

## **Electronic supplementary information for**

# **Injectable oligomer-cross-linked chitosan hydrogels for biomedical applications**

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## **ADDITIONAL RESULTS**

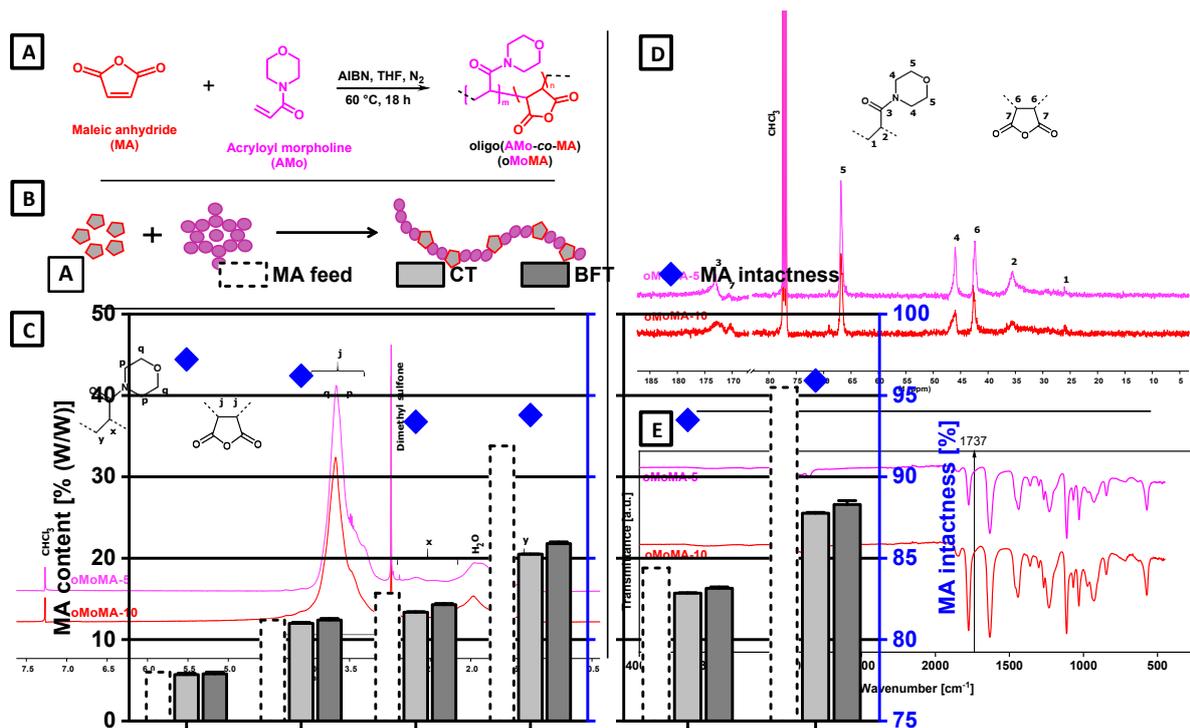
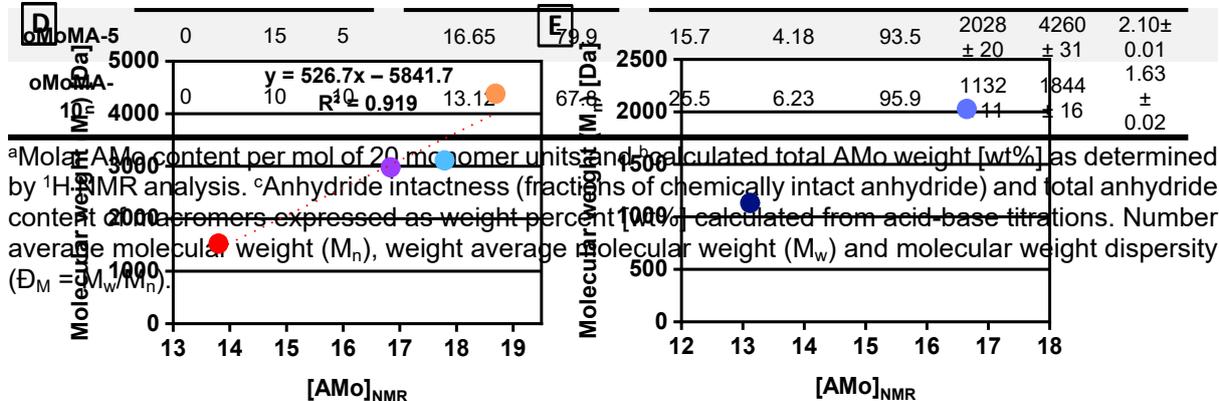
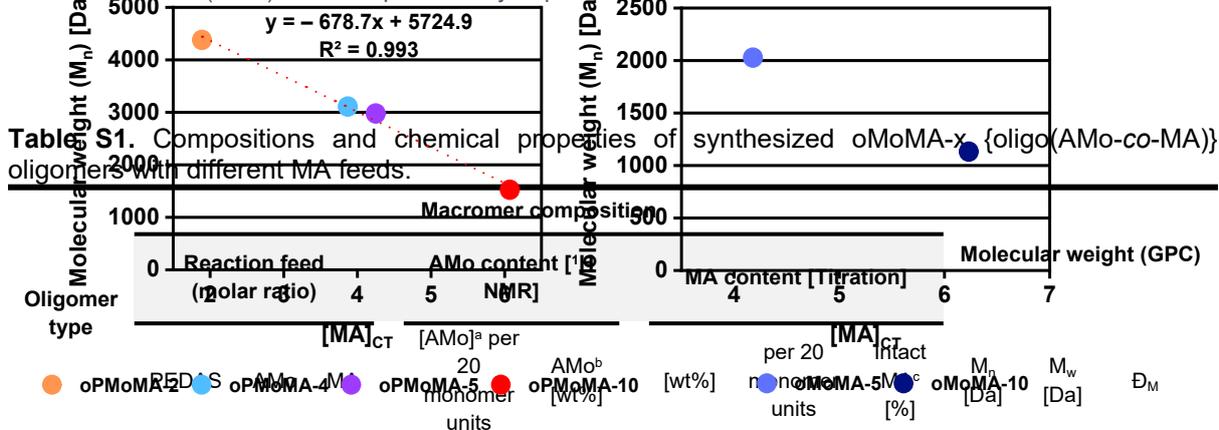
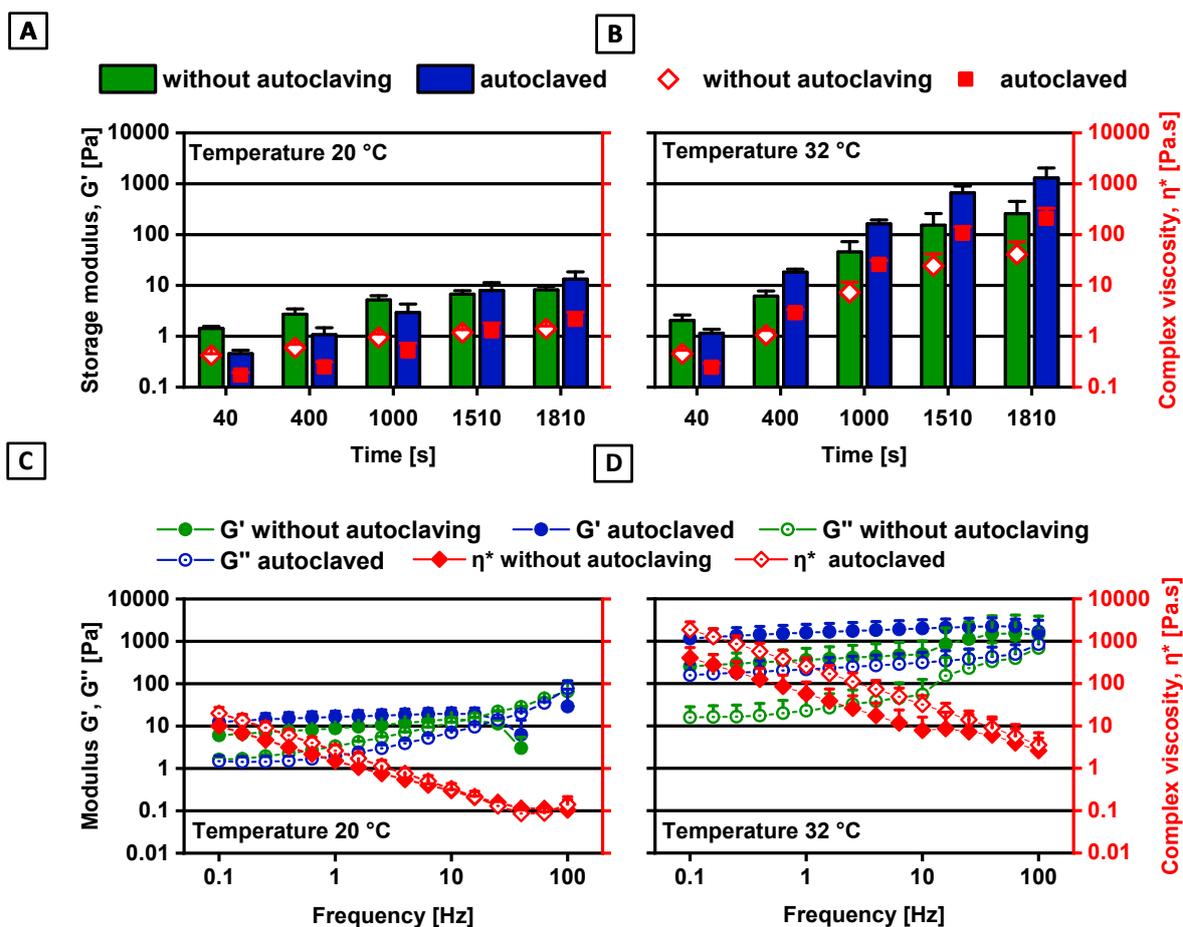


