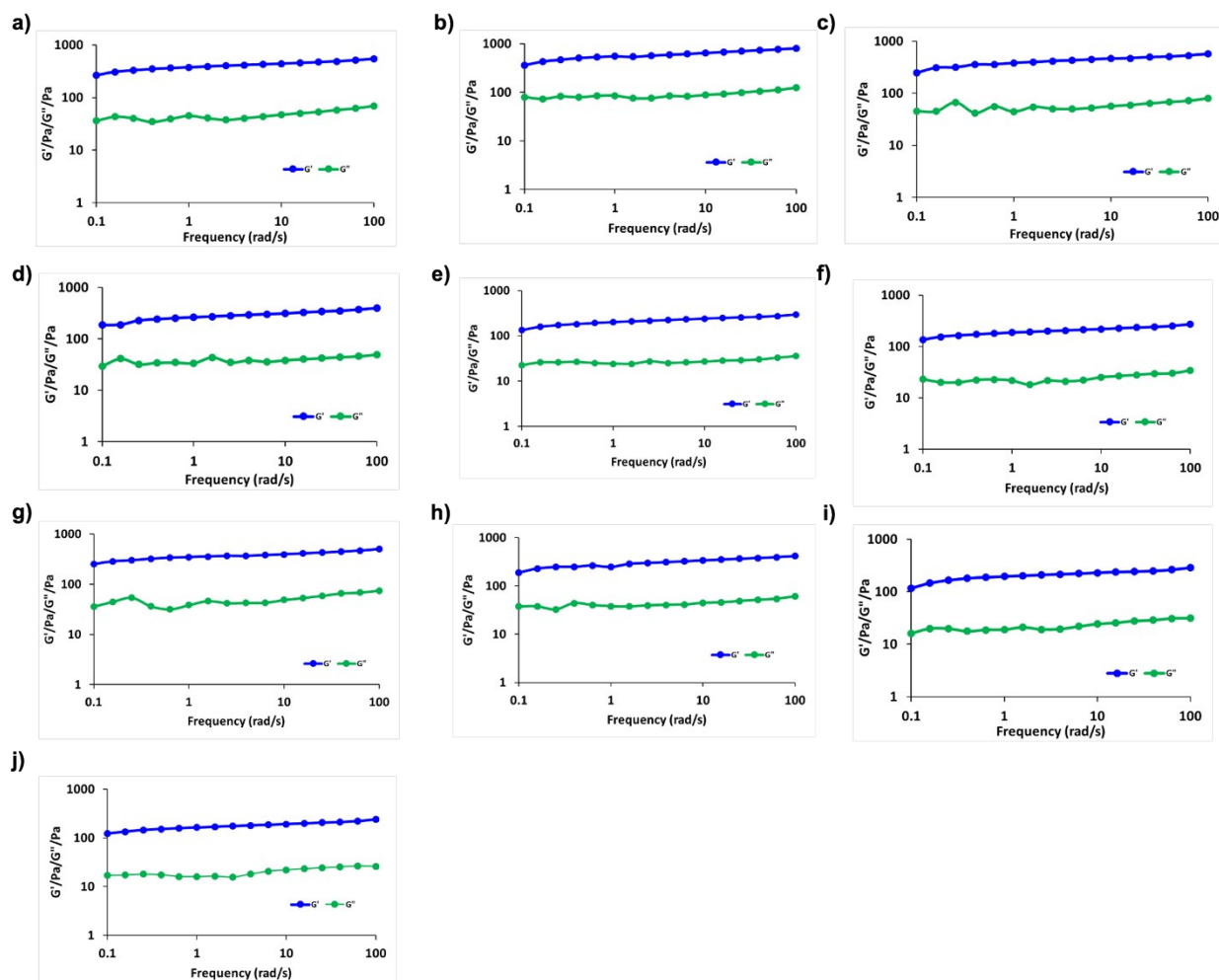
**Figure S6:** Histogram of fiber thicknesses for C14-FF from TEM method.



