

## SUPPORTING INFORMATION

### **Covalent co-assembly between resilin-like polypeptide and peptide amphiphile into hydrogels with controlled nanostructure and improved mechanical properties**

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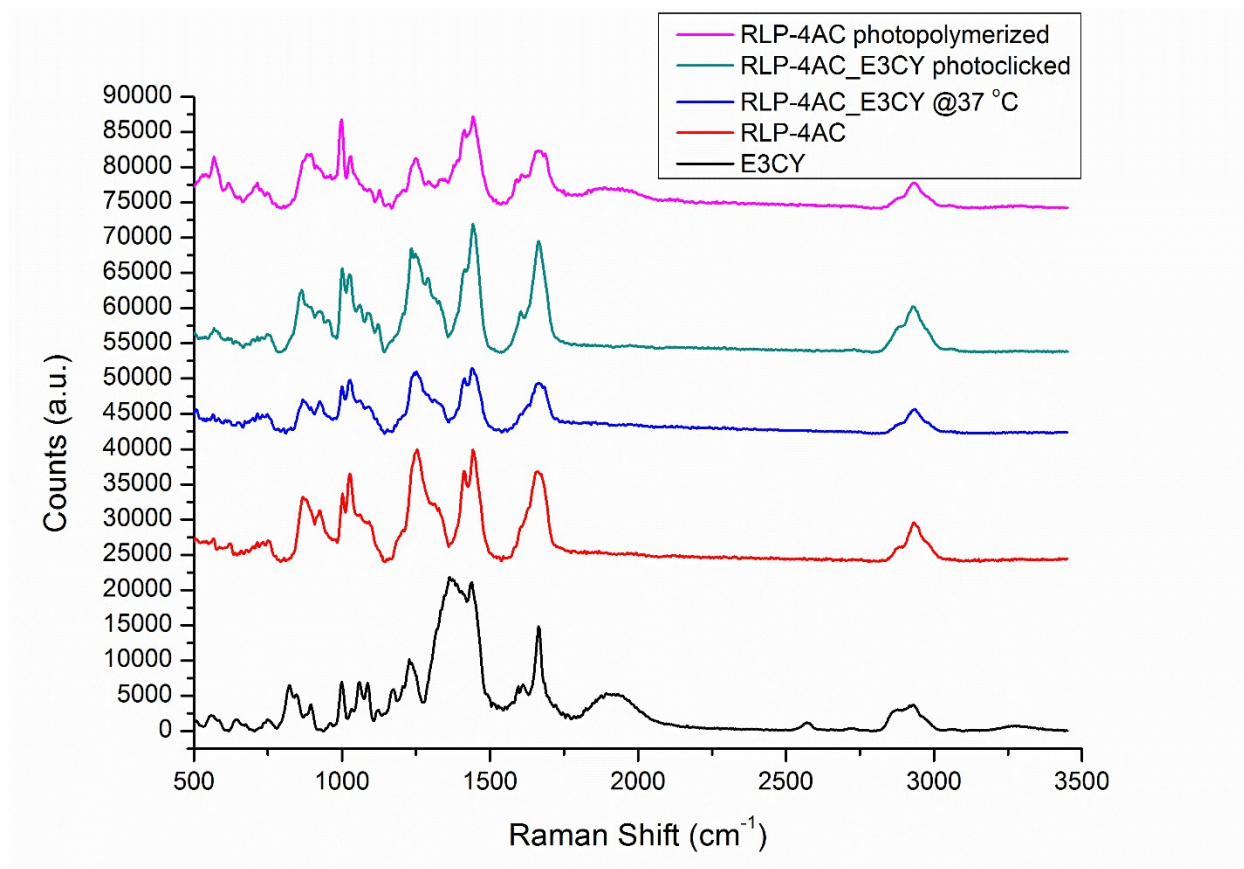
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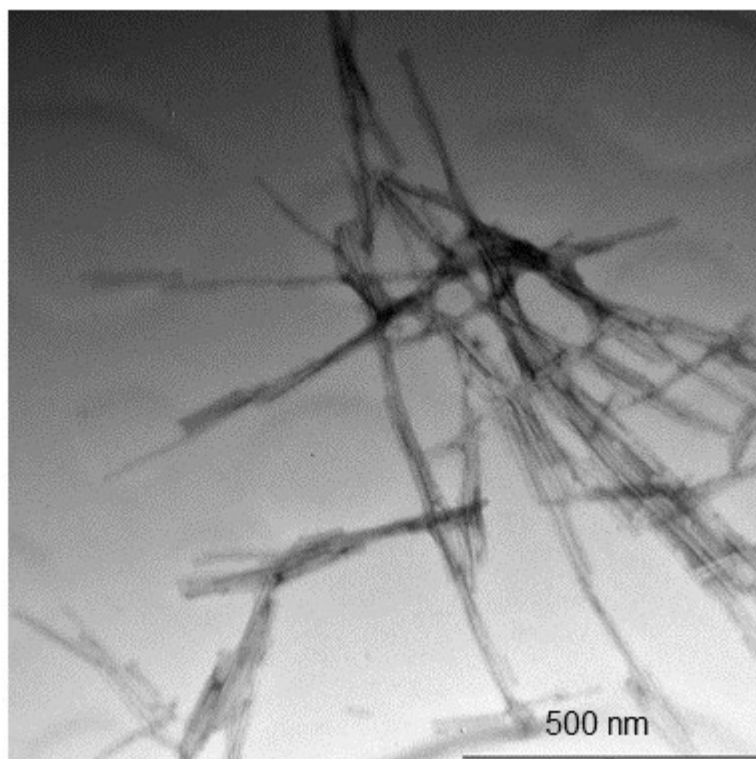
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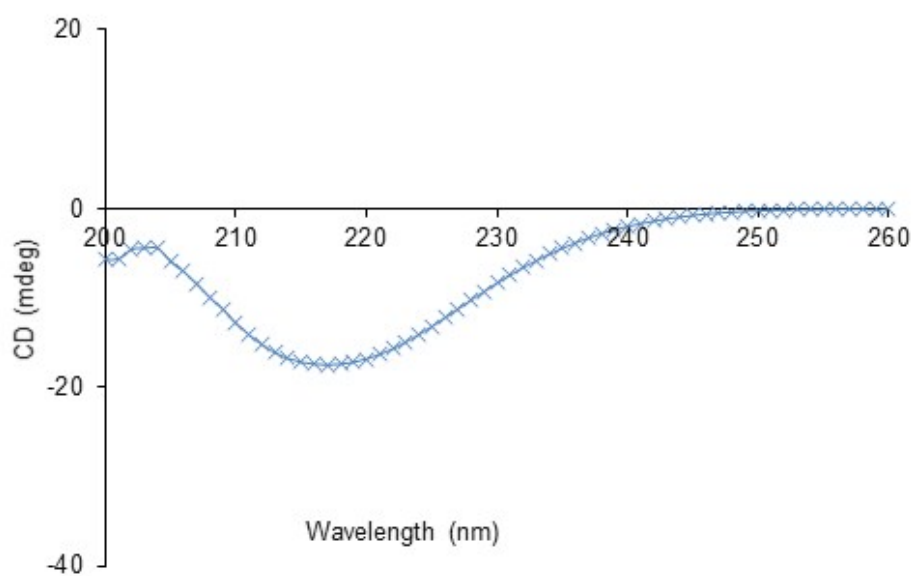
- 1. Raman spectra**
- 2. TEM image**
- 3. CD spectra**
- 4. Frequency sweep rheographs**
- 5.  $^1\text{H}$  NMR spectrum**
- 6. ESI-MS spectrum**
- 7. Analytical HPLC trace**