Figure S1: Set of hydrophilic oligomeric macromer without P2DAS. (A) Chemistry of oligomeric macromers, oMoMA {oligo(AMo-co-MA)} synthesized from AMo and MA free radical polymerization. (B) Scheme of oMoMA-x structure. (C) Stacked  $^1\text{H}$  NMR spectra of oMoMA-5 and oMoMA-10 with dimethyl sulfone as an internal reference. 11 and 12 represent the integrals used to determine macromer composition. (D) Stacked, representative  $^{13}\text{C}$  NMR spectra of oMoMA-x with peaks labeled with digits representing specific carbons of the structural components. (E) Fourier transform infrared (FTIR) stacked spectra of hydrophilic oligomeric macromers oMoMA-x.



<sup>a</sup>Molar AMo content per mol of 20-monomer units and calculated total AMo weight [wt%] as determined by  $^1\text{H}$  NMR analysis. <sup>c</sup>Anhydride intactness (fractions of chemically intact anhydride) and total anhydride content in macromers expressed as weight percent [%] calculated from acid-base titrations. Number average molecular weight ( $M_n$ ), weight average molecular weight ( $M_w$ ) and molecular weight dispersity ( $\bar{M}_w = M_w/M_n$ ).

Figure S2: (A) Anhydride content (left y-axis) of hydrophilic oligomeric macromers, theoretical  $MA_{feed}$  and experimentally determined by conductometric ( $MA_{CT}$ ) and Brown Fujimori titration ( $MA_{BFT}$ ) as well as calculated chemically intact MA content (right y-axis). Columns with error bars represent means  $\pm$  standard deviations ( $n = 3$ ). Correlation analysis of [MA]CT (anhydride content as determined by conductometric titrations) vs molecular weight (Mn) (as determined by GPC of (B) oPMoMA-x, and (C) oMoMA-x. AMo content as determined by proton NMR vs molecular weight (Mn) of newly synthesized hydrophilic macromers (D) oPMoMA-x, and (E) oMoMA-x. An indirect relation was found between copolymerized MA content and corresponding synthesized oligomeric macromers molecular weight ( $R2 = 0.993$  indicating linear regressions for oPMoMA-x) whereas a direct correlation was seen between obtained AMo content and molecular weight ( $R2 = 0.919$  indicating linear regressions for oPMoMA-x).



Parameter	Time sweep rheology	Frequency sweep rheology
Measuring Device: (Anton Paar Rheometer)	MCR301 SN80578772	
Measuring Geometry:	CP50-1-SN18839 d = 0.103 mm	
Time Setting:	60 Measurements Points Measurements Points Duration 30 s Amplitude gamma = 1 %	16 Measurements Points Amplitude gamma = 1 %
Measuring Profile:	Frequency f = 1 Hz T [-1] = 20 °C and 32 °C	Frequency f = 0.1 ... 100 Hz log;  Slope  = 5 Pt. / dec T [-1] = 20 °C and 32 °C

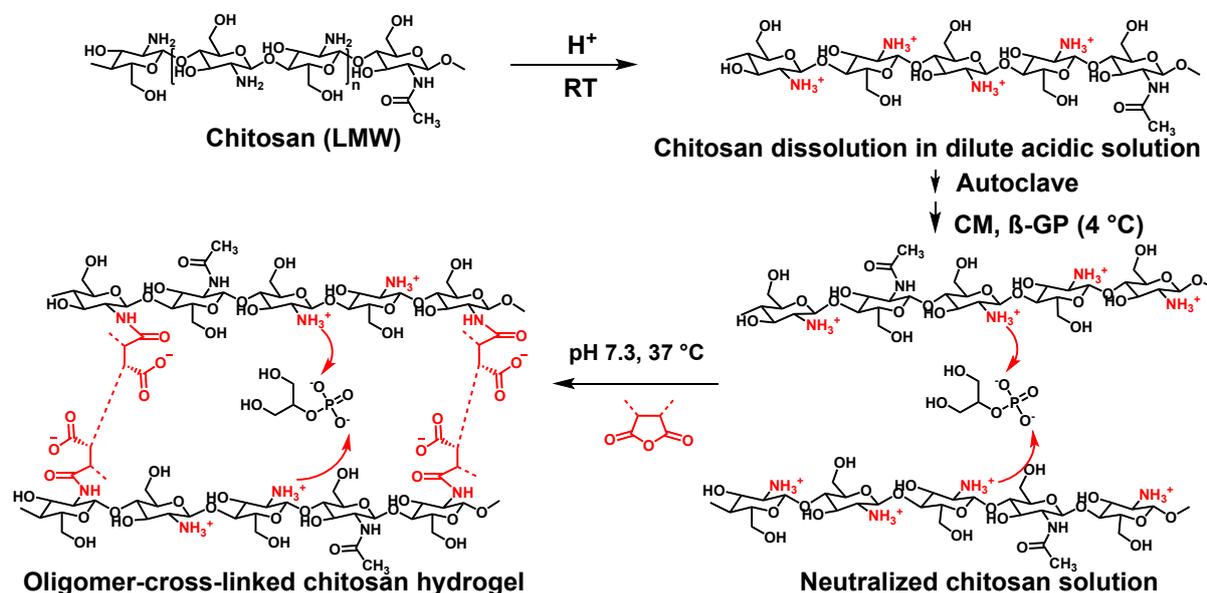
Figure S3: Rheological characterization of pH adjusted pristine chitosan solution with and without autoclaving to investigate effect of autoclaving on viscoelastic properties of chitosan. Time sweep profiles (logarithmic plot) storage modulus ( $G'$ ) (left y-axis) and complex viscosity ( $\eta^*$ ) (right y-axis) are shown as determined at 1 Hz at (A)

20 °C and (B) 32 °C. Frequency sweep profiles (logarithmic scale) representing storage ( $G'$ ) and loss modulus ( $G''$ ) (left y-axis) and complex viscosity ( $\eta^*$ ) (right y-axis) measured as a function of the oscillation frequency at (C) 20 °C and (D) 32 °C. Columns and data points with error bar represent means  $\pm$  standard deviations ( $n = 4$ ). Bottom: Table with the experimental set up (parameter, instrument description and method summary).

