Supplementary Figures

Shining a Light on the Hidden Structure of Gelatin Methacryloyl Bioinks Using Small-Angle X-ray Scattering (SAXS)

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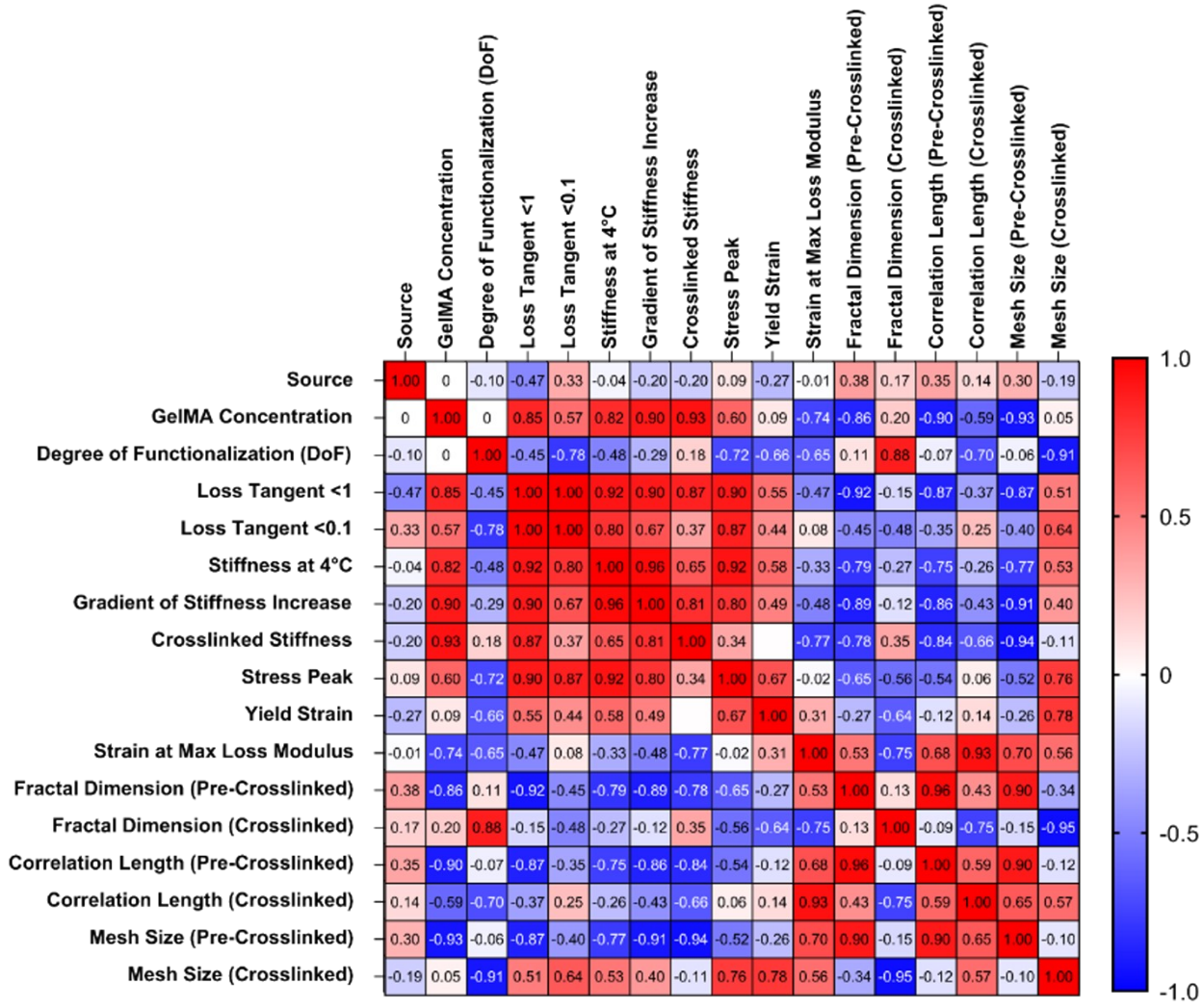
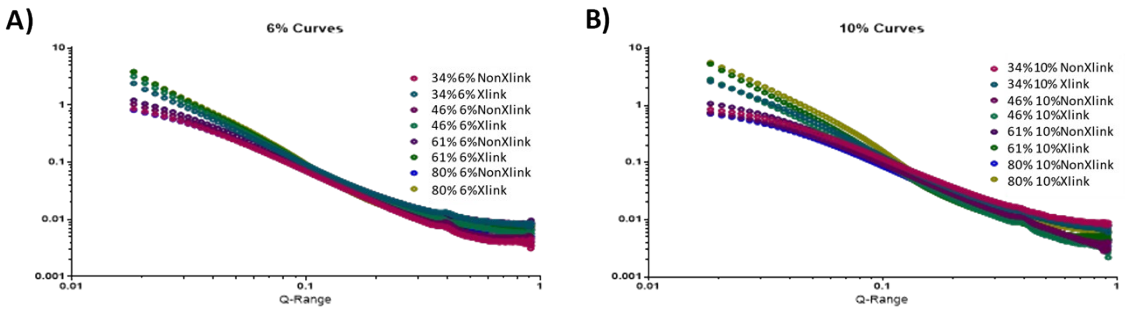
Figure S1. Correlation Heatmap with Values