**Figure S7:** Histogram of fiber thicknesses for C14-FF from AFM method.

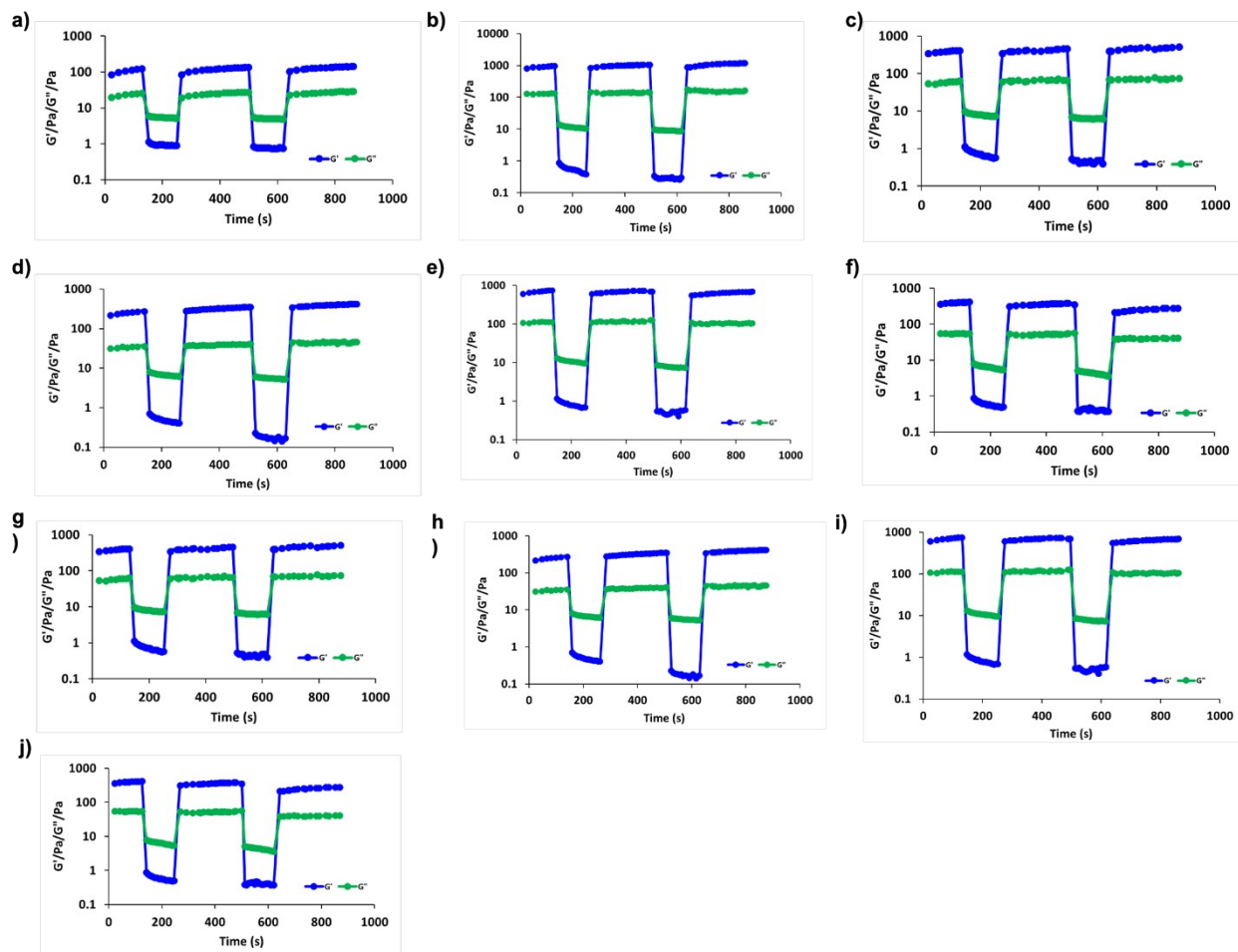