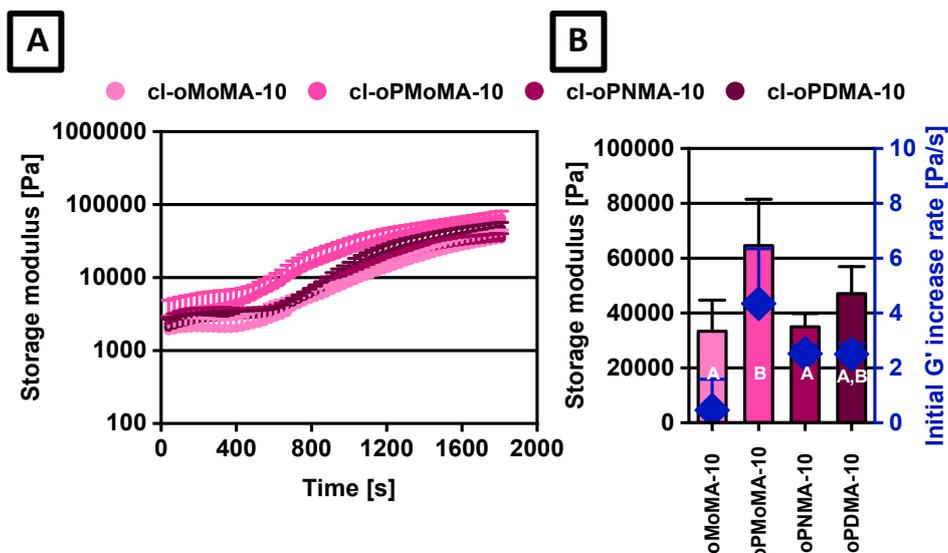
**Table S2:** Reaction conditions (70  $\mu$ l of 1.65 % chitosan solution and 7  $\mu$ l of 40% macromer solution), calculation of cross-linking group ratio and storage modulus (oscillation rheology) at 37 °C, 1 Hz

Chitosan Total free $-\text{NH}_2$ [ $\mu\text{mol}$ ] <sup>a</sup>	Intact MA moiety [ $\mu\text{mol}$ ] <sup>b</sup>			
	oPMoMA-10	oPMoMA-5	oPMoMA-4	oPMoMA-2
5.47	5.38	3.49	3.22	1.54
	Cross-linking group ratio [MA] / [ $\text{NH}_2$ ]			
	0.98	0.64	0.59	0.28
	Storage modulus, $G'$ [Pa]			
405	64,565	140,150	112,750	47,580

<sup>a</sup>Chitosan total free  $-\text{NH}_2$  group calculated from low molecular weight chitosan ( $4.718 \text{ mmol g}^{-1}$ ), considering 4  $-\text{NH}_2$  group per pentasaccharide moiety of 80% deacetylated chitosan. <sup>b</sup>Total intact MA content was calculated from acid-base titration values of macromers.



**Figure S4:** Chemical illustration of injectable oligomer-cross-linked chitosan hydrogel (iCsgel) fabrication from low molecular weight (LMW) chitosan and reactive anhydride-containing oligomer.



	Time [s]								
	40 s			400 s			1810 s		
	<sup>a</sup> S.M. [Pa]	<sup>b</sup> L.M. [Pa]	<sup>c</sup> C.V. [Pa·s]	S.M. [Pa]	L.M. [Pa]	C.V. [Pa·s]	S.M. [Pa]	L.M. [Pa]	C.V. [Pa·s]
<b>oMoMA-10</b>	1861.5	552.9	309.1	2025.3	522.2 ±	333 ±	33353.3	7044.7 ±	5426 ±
	±	±	±	± 741.9	283.2	125.7	±	± 2140.9	± 1836.8
	1073.9	300.1	177.2	4440.0	1035.3	726.1	11348.7	15905.0	10584.3
<b>oPMoMA-10</b>	2875.8	997.2	484.9	±	± 790.5	±	±	± 2945.0	± 2718.0
	±	±	±	±	±	±	±	±	±
	2000.7	873.7	346.2	2479.4	± 790.5	412.8	16928.3	± 2945.0	± 2718.0
<b>oPNMA-10</b>	2321.3	608 ±	382 ±	3227 ±	767.6 ±	528.4	34986.7	7921.3 ±	5709.3 ±
	±	± 139.4	± 91.4	± 539.8	± 42.7	± 81.9	± 4828.2	± 585.2	± 769.9
	557.2	139.4	91.4	539.8	42.7	± 81.9	± 4828.2	585.2	769.9
<b>oPDMA-10</b>	2084 ±	925.6	363.6	2984.3	1316.9	520.2	47053.3	15476.7	7883 ±
	± 598.4	± 135.7	± 93.6	± 489.6	± 339	± 85.7	± 9877.2	± 3296.3	± 1656.1
	598.4	135.7	± 93.6	± 489.6	± 339	± 85.7	± 9877.2	± 3296.3	1656.1