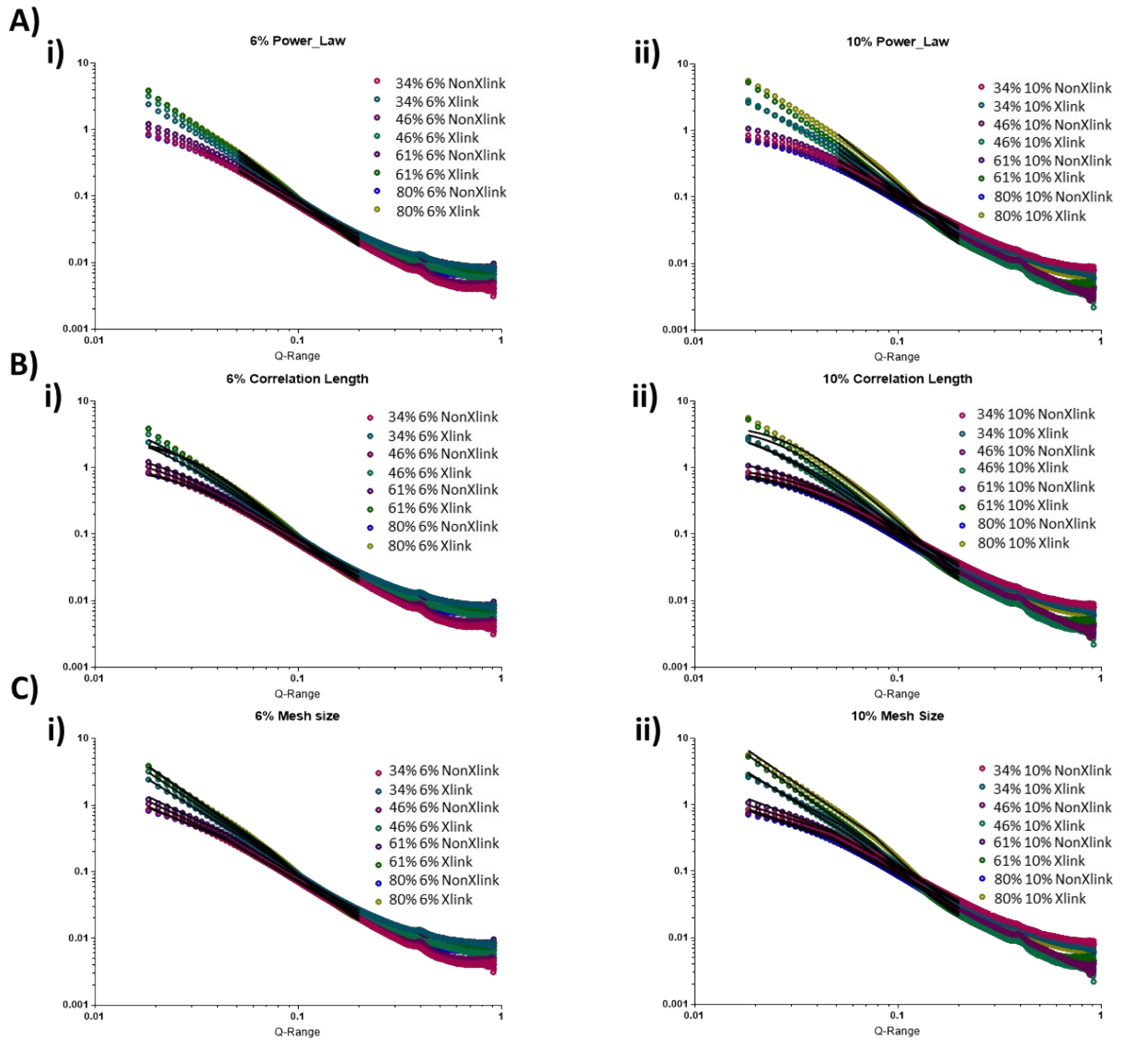
Heatmap showing Pearson r values as an indication of correlations between investigated variables as calculated through Pearson correlation test.