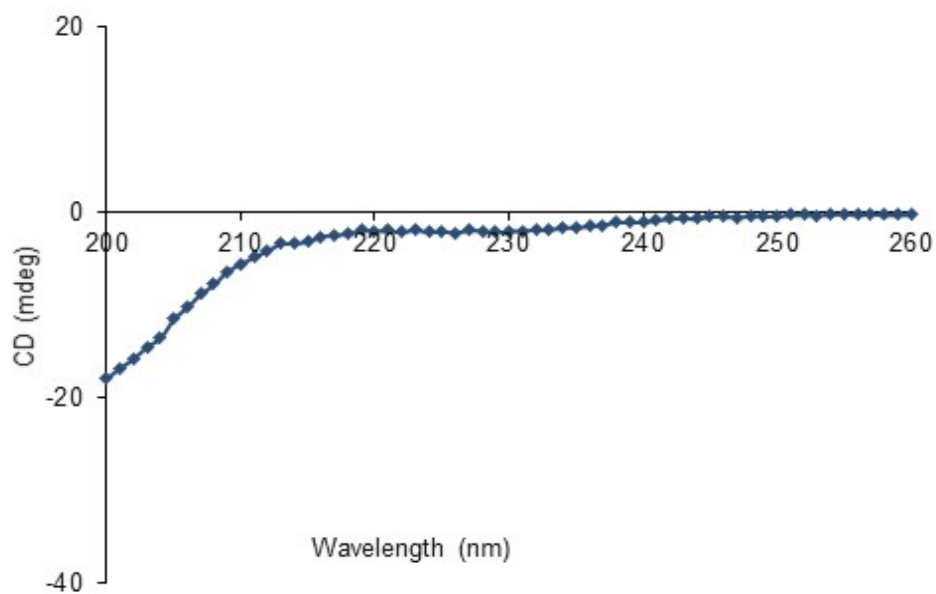
**Figure S1.** Raman spectra for E3CY, RLP-4AC, RLP\_E3CY@37°C, photoclicked RLP-4AC\_E3CY and photopolymerized RLP-4AC. Laser excitation wavelength: 785 nm. Laser power ~63 mW. Integration times: E3CY (25s); RLP-4AC (90s); RLP\_E3CY@37°C (45s); photoclicked RLP-4AC\_E3CY (45s); photopolymerized RLP-4AC (60s). Spectra are baseline corrected and spatially offset for clarity.



