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Supplementary Information

Hierarchical Stabilization of Bioactive Hydrogels by Multi-Arm Peptide–Polymer Supramolecular Staples

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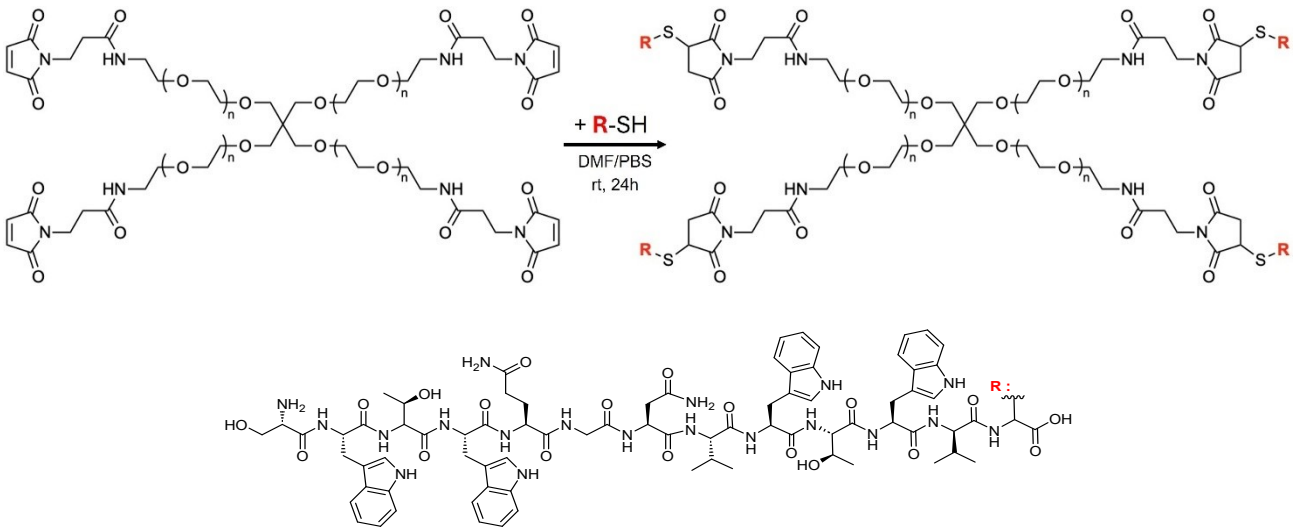
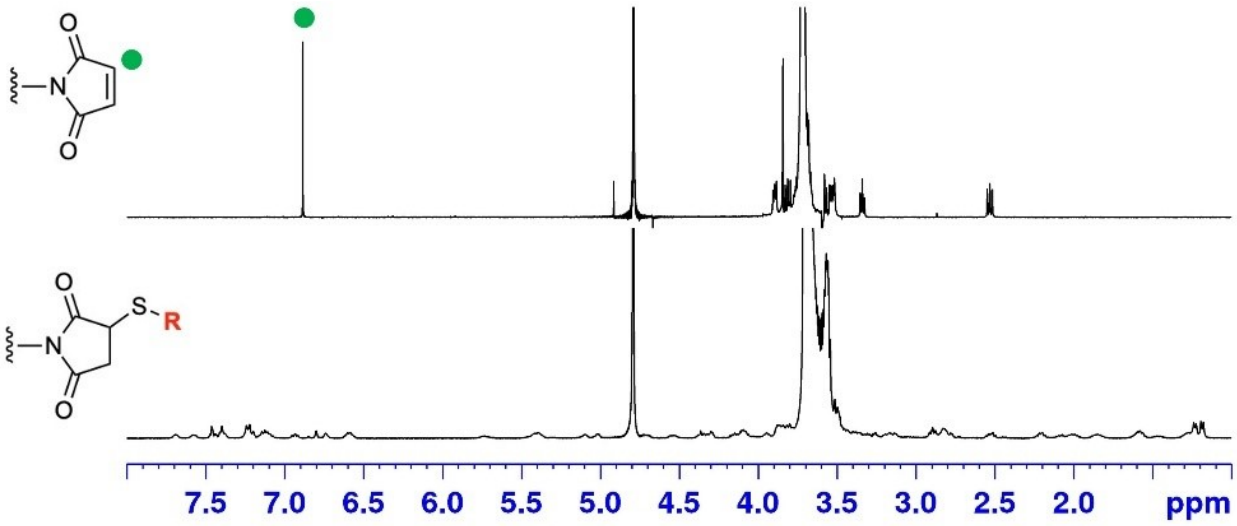