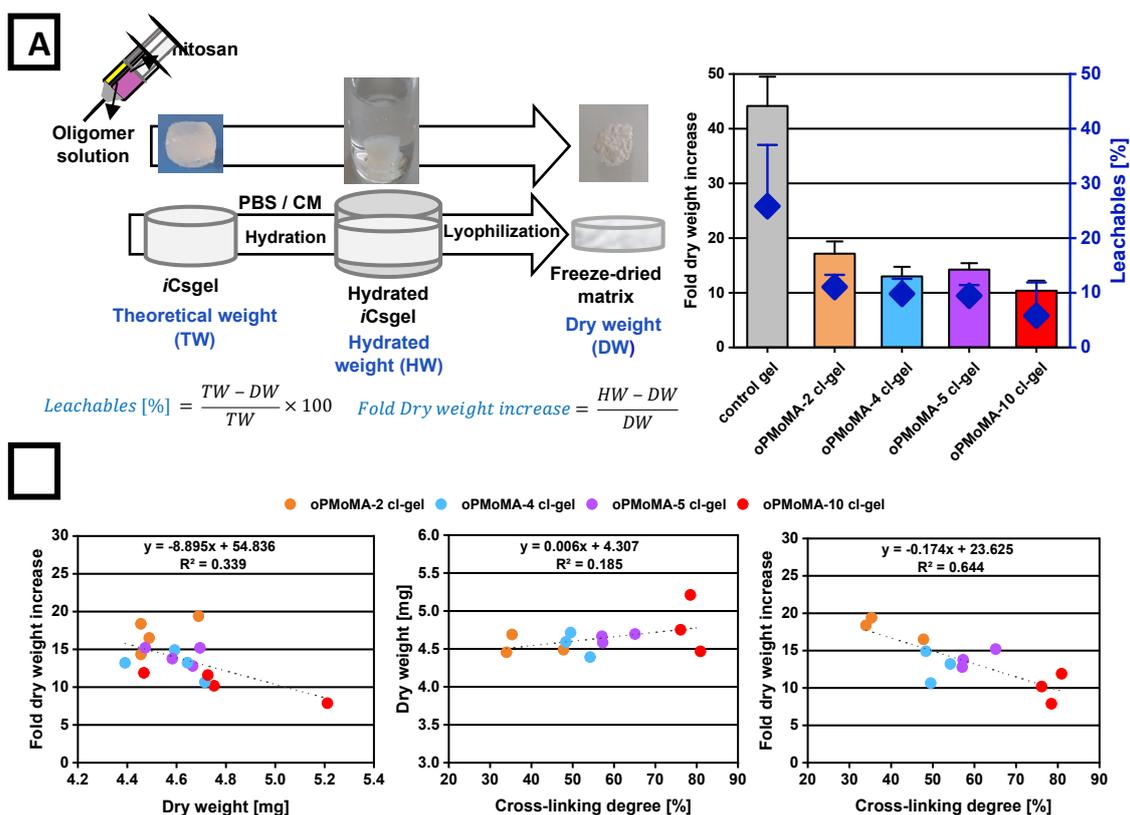
<sup>a</sup>Storage modulus (G'), <sup>b</sup>Loss modulus (G''), <sup>c</sup>Complex Viscosity ( $\eta^*$ )

The last point of the profiles (G' at 1810 s)

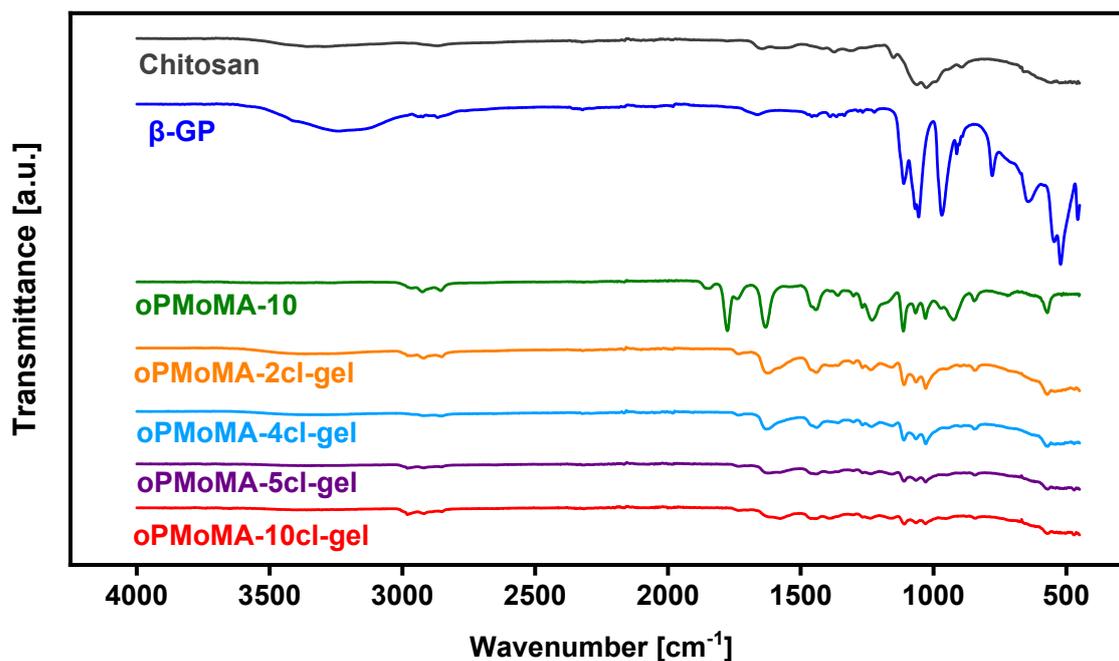
	1	2	3	Mean	S.D.
<b>oMoMA-10</b>	46150	24510	29400	33353.3	11348.7
<b>oPMoMA-10</b>	25630	28050	24790	26156.7	1692.6
<b>oPNMA-10</b>	31630	40520	32810	34986.7	4828.2
<b>oPDMA-10</b>	48690	56010	36460	47053.3	9877.2