Figure S2. SAXS Acquired Scattering Profiles of GelMA Hydrogels



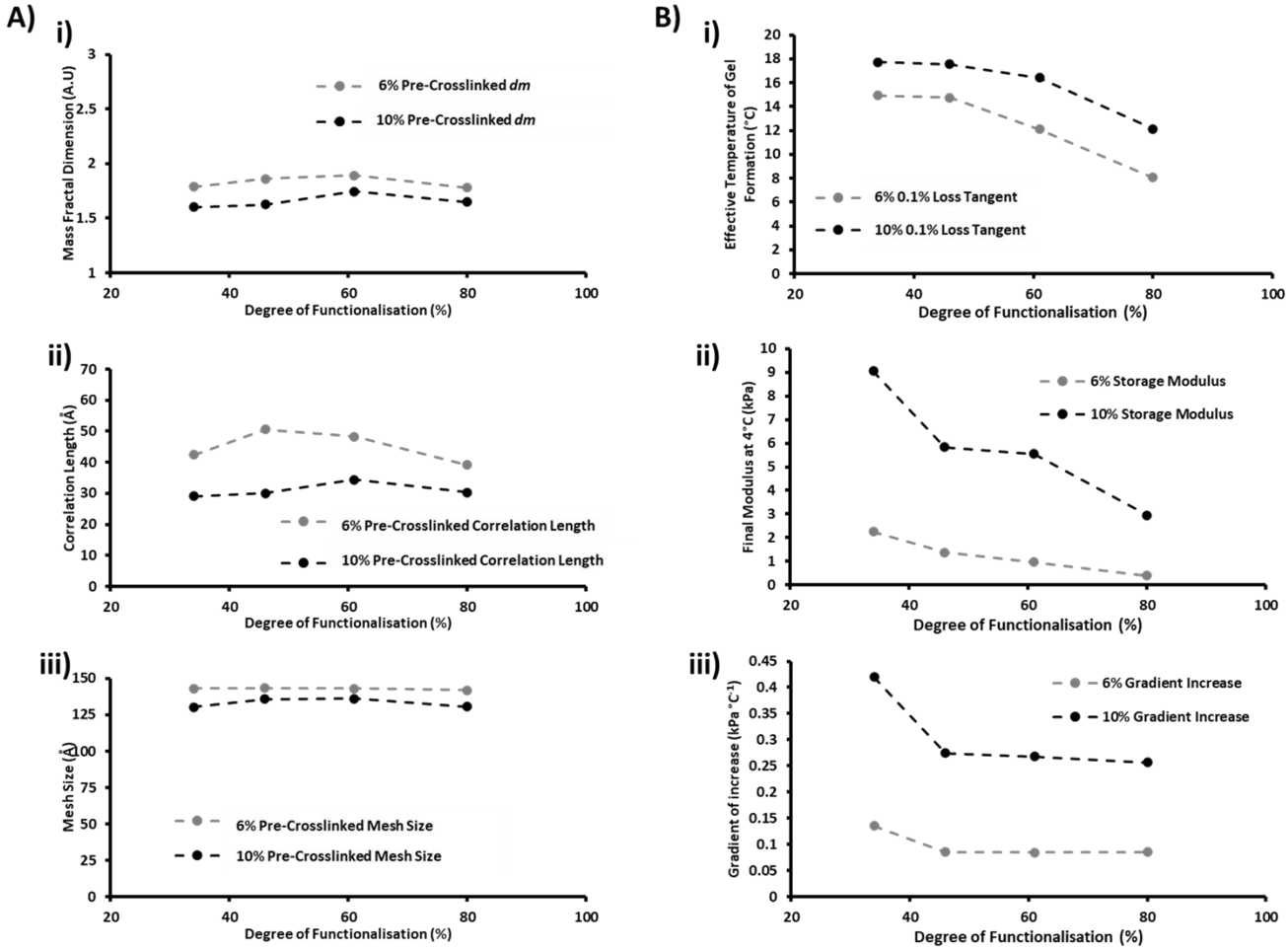
A) 6% GelMA Hydrogels, and B) 10% GelMA hydrogels.