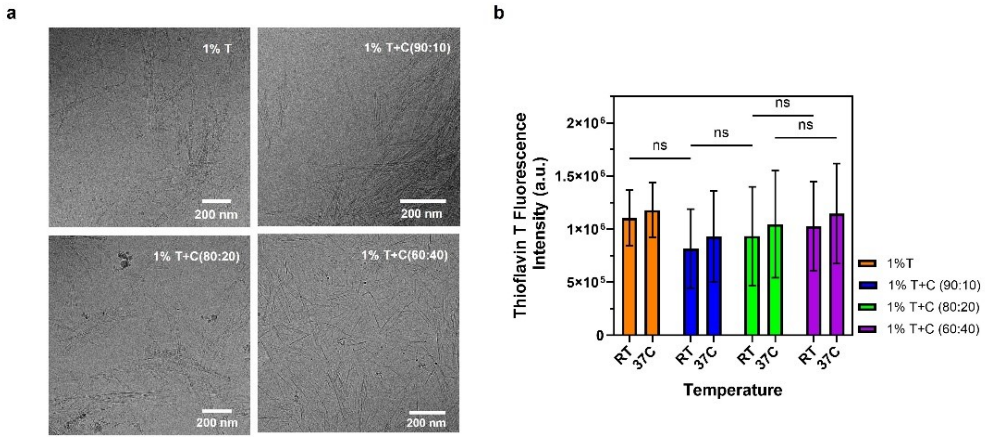
Supplementary Figure 1. Reaction of thiols in the cysteine residues of Trpzip peptides with 4 Arm-PEG-MAL



Supplementary Figure 2. ^{1}H NMR spectrum of 4 arm PEG-Mal (top) and Trpzip_q5_cys / 4-arm PEG conjugate (bottom).

Supplementary Table 1. Material compositions of hydrogel samples.

Sample code	**Trpzip: Trpzip-PEG Conjugate ratio (w/w)**	**Trpzip concentration (w/v, mg in v (µl of final volume of the hydrogel))**	**Trpzip–PEG concentration (w/v, mg in v (µl of final volume of the hydrogel))**
1% T	-	0.01×v	-
1% T+ C (90:10)	90:10	0.01×v	0.01×v×10/90
1% T+ C (80:20)	80:20	0.01×v	0.01×v×20/80
1% T+ C (60:40)	60:40	0.01×v	0.01×v×40/60



Supplementary Figure 3. Physicochemical characterization of Trpzip/Trpzip-PEG hydrogels. a. Cryo-TEM micrographs of 1% (w/v) Trpzip hydrogels and co-assembled systems with varying ratios of Trpzip to Trpzip-PEG: 90:10, 80:20, and 60:40 acquired 10 h post-gelation at 37 °C, revealing the nanoscale network morphology **b.** Thioflavin T fluorescence intensity of the corresponding hydrogels measured at room temperature (RT) and 37 °C, indicating the extent of β-sheet formation. Data are presented as mean ± standard deviation, with statistical analysis showing no significant differences (ns) between groups.

Supplementary Table 2. Quantitative Morphology of Filamentous Structure in Cryo-Electron Tomograms.

Sample Codes	**1% T**	**1% T+C (90:10)**	**1% T+C (80:20)**	**1% T+C (60:40)**
Length of ribbon bundle (nm)	238-486	321-700	115-373 200-536	350
Thickness of bundle (nm)	14-78	13.6-42	19- 65	16.2-19.8
Length of single filament (nm)	-	-	-	55.9-200
Thickness of single filament (nm)	-	-	-	7.24-7.75

Note: Values represent measurements of individual filaments or bundle structures extracted from cryo-TEM tomograms and therefore reflect the distribution of observed sizes within each sample. Discrete values correspond to measurements of individual structures rather than distinct structural populations.

Supplementary Table 3. Parameter values of 0.5% Trpzip hydrogel fitted with Power Law + Flexible cylinder model.