**Figure S8:** CD spectra of ThT assay of C14-FF compound in non-gelling solvents; a) MeOH, b) ACN.

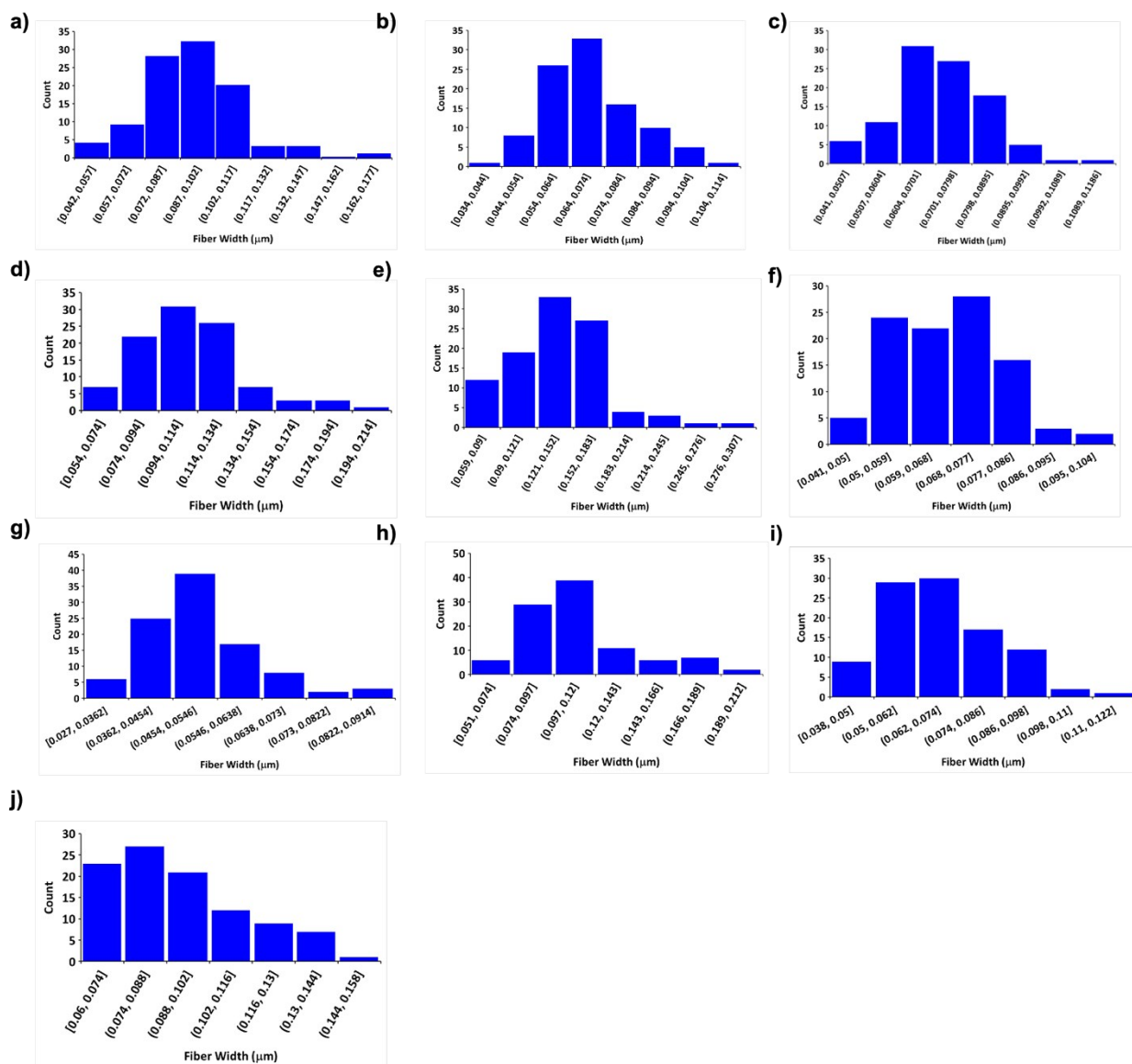


**Figure S9:** Frequency sweep rheology experiments on multi-component hydrogels. a) C14-FF + collagen, b) C14-FF + glucose, c) C14-FF + glutamine, d) C14-FF + mix 1, e) C14-FF + mix 2, f) C14-FF + mix 3, g) C14-FF + NAG+GA, h) C14-FF + polylysine, i) C14-FF + starch, j) C14-FF + vitamins.