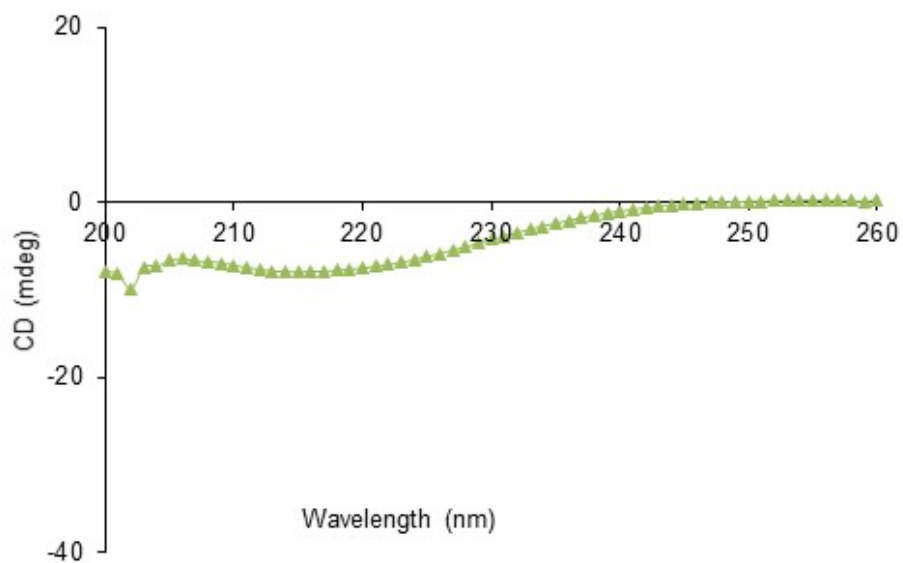
**Figure S2.** TEM micrograph of E3CY after photo irradiation showing the stability of the PA nanofibers to photodegradation.