Figure S3. Fitting Various Models to Scattering Profiles of GelMA Hydrogels



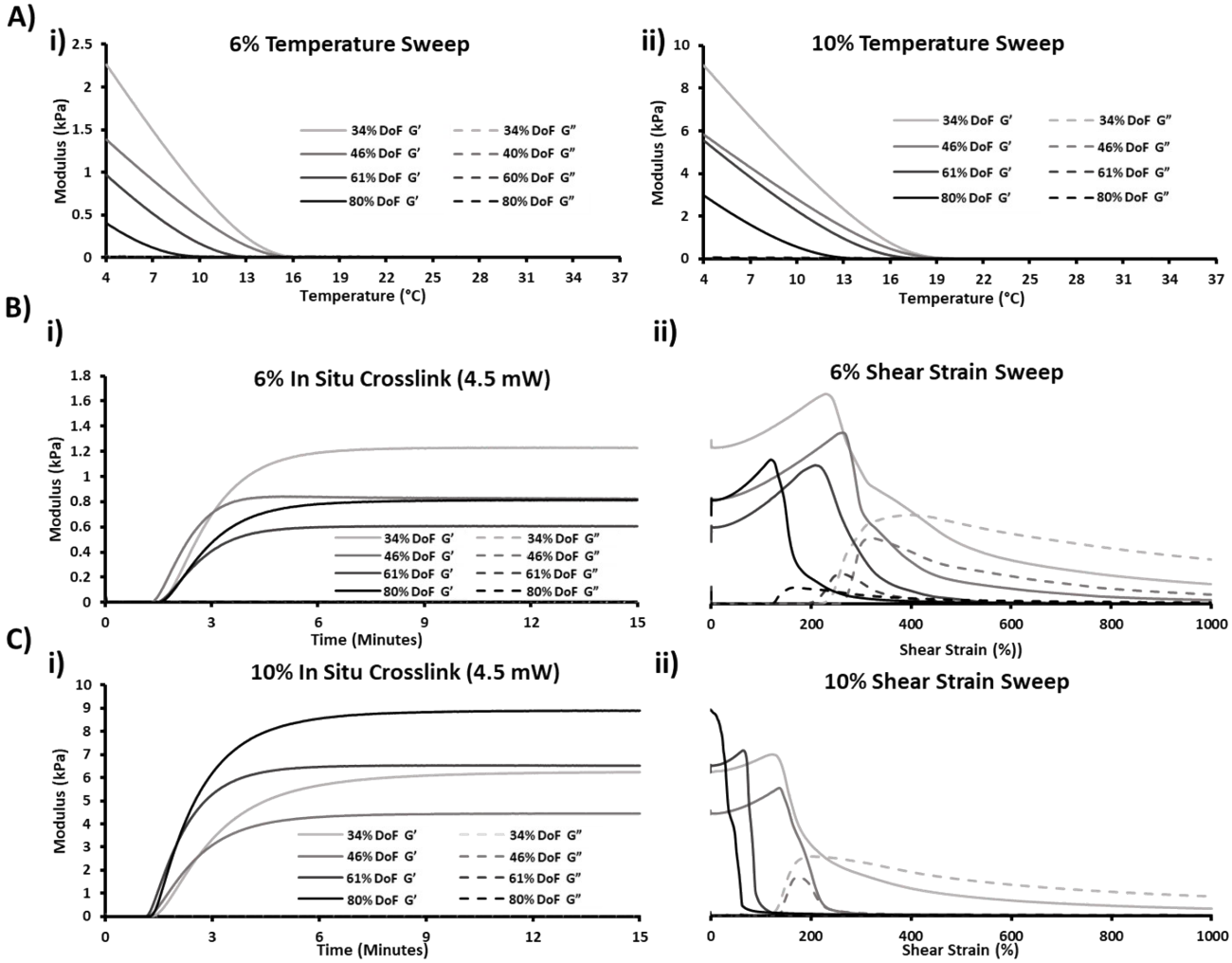
A) Fitting of power-law model (mass fractal dimension) in i) 6% GelMA and ii) 10%. B) Fitting for correlation length in i) 6% GelMA and ii) 10% GelMA. C) Fitting for two-power-law model (for calculation of mesh size) in i) 6% GelMA and ii) 10% GelMA. (Black lines indicate fit)