Parameter	Instrument description and method summary
Measuring Device: (Anton Paar Rheometer)	MCR301 SN80578772
Measuring Geometry:	CP25-1-SN18554 d=0.048 mm
Time Setting:	60 Measurements Points Measurements Points Duration 30 s
Measuring Profile:	Amplitude $\gamma = 1\%$ Frequency $f = 1$ Hz T [-1] = 37 °C

**Figure S5:** Rheological characterization of the cross-linking reaction between reactive anhydride (MA-10) containing macromers and amine containing chitosan solution as determined at 1 Hz. (A) Rheograms depicting reaction between chitosan and MA-10 containing oligomers. Time sweep profiles (logarithmic scale) of storage moduli ( $G'$ ) for cross-linking reaction with oPMoMA-10, oMoMA-10, oPNMA-10 and oPDMA-10. (B) The last point of the profiles ( $G'$  at 1810 s) are plotted separately (columns, left y-axis) and initial rate of modulus ( $G'$ ) increase calculated for each reaction mixtures (diamonds, right y-axis). Data points, columns, diamonds with error bars represent means  $\pm$  standard deviations ( $n = 4$ ). Columns with different letters are statistically significantly different ( $p < 0.05$ ). Bottom: 1st Table with the experimental results at different time points. Bottom: 2nd Table with the experimental set up.



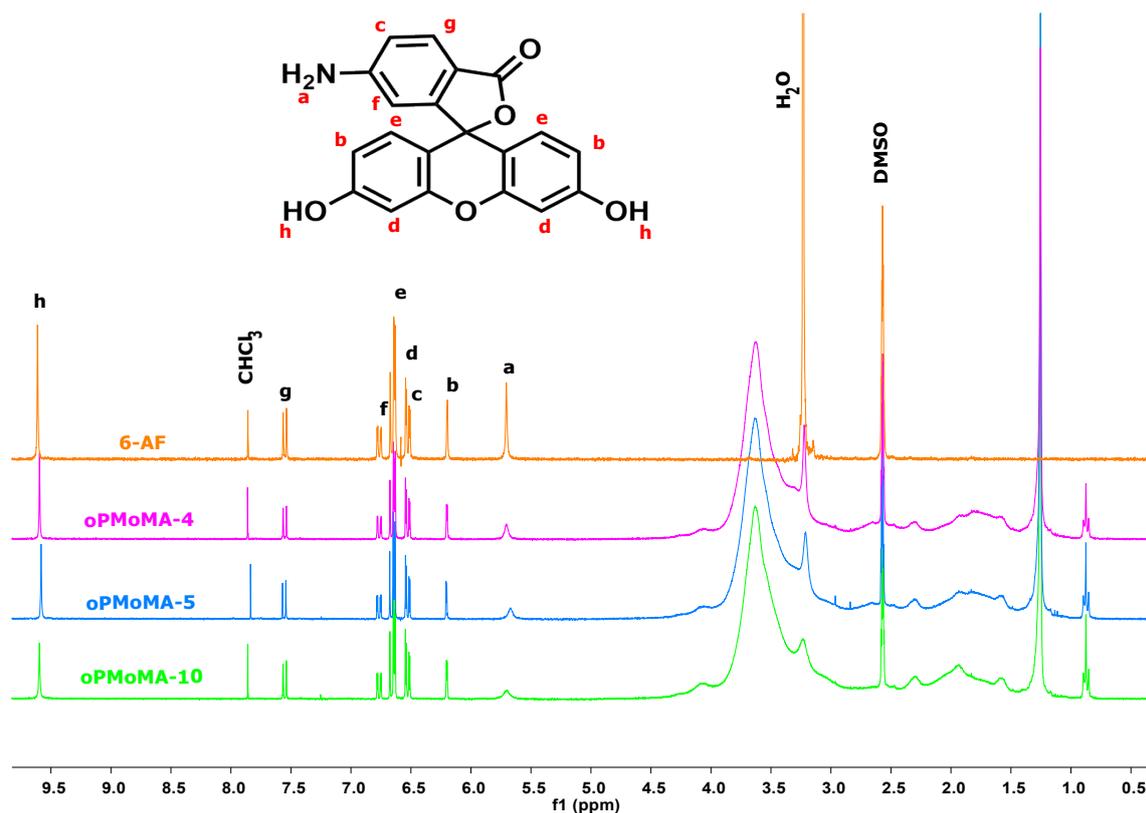
**Figure S6:** (A) Physico-chemical properties of *i*Csgel cross-linked with a selected set of hydrophilic oligomers (oPMoMA-x) in comparison to control gel (physically gelled chitosan). Fold dry weight increase (equilibrium water content) calculated from the hydrated gel and leachables were determined from the lyophilized gel discs. Columns and data points with error bars represent means  $\pm$  standard deviations ( $n = 4$ ). (B) Correlation analysis between water content vs dry weight, dry weight vs cross-linking degree and water content vs cross-linking degree (from left to right) of all tested *i*Csgel cross-linked with oPMoMA-x. An indirect correlation was found between water content and dry weight ( $R^2 = 0.339$ , linear regression), relation between dry weight and cross-linking degree ( $R^2 = 0.185$ , linear regression) as well as indirect correlation between water content and cross-linking degree ( $R^2 = 0.644$ , linear regression).