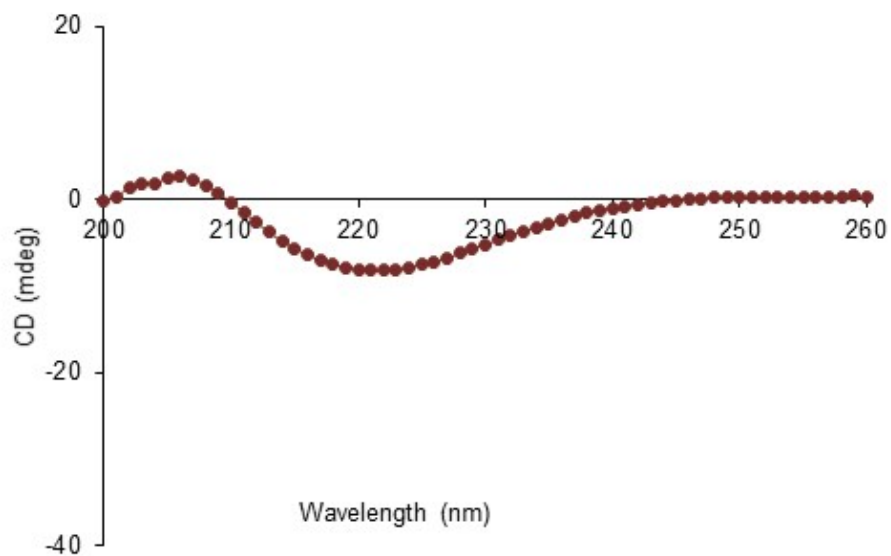
**Figure S3.** Circular dichroism spectra for E3CY solution (0.05% wt) in PBS 1x at 25 °C.



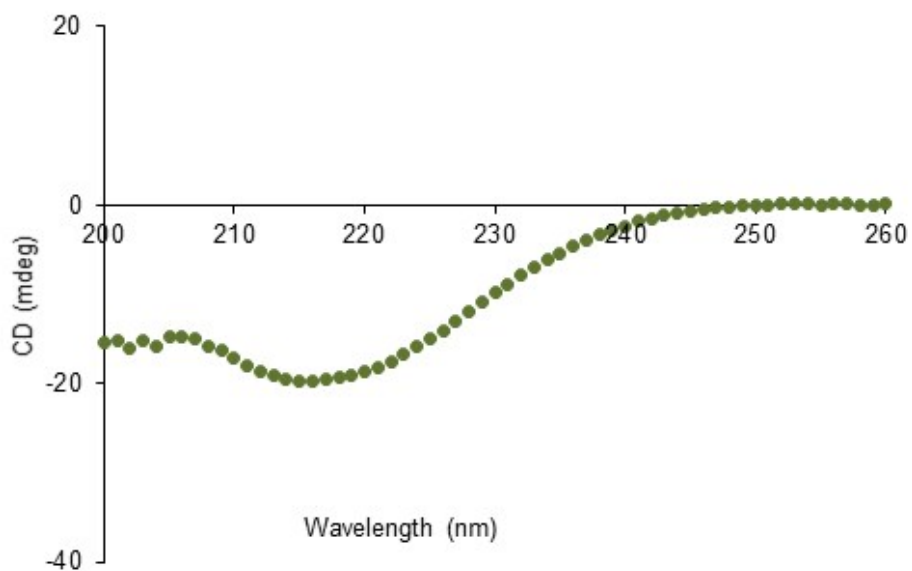
**Figure S4.** Circular dichroism spectra for RLP-4AC solution (0.25% wt) in PBS 1x at 25 °C.