Figure S4. Nanostructured and Viscoelastic Properties of Pre-Crosslinked GelMA Hydrogels



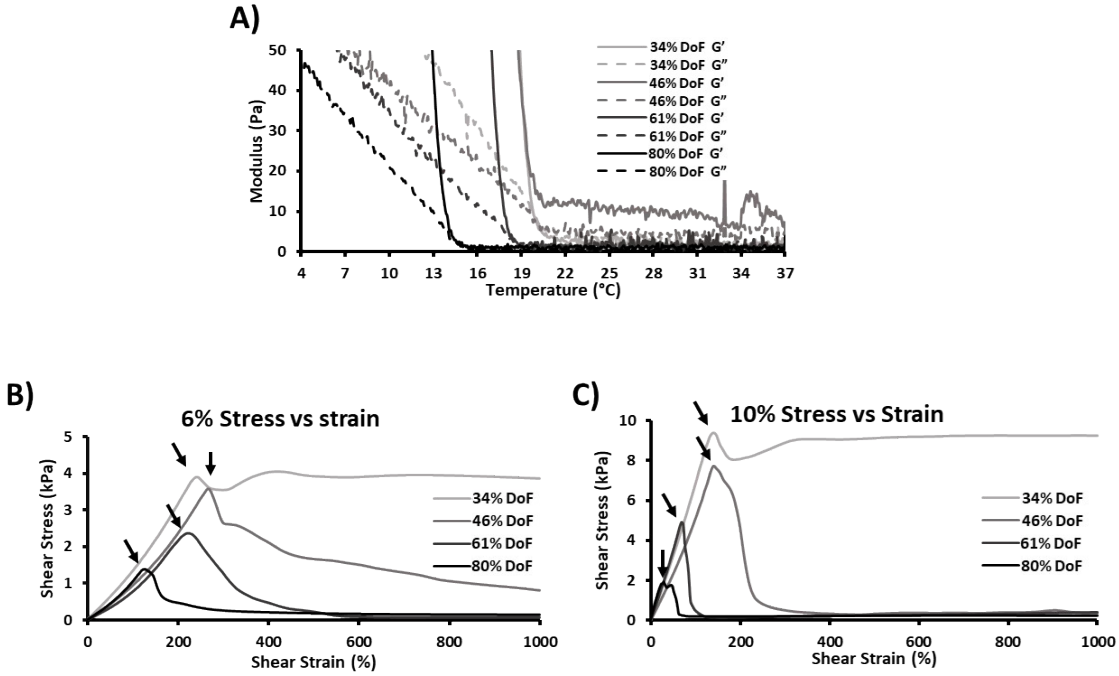
A) Nanostructured properties as determined through SAXs regarding i) mass fractal dimension, ii) correlation length, and iii) mesh size. B) Thermo-dependent viscoelastic properties of pre-crosslinked GelMA, including i) effective temperature of gel formation, ii) storage modulus at 4°C, and iii) gradient of storage modulus increase.

