Supporting Information

Incorporation of PEG diacrylates (PEGDA) generates hybrid Fmoc-FF matrices.

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Figure S1: Self-supporting hydrogel of Fmoc-FF/PEGDA1 1/50 *mol/mol* ratio.

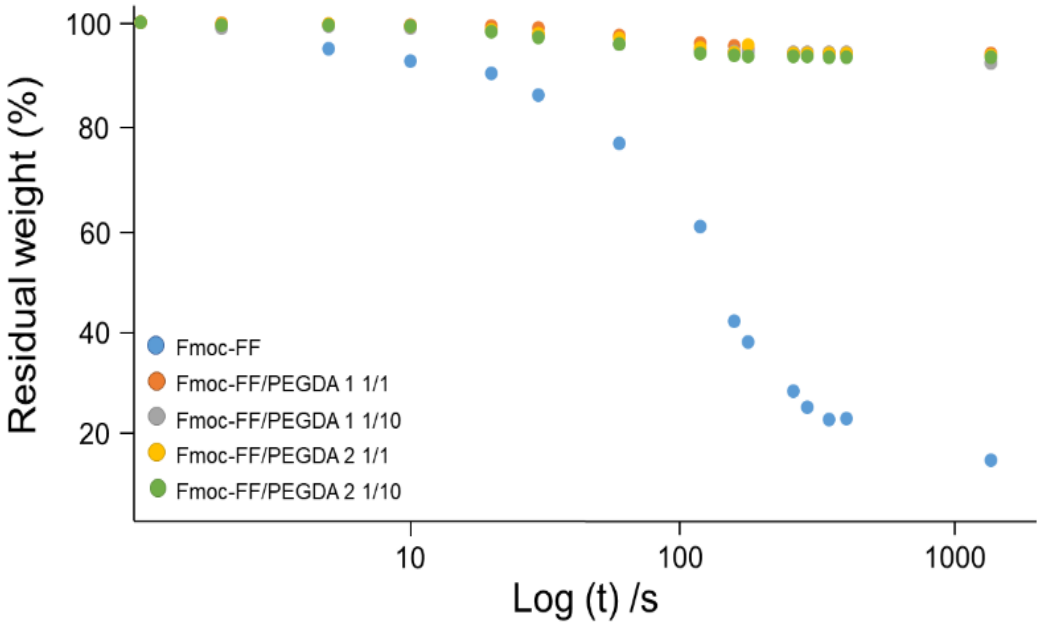

Figure S2: Water retention curve of mixed Fmoc-FF/PEGDA hydrogels.