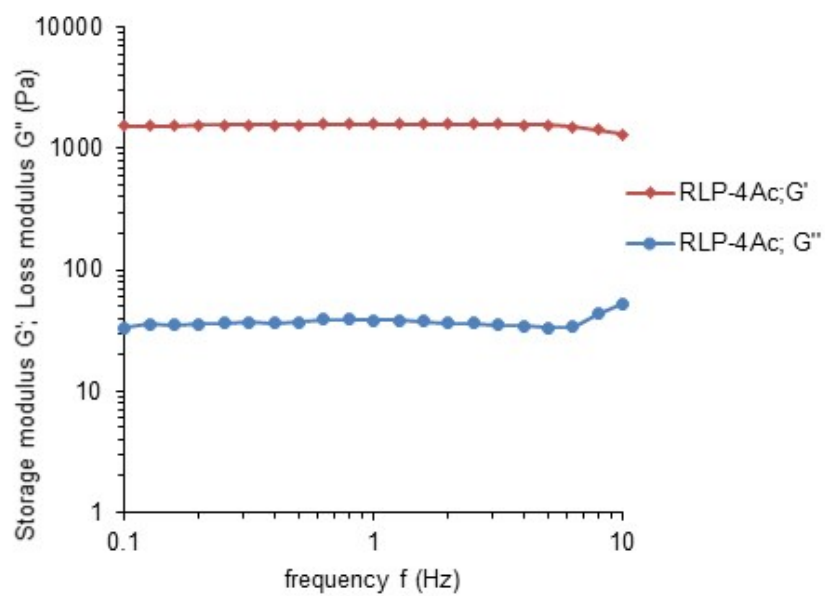
**Figure S5.** Circular dichroism spectra for the mixture of RLP-4AC (0.25%) with E3CY (0.01% wt) after 6 h of incubation at 37 °C.



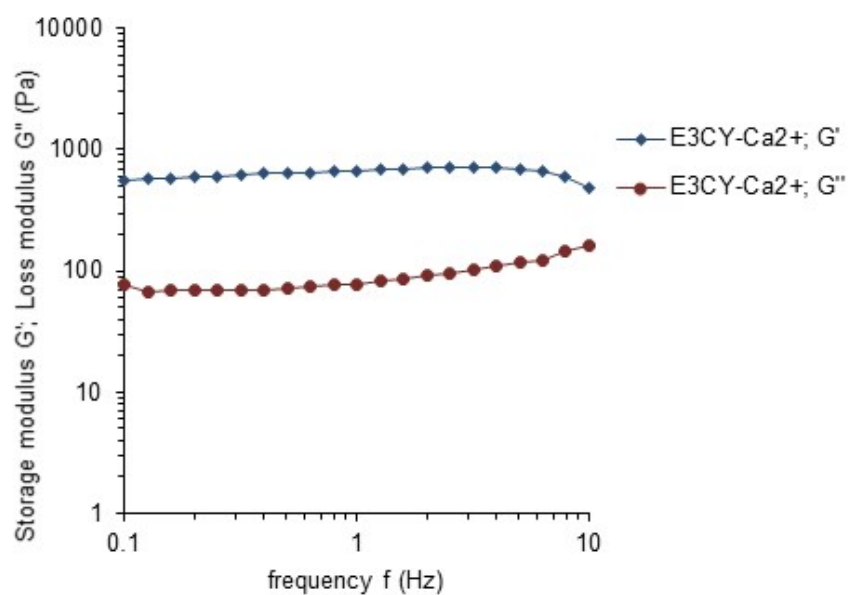
**Figure S6.** Circular dichroism spectra for the mixture of RLP-4AC (0.25%) with E3CY (0.025% wt) after 6 h of incubation at 37 °C.