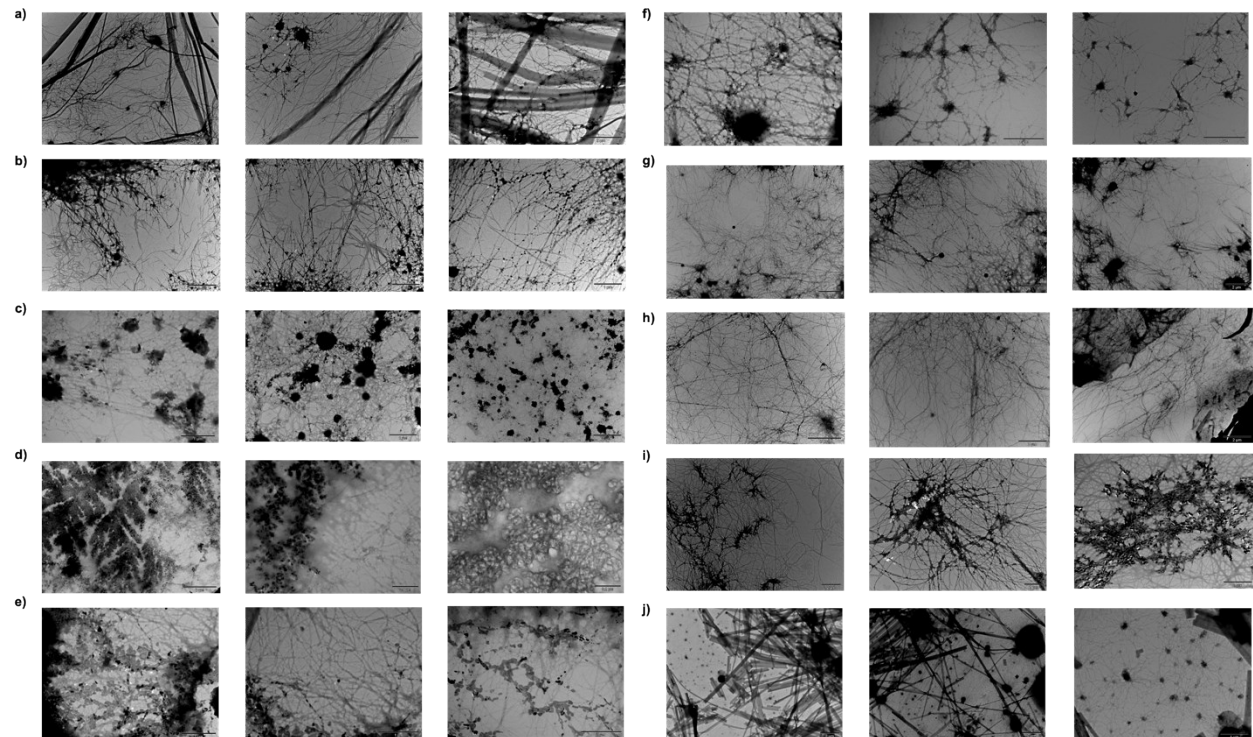




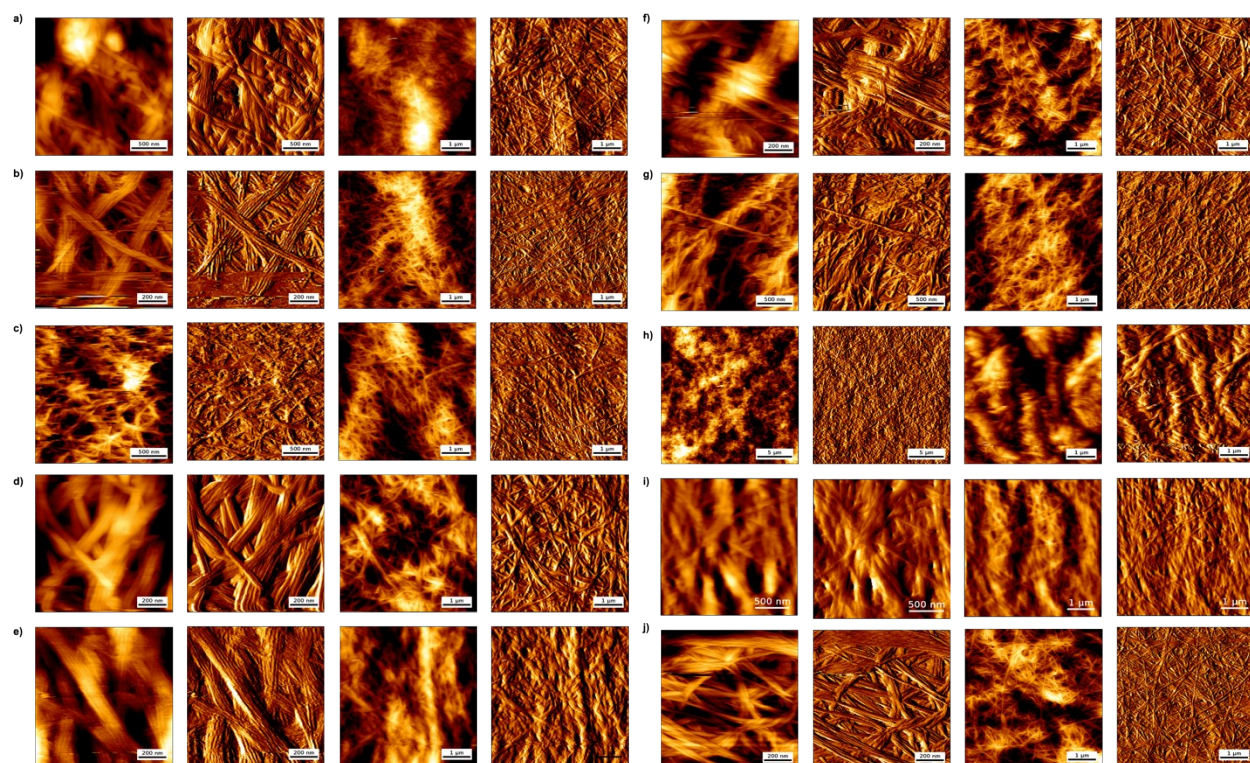
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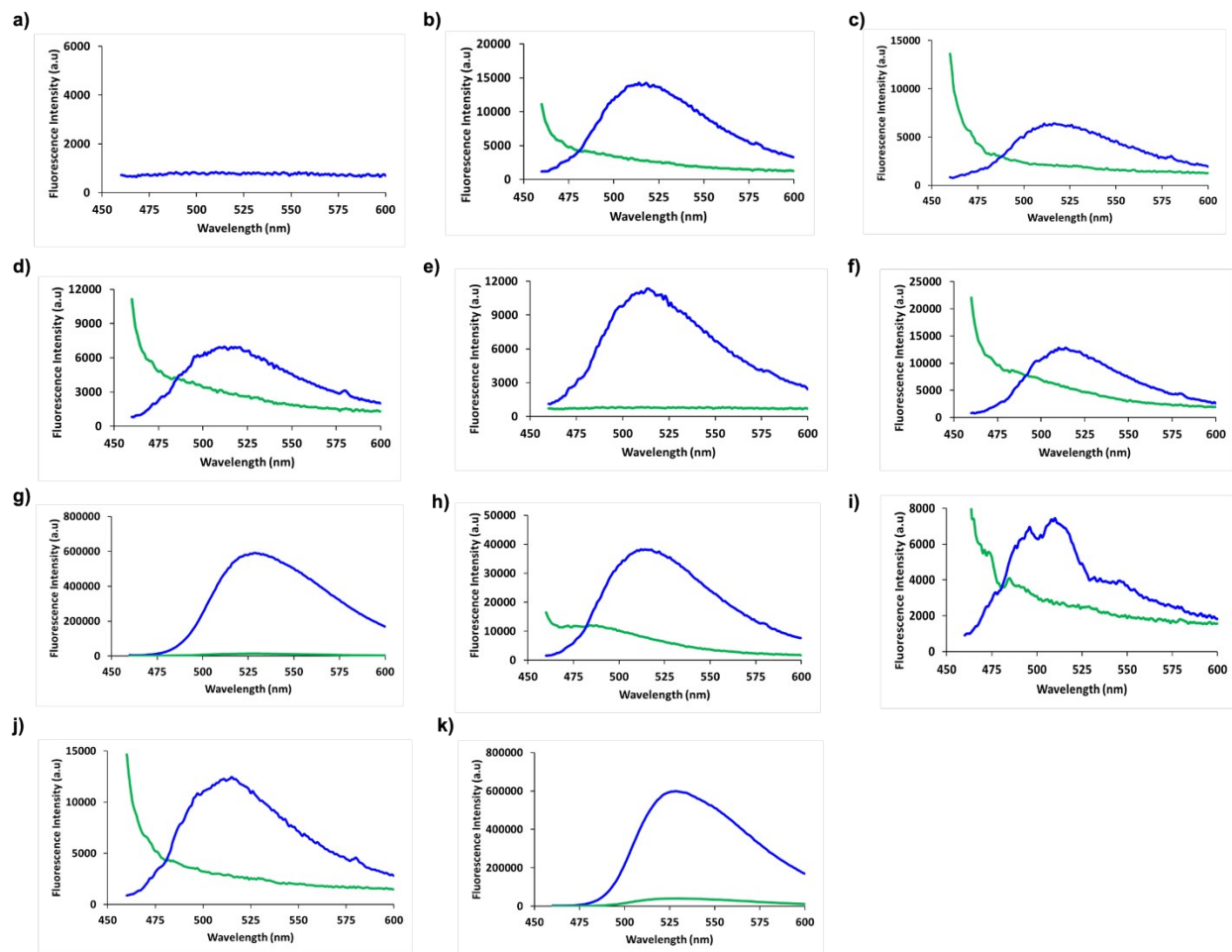