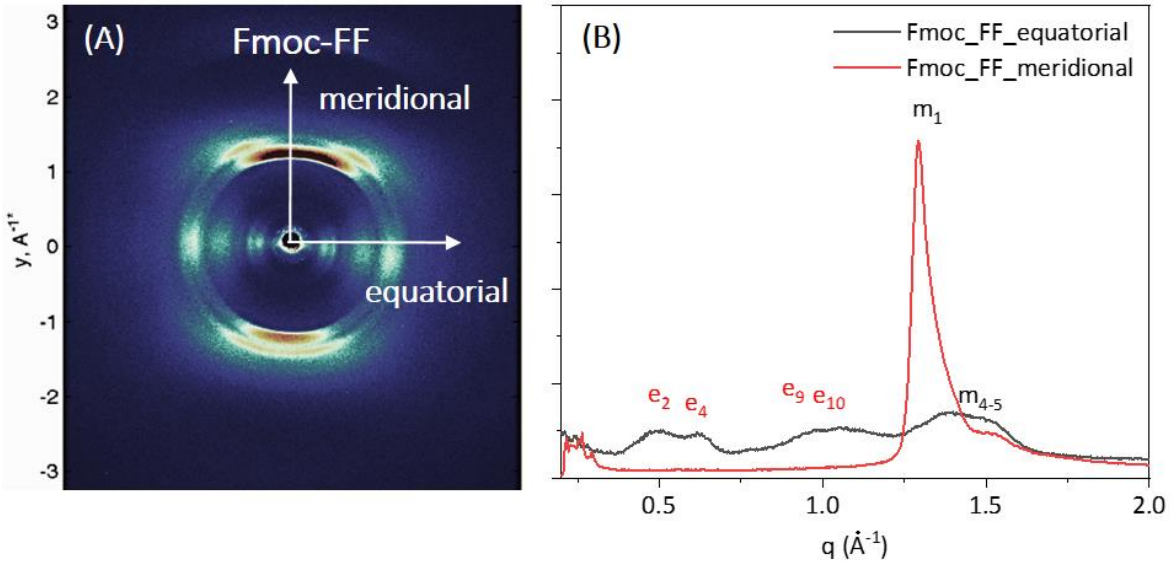

Figure S3: WAXS characterization of the mixed hydrogels Fmoc-FF: 2D WAXS data (on the left), and 1D WAXS meridional/equatorial profiles (on the right).

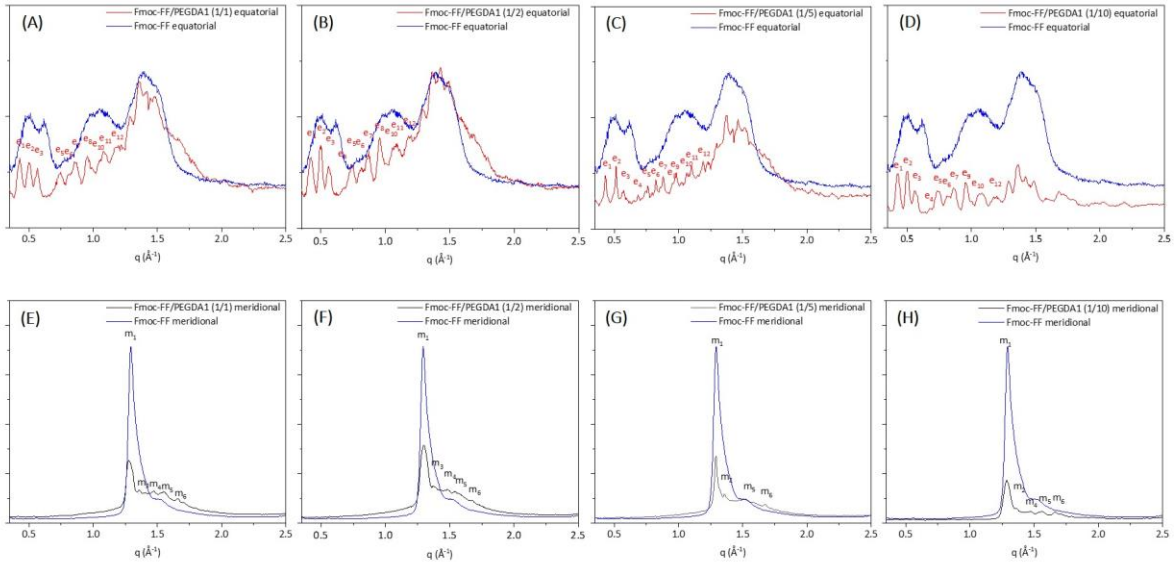

Figure S4: Superimposition of 1D WAXS equatorial (A, B, C, D) and meridional (E, F, G, H) profiles for Fmoc-FF HG and PEGDA1 based HGs at the studied molar ratio.