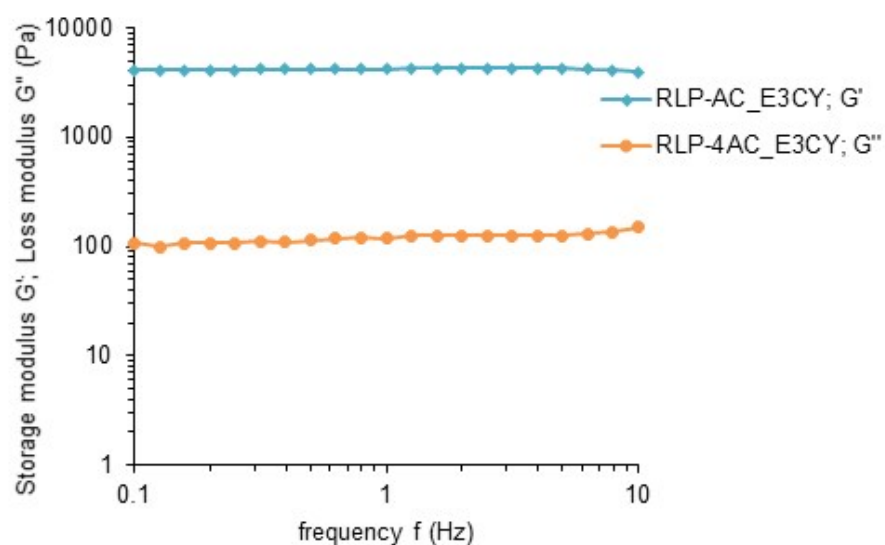
**Figure S7.** Circular dichroism spectra for the mixture of RLP-4AC (0.25%) with E3CY (0.05% wt) after 6 h of incubation at 37 °C.



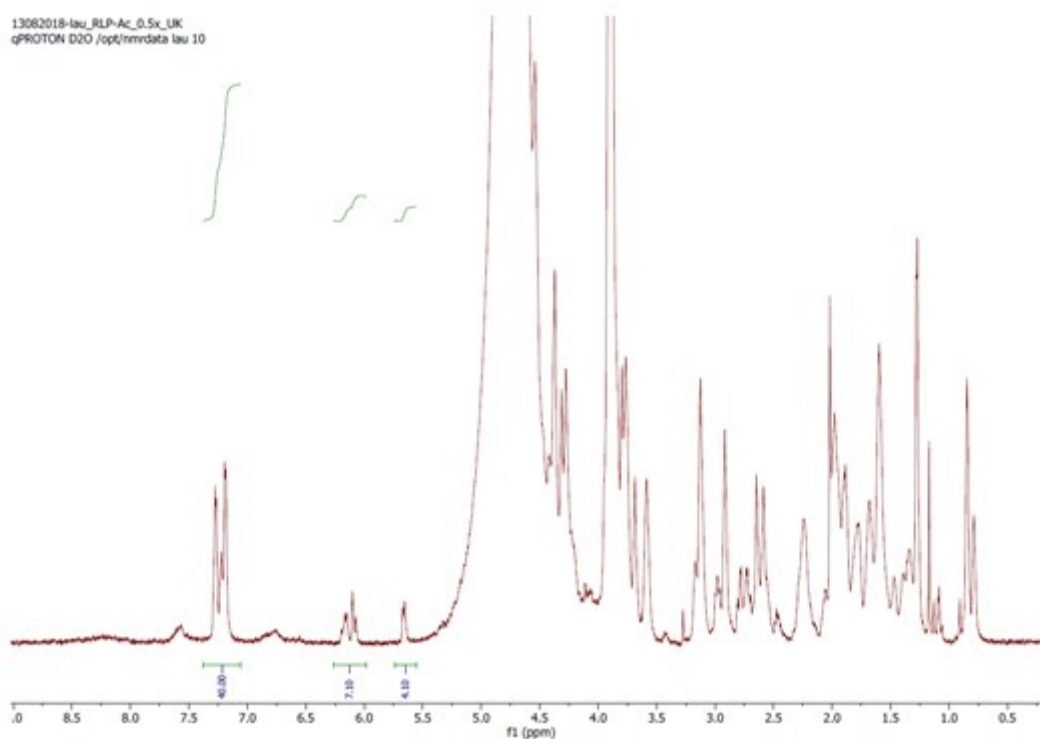
**Figure S8.** Frequency sweep rheographs for 5% wt RLP-4AC photopolymerized hydrogels.