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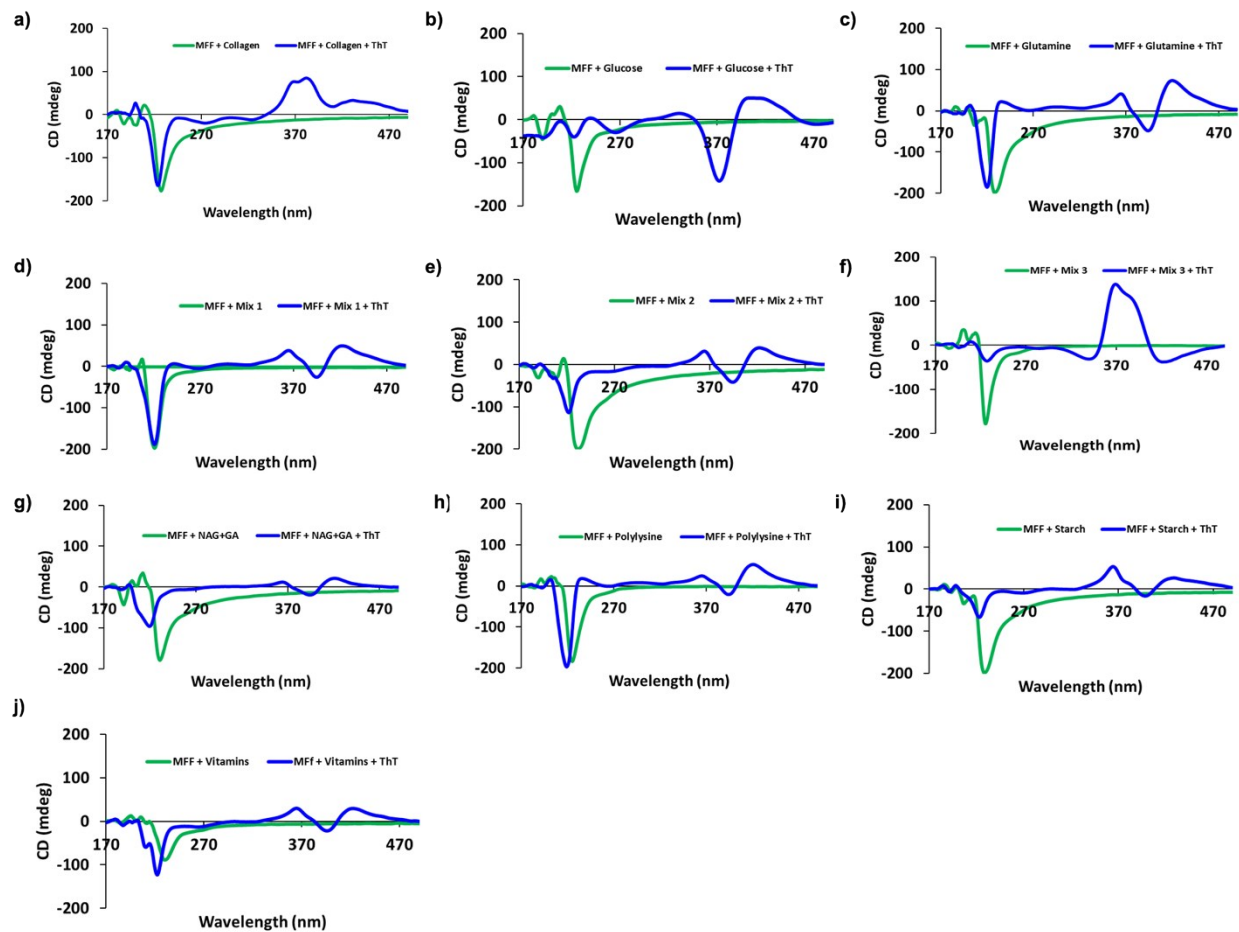


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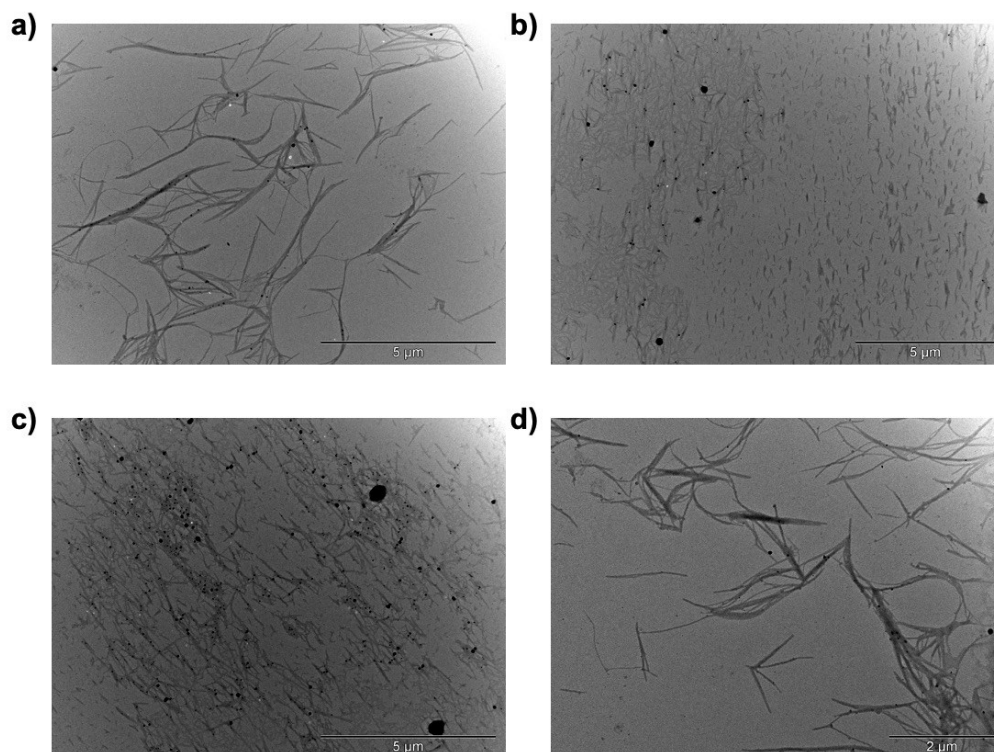


**Figure S14:** Fluorescence spectra of multi-component hydrogels with ThT in gel (blue) and non-gel (green) form. a) ThT, b) C14-FF + collagen, c) C14-FF + glucose, d) C14-FF+ glutamine, e) C14-FF + mix 1, f) C14-FF + mix 2, g) C14-FF + mix 3, h) C14-FF + NAG+GA, i) C14-FF + polylysine, j) C14-FF + starch, k) C14-FF + vitamins.