Before covalent bond formation	Wavenumber [cm <sup>-1</sup> ]		Functional group
Chitosan	1548 cm <sup>-1</sup>		amino group of chitosan
Anhydride of the macromer (Saturated cyclic anhydrides e.g., succinic anhydride)	1650 cm <sup>-1</sup>		amide I peak
	1777 cm <sup>-1</sup>		asymmetric C=O stretch
	1850 cm <sup>-1</sup>		symmetric C=O stretch
Bands from anhydrides and amines yielding chemically cross-linked chitosan-based hydrogel (oPMoMA-x cl-gels)			
After covalent bond formation	Wavenumber [cm <sup>-1</sup> ]		Functional group
Chitosan peak disappears	1548 cm <sup>-1</sup>	No peak	amino group of chitosan
	1650 cm <sup>-1</sup>	1628 cm <sup>-1</sup>	amide I peak
<sup>1</sup> Anhydride peaks disappear	1777 cm <sup>-1</sup>	No peak	asymmetric C=O stretch
	1850 cm <sup>-1</sup>	No peak	symmetric C=O stretch
New or more prominent peaks appear in anhydride containing macromer cross-linked chitosan-based hydrogel *oPMoMA-x cl-gels		1628 cm <sup>-1</sup>	-O=C-N [Amide I]
		1445 cm <sup>-1</sup>	primary amide [Amide VI]
		1235 cm <sup>-1</sup>	(NH) + (-O=C-N) [Amide III]
		573 cm <sup>-1</sup>	(C-C) + (-O=C-N) [Amide IV]

<sup>1</sup>See Figure 1E and S5E for original anhydride peaks in oPMoMA-x and oMoMA-x FTIR spectra.

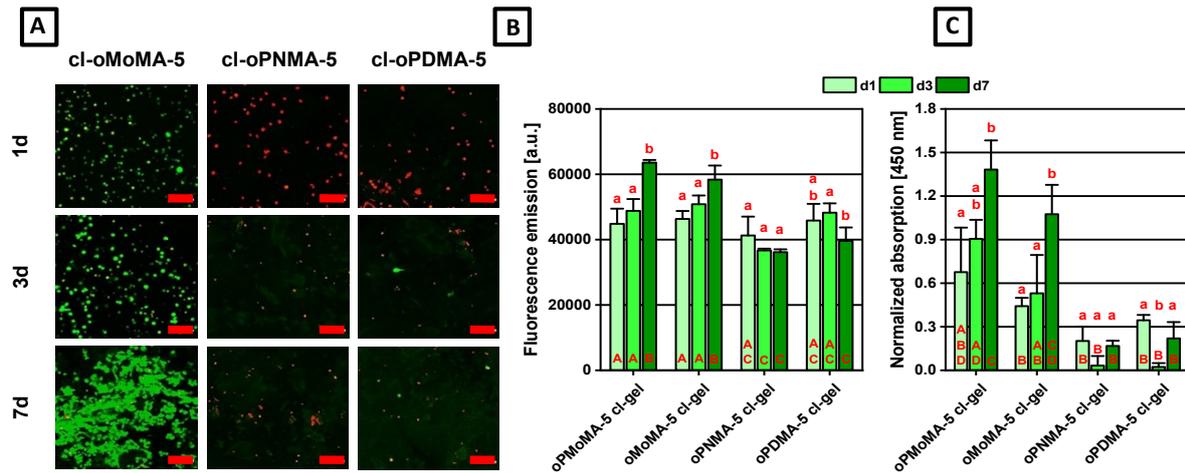
Figure S7: Stacked FTIR spectra of oPMoMA-x cross-linked chitosan hydrogel in comparison to pristine chitosan,  $\beta$ -glycerophosphate ( $\beta$ -GP) and oPMoMA-10 oligomer. Bottom: Table with the experimental results.