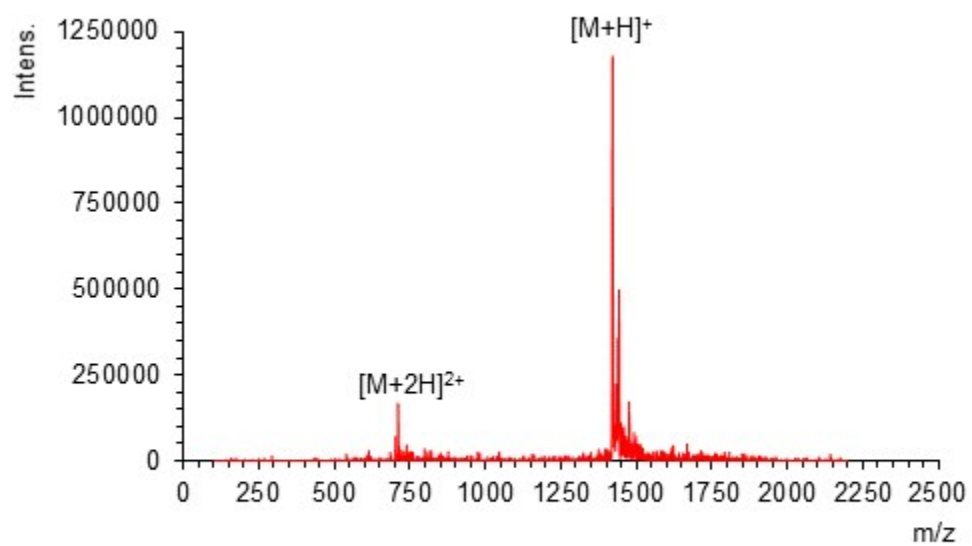
**Figure S9.** Frequency sweep rheographs for 1% wt E3CY- $\text{Ca}^{2+}$  hydrogels.



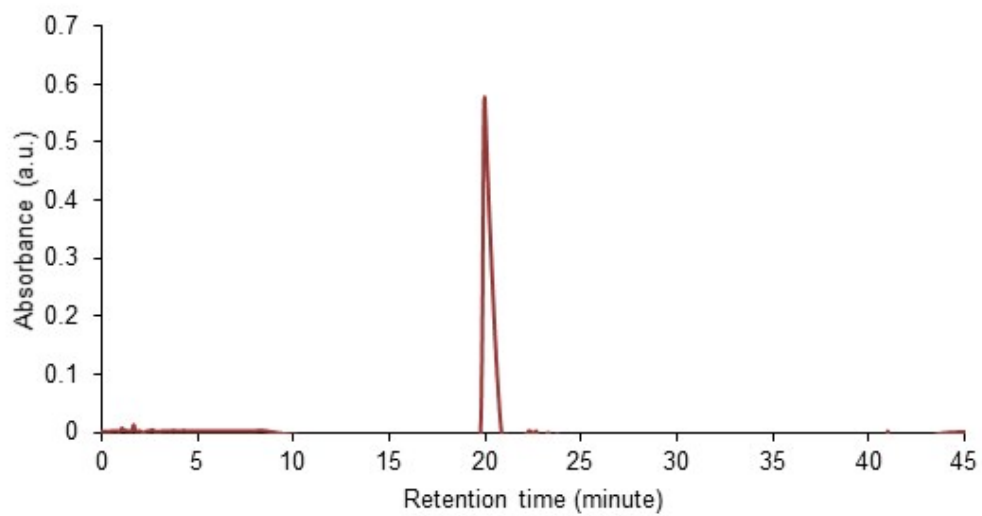
**Figure S10.** Frequency sweep rheographs for RLP-4AC\_E3CY hydrogels synthesized by thiol-ene photoclicked at 1 and 5% wt final concentrations of E3CY and RLP-4AC.



**Figure S11.**  $^1\text{H}$  NMR spectrum showing the vinylic protons of acrylamide integrated against the lysine amine protons of RLP backbone.



**Figure S12.** ESI-MS spectrum of E3CY peptide amphiphile



**Figure S13.** HPLC trace of E3CY peptide amphiphile