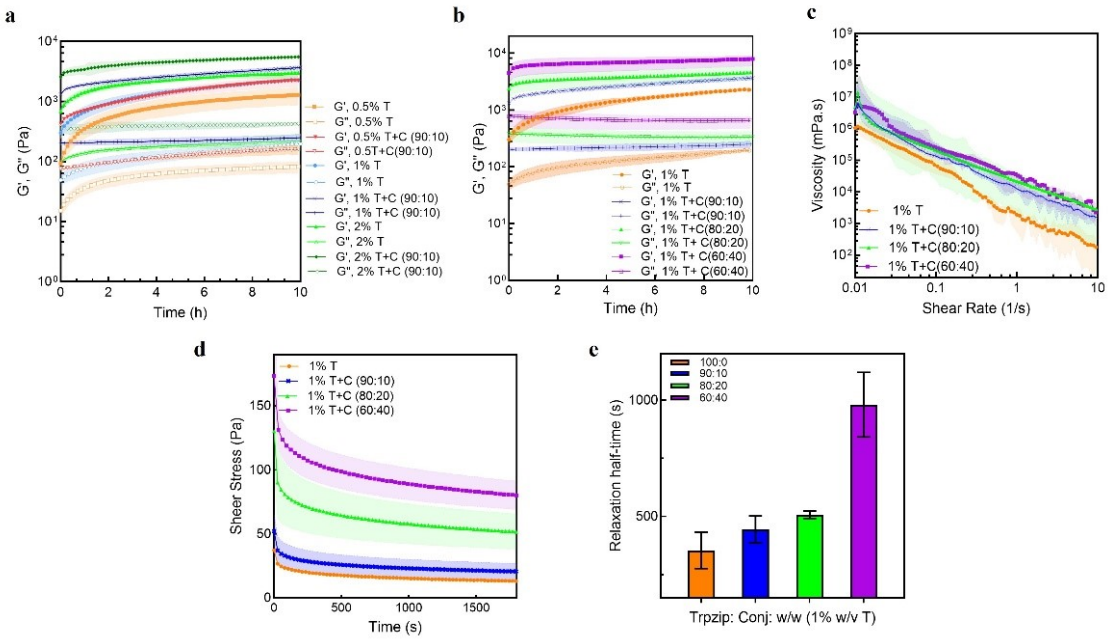
Hydrogel		Background	Power Scale	Power exponent	Flexible Cylinder Scale	Flexible Cylinder length, nm	Kuhn length, nm	Radius, nm	Polymer SLD	D2O SLD	Kuhn Length poly dispersity	Radius poly dispersity
0.5%T	Absolute	1.06E-02	8.58E-09	3.58	7.06E-04	1000	6.50	2.23	3.69	6.39	0.1	0.1
	Error	±6.93E-05	±2.22E-9	±3.97E-02	±7.41E-06		±0.23	±0.02				

Supplementary Table 4. Parameter values of Trpzip/Trpzip-PEG hydrogel fitted with Power Law and Ellipsoid model.

Hydrogel		**Background**	**Power Scale**	**Power exponent**	**Ellipsoid Scale**	**Polar Radius, nm**	**Equatorial Radius, nm**	**Peptide SLD**	**D_2O SLD**
0.5T+ C (90:10)	**Absolute**	6.76E-03	8.30E-05	2.37	6.10E-04	6.76	45.74	3.29	6.39
	Error	±2.09E-05	±7.12E-07	±2.62E-03	±5.03E-6	±0.03	±0.57		
0.5T+ C (80:20)	**Absolute**	6.37E-03	8.74E-05	2.38	6.36E-04	7.954	48.24	2.95	6.39
	Error	±2.08E-05	±7.24E-07	±2.65E-03	±4.17E-06	±0.03	±0.48		
0.5T+ C (60:40)	**Absolute**	8.83E-03	1.63E-04	2.27	8.15E-04	9.04	58.03	2.43	6.39
	Error	±2.29E-05	±1.60E-06	±3.60E-03	±4.45E-06	±0.02	±0.44		

Supplementary Table 5. Parameter values obtained from Power-law fitting of USANS data for Trpzip/Trpzip-PEG hydrogels at 90:10, 80:20, and 60:40 formulations.

Hydrogel		**Scale**	**Background**	**Power**
0.5T+C (90:10)	**Absolute**	3.17E-07	6.76E-03	**2.98**
	Error	3.60E-08		0.01
0.5T+ C (80:20)	**Absolute**	2.61E-07	6.37E-03	**3.03**
	Error	2.57E-08		0.01
0.5T+ C (60:40)	**Absolute**	3.17E-07	8.83E-03	**3.07**
	Error	2.86E-08		0.01



Supplementary Figure 4. The Mechanical properties of Trpzip-based hydrogels. a, b Oscillatory time sweep measurements of the storage modulus (G′) and loss modulus (G″) of 0.5%, 1% and 2% w/v of Trpzip-based hydrogels **c** Viscosity flow curves of 1% w/v Trpzip-based hydrogels conducted using a logarithmic ramping rate from 0.01 to 10 s^{-1} shear rate over 10 min **d** Stress-relaxation profiles of 1% w/v Trpzip hydrogel and its hydrogels with Trpzip-PEG performed at 5% strain for 0.5 h **e** Relaxation half-time ($\tau_{1/2}$) of 1% w/v Trpzip-based hydrogels.