Figure S5. Viscoelastic Properties of GelMA Hydrogels at Different DoF and Concentration



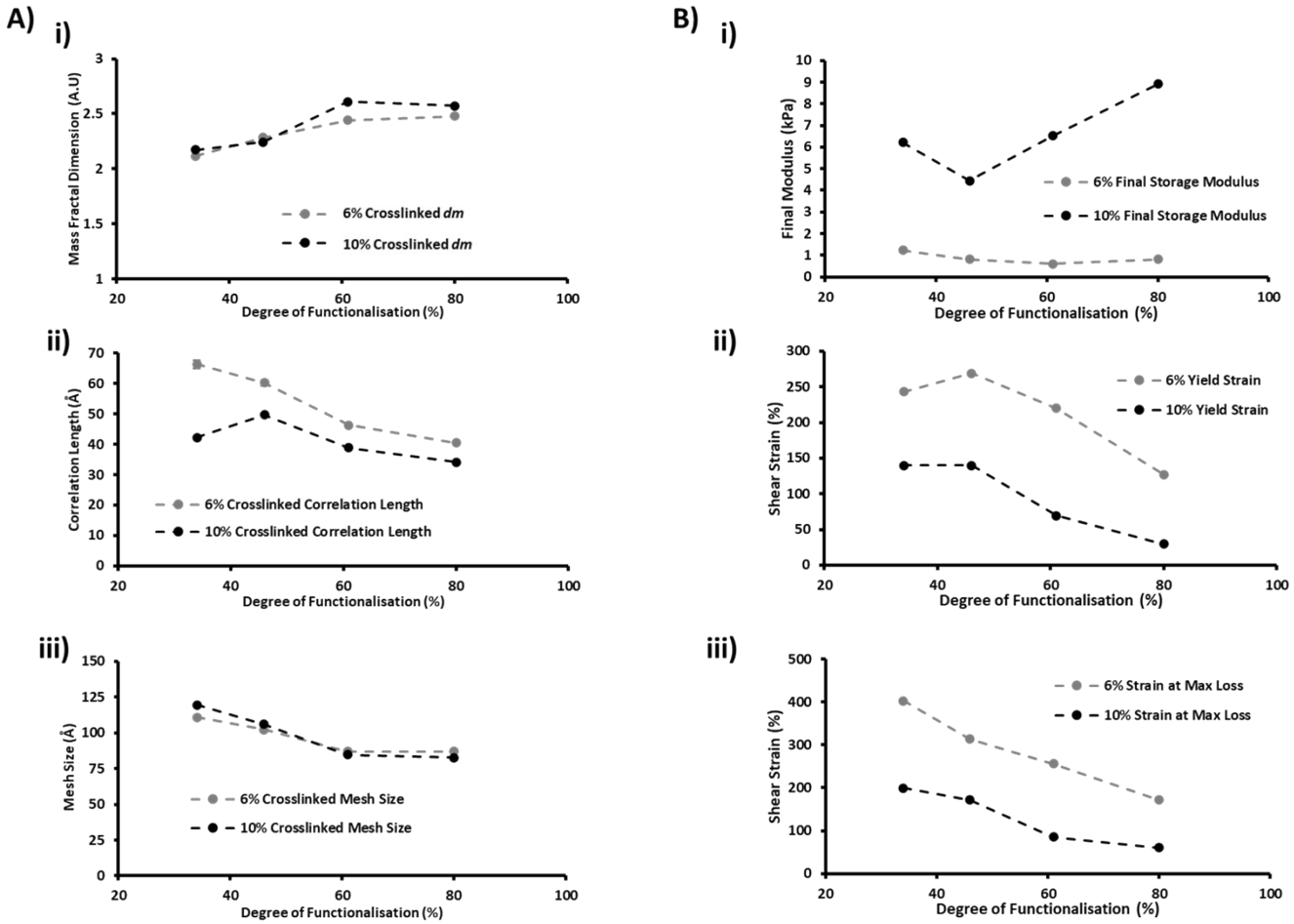
A) G' and G'' response to cooling in i) 6% GelMA and ii) 10% GelMA pre-crosslinked samples. B) G' and G'' response to i) crosslinking in 6% GelMA samples and ii) shear-strain in 6% crosslinked GelMA samples. C) G' and G'' response to i) crosslinking in 10% GelMA samples and ii) shear-strain in 10% crosslinked GelMA samples.