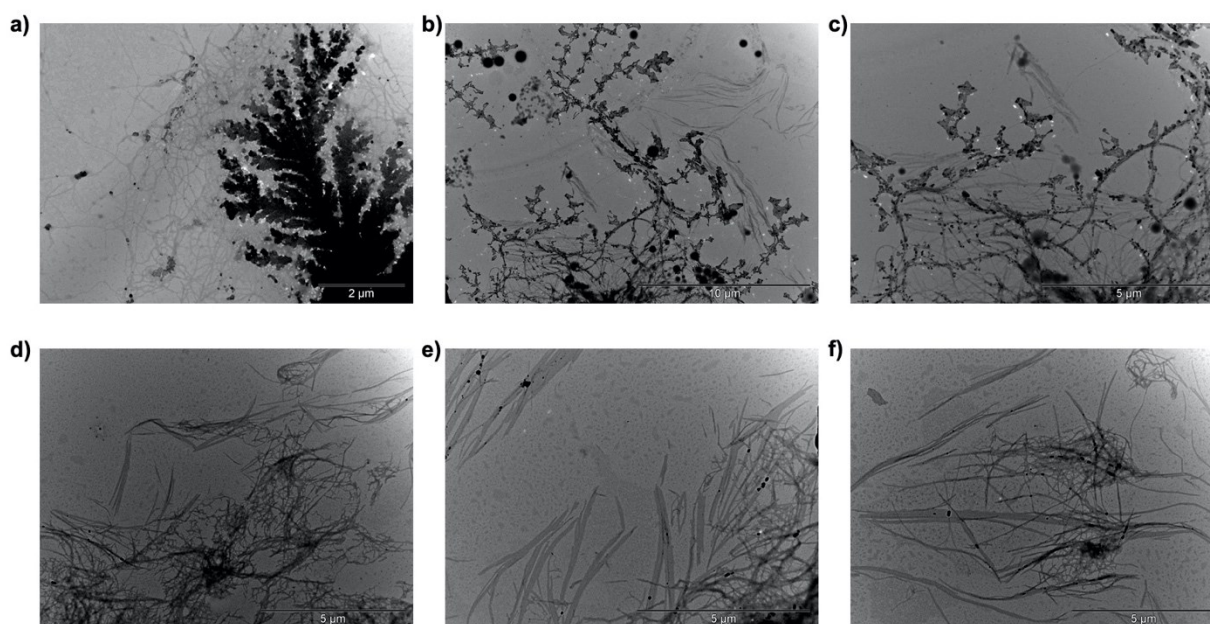




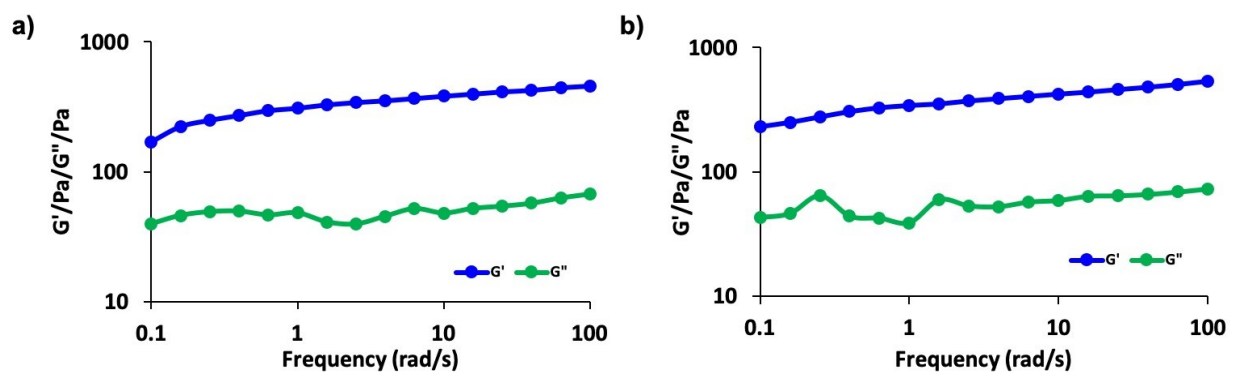
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**Table S1:** Change in matrix properties upon addition of components.

Component added to C14-FF	Mechanical increase/decrease in relation to C14-FF (264 Pa) (Pa)	Fiber Width increase/decrease in relation to C14-FF (32 nm) (nm)	Morphology Changes in TEM in comparison to C14-FF nanofibers alone
Collagen	+ 146	+58	Nanofiber matrix + collagen fibers
Glucose	+336	+38	Mixtures of thin and thick fibers within matrix
Glutamine	+156	+38	Dense nanofiber matrix + plaques
Mix 1	+26	+78	Nanofiber matrix + branched starch (proteoglycan mimic)
Mix 2	-44	+108	Nanofiber matrix + ordered nanocrystals
Mix 3	-64	+28	Dense thin and thick nanofiber matrix
NAG + GA	+106	+18	Nanofiber matrix + plaques
Polylysine	+36	+78	Dense thin and thick nanofiber matrix
Starch	-54	+38	Nanofiber matrix + starch nanocrystals within
Vitamins	-84	+58	Nanofiber matrix + thick nanosheets + spheres