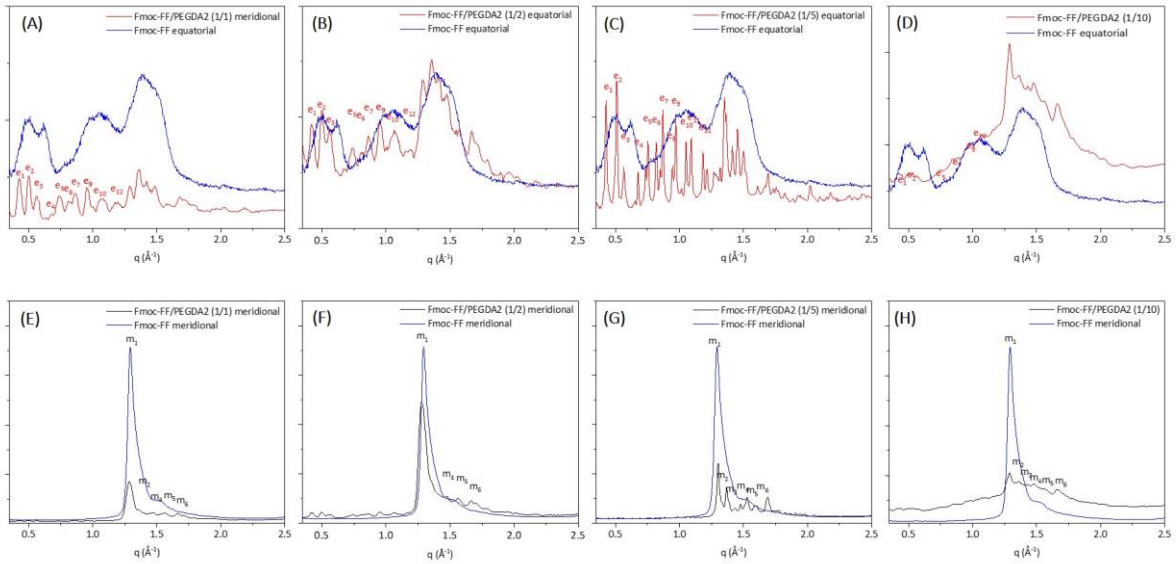

Figure S5: Superimposition of 1D WAXS equatorial (A, B, C, D) and meridional (E, F, G, H) profiles for Fmoc-FF HG and PEGDA2 based HGs at the studied molar ratio.

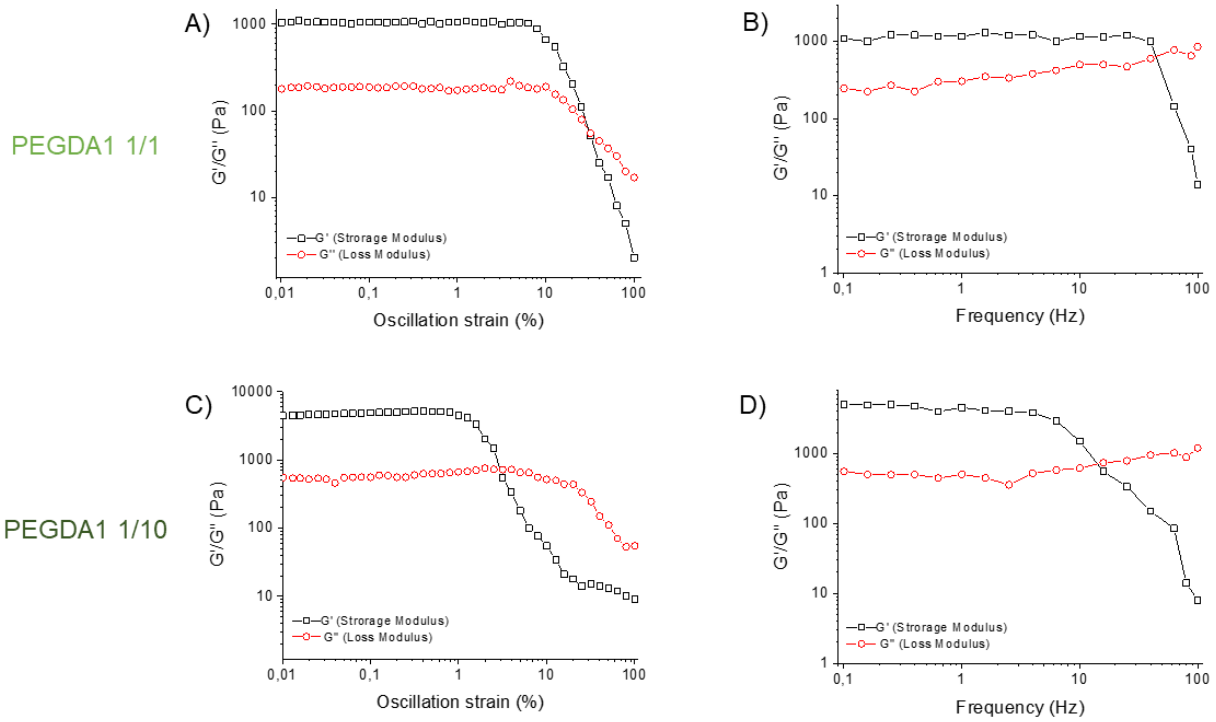

Figure S6: Oscillation time sweeps tests for PEGDA1 1/1 (A) and PEGDA1 1/10 (C); frequency time sweeps tests for PEGDA1 1/1 (B) and PEGDA1 1/10 (D).