Figure S6. Viscoelastic Properties of GelMA with Differing DoFs



A) G' and G'' response to temperature change showing lack of traditional fluidic state in some DoF samples. B) Complex stress response to strain in 6% GelMA samples showing effective yield point (arrows). C) Complex stress response to strain in 10% GelMA samples showing effective yield point (arrows).

Figure S7. Nanostructured and Viscoelastic Properties of Crosslinked GelMA



A) Nanostructured propertied as determined though SAXS regarding i) mass fractal dimension, ii) correlation length, and iii) mesh size. B) Crosslinked viscoelastic propertied including final modulus, yield strain, and strain at max loss modulus.

Table S1. Comparison of Loss Tangent < 1 and Loss Tangent < 0.1 in GelMA Hydrogels

A)

DoF	Loss tangent < 1	Loss tangent < 0.1
34	16.41666667	14.92667
46	16.41333333	14.76
61	13.68333333	12.11333
80	N/A	8.066667

6% wt. vol.$^{-1}$

B)

DoF	Loss tangent < 1	Loss tangent < 0.1
34	19.96333333	17.73333
46	N/A	17.56667
61	18.39666667	16.41333
80	N/A	12.12

10% wt. vol.$^{-1}$

A) Comparison of loss tangent values in 6% (w/v) GelMA samples. B) Comparison of loss tangent in 10% (w/v) GelMA samples.

Table S2. Molecular Weight and Number of 34% DoF and 80% DoF GelMA Samples

C)

	34% DoF	80% DoF
Molecular Weight (Da)	107,464	62,688
Molecular Number	39,171	19,666
PDI (MW/MN)	2.743	3.188

Table S3A – Values used in Correlation Study

Source	GelMA conc.	DoF	Loss Tan <1	Loss Tan <0.1	Stiffness at 4°C	Grad. of Stiffness Increase	Crosslinked Stiffness	Stress Peak	Yield Strain	Strain at Max Loss
1	6	34	16.41667	14.92667	2264.367	135.5769	1230.267	3911.2	243	403
2	6	46	16.41333	14.76	1376.767	85.8837	823.1667	3574.333	269	313
2	6	61	13.68333	12.11333	964.9867	84.71998	603.4867	2358.067	220	256
1	6	80	N/A	8.066667	400.6167	85.83634	815.4967	1402.233	127	171
1	10	34	19.96333	17.73333	9055.8	419.9832	6221.7	9369.967	778	199
2	10	46	N/A	17.56667	5838.033	274.4133	4437.533	7713.7	140	171
2	10	61	18.39667	16.41333	5545.067	267.8109	6515.467	4919.633	69.5	84.96667
1	10	80	N/A	12.12	2962.033	256.7342	8904.9	1939.51	29.7	59.93333

Table S3B – Values used in Correlation Study (Continued)

Frac. Dimension (Pre-Crosslinked)	SD	Frac. Dimension (Crosslinked)	SD	Correlation Length (Pre-Crosslinked)	SD	Correlation Length (Crosslinked)	SD	Mesh Size (Pre-Crosslinked)	SD	Mesh Size (Crosslinked)	SD
1.7889	0.005875	2.1153	0.004937	42.469	0.56841	66.279	1.327	143.0142	1.06804	110.797	1.282603
1.8621	0.00584	2.2854	0.004671	50.637	0.81012	60.255	0.96307	143.2947	1.311241	102.3203	0.891267
1.8928	0.005391	2.4414	0.004479	48.303	0.65922	46.304	0.47106	143.0435	1.155761	86.91638	0.390993
1.7796	0.005901	2.4781	0.004216	39.188	0.47049	40.53	0.31737	141.9833	0.966534	87.07538	0.314095
1.6009	0.00445	2.1714	0.003799	29.114	0.19861	42.276	0.31659	130.3349	0.504816	119.459	0.541974
1.624	0.004739	2.2436	0.004076	30.071	0.22889	49.711	0.51078	135.7265	0.593627	105.9095	0.616954
1.747	0.004378	2.6103	0.003603	34.417	0.26345	38.842	0.23372	136.1323	0.601951	84.6414	0.232579
1.6498	0.00507	2.5738	0.003056	30.321	0.24168	34.13	0.14916	130.5271	0.584045	82.63544	0.160888