PEGDA2 1/1

PEGDA2 1/10

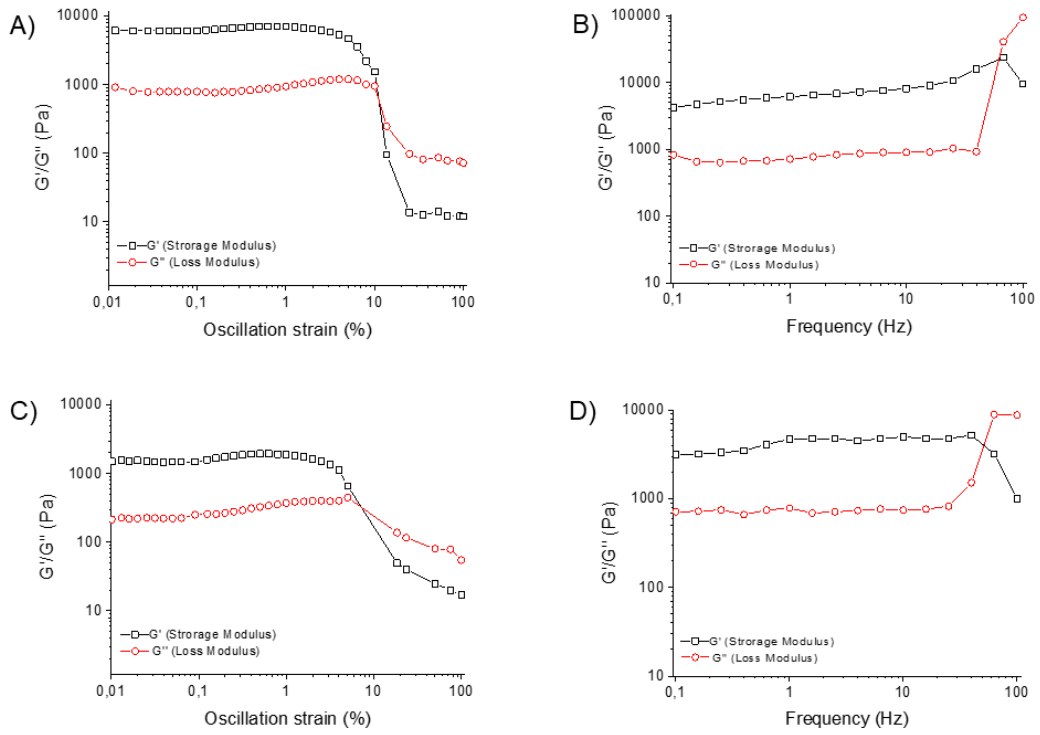

Figure S7: Oscillation time sweeps tests for PEGDA2 1/1 (A) and PEGDA2 1/10 (C); frequency time sweeps tests for PEGDA2 1/1 (B) and PEGDA2 1/10 (D).

Table S1: Meridional and equatorial peak positions in q ($Å^{-1}$) and corresponding distance d = 2π/q (Å) of the mixed hydrogels; in red the common peak with the Fmoc-FF reference fiber.

Reflections	*q* [$Å^{-1}$] ± 0.02	*d* [Å] ± 0.5
Equatorial	e_1 = 0.42	d_{e1}=14.96
	e_2 = 0.50	**d_{e2}=12.57**
	e_3 = 0.56	d_{e3}=11.22
	e_4 = 0.65	**d_{e4}=9.67**
	e_5 = 0.74	d_{e5}=8.49
	e_6 = 0.81	d_{e6}=7.76
	e_7 = 0.87	d_{e7}=7.22
	e_8 = 0.94	d_{e8}=6.68
	e_9 = 0.97	**d_{e9}=6.48**
	e_{10} = 1.05	**d_{e10}=5.98**
	e_{11} = 1.09	d_{e11}=5.76
	e_{12} = 1.18	d_{e12}=5.32
Meridional	**m_1 = 1.29**	**d_{m1}=4.91**
	m_2 = 1.32	d_{m2}=4.76
	m_3 = 1.42	d_{m3}=4.42
	m_4 = 1.46	**d_{m4}=4.30**
	m_5 = 1.54	**d_{m5}=4.08**
	m_6 = 1.62	d_{m6}=3.88