Macromer	MA <sub>eq</sub>	Ratio of oPMoMA derivatized
Unmodified macromer cl-gel	0	0
oPMoMA-4 <sup>+6-AF</sup> cl-gel	1	0.25
oPMoMA-5 <sup>+6-AF</sup> cl-gel	0.5, *1.0, 1.5, 2.0	0.1, *0.2, 0.3, 0.4
oPMoMA-10 <sup>+6-AF</sup> cl-gel	1	0.1

\* oPMoMA-5 MA<sub>eq</sub> = 1.0 of 6-AF has 0.2 Ratio of oPMoMA-5 derivatized.

**Figure S8:** Representative <sup>1</sup>H NMR spectra of 6-aminofluorescein (6-AF) and 6-AF-modified oPMoMA-x macromers with MA<sub>eq</sub> = 1. The letter assigned to the peaks correspond to the protons at the positions labeled in the structural element of 6-AF. Label 'a' in spectra corresponds to -NH<sub>2</sub> group of pristine 6-AF and 6-AF modified oPMoMA-x. Significant sharp peak reduction of amino group together with significant reduction in hydroxyl peak at 9.6 ppm in spectra of derivatized oPMoMA-x<sup>+6-AF</sup> clearly indicates successful covalent attached of 6-AF to macromers through amide bond formation.



**Figure S9:** Cytocompatibility of encapsulated L929 fibroblast cells in *iCsgel* upon cross-linking with MA-5-type oligomers. (A) Stacked CLSM images of cell-laden *iCsgel* at day 1, 3 and 7 after staining with Calcein-AM and EthD-1 (Live/Dead<sup>®</sup> assay) (scale bar 100  $\mu\text{m}$ ). (B) Metabolic activity as a measure of fluorescence emission (AlamarBlue<sup>™</sup> assay, left image) and (C) normalized absorption (WST-8 assay, right image) of encapsulated L929 fibroblast cells at day 1, 3 and 7. Columns with different letters represent means  $\pm$  standard deviations ( $n = 4$ ). Columns with different letters are statistically significantly different, small letters indicate differences within groups and capital letters indicate difference of respective formulations between groups,  $p < 0.05$ .

**DATA PRESENTED IN THE MANUSCRIPT**

Table S3: Time sweep rheology instrument description and method summary. Bottom: Tables with the experimental results at different time points.

Parameter	Instrument description and method summary
Measuring Device: (Anton Paar Rheometer)	MCR301 SN80578772
Measuring Geometry:	CP25-1-SN18554 d=0.048 mm
Time Setting:	60 Measurements Points Measurements Points Duration 30 s
Measuring Profile:	Amplitude gamma = 1 % Frequency f = 1 Hz T [-1] = 37 °C

The last point of the profiles (G' at 1810 s)

	1	2	3	4	Mean	S.D.
<b>Control</b>	352.4	507.2	300.3	456	404.0	94.5
<b>oPMoMA-2</b>	61700	51940	36040	40630	47577.5	11545.0
<b>oPMoMA-4</b>	115100	150900	76200	108800	112750.0	30618.3
<b>oPMoMA-5</b>	140100	140800	135800	143900	140150.0	3337.2
<b>oPMoMA-10</b>	86550	67530	57380	46790	64562.5	16928.3
<b>oMoMA-5</b>	141400	126100	147300	110400	131300.0	16551.5
<b>oPNMA-5</b>	85270	111600	140800	106800	111117.5	22862.0
<b>oPDMA-5</b>	48610	59560	66100	32000	51567.5	14907.6

Initial G' increase rate [Pa/s]

	1	2	3	4	Mean	S.D.
<b>Control</b>	0.12	0.20	0.16	0.19	0.17	0.04
<b>oPMoMA-2</b>	5.57	1.09	0.84	1.04	2.14	2.29
<b>oPMoMA-4</b>	4.52	2.05	2.29	2.57	2.86	1.13
<b>oPMoMA-5</b>	19.16	43.68	9.50	7.85	20.05	16.52
<b>oPMoMA-10</b>	5.10	5.12	1.38	5.78	4.35	2.00
<b>oMoMA-5</b>	5.44	4.73	5.90	19.85	8.98	7.27
<b>oPNMA-5</b>	35.18	10.74	19.16	15.53	20.15	10.59
<b>oPDMA-5</b>	9.68	19.52	12.17	10.05	12.85	4.58

*Table S4:* Complex viscosities ( $\eta^*$ ) (columns left y-axis) and loss modulus ( $G''$ ) (diamonds, right y-axis) determined at different time points from rheological data as a measure of gelation kinetics after initiation of cross-linking reaction with oPMoMA-x in comparison to control.

	Time [s]	loss modulus ( $G''$ ) [Pa]					Complex viscosities ( $\eta^*$ ) [Pa*s]						
		1	2	3	4	Mean	S.D.	1	2	3	4	Mean	S.D.
<b>Control</b>	40	2.9	13.5	19.2	4.2	10.0	7.8	6.2	17.1	23.2	7.4	13.5	8.2
	400	4.6	16.1	22.4	6.8	12.4	8.3	12.8	28.3	32.2	18.5	22.9	8.9
	1000	8.3	25.4	24.5	12.0	17.5	8.7	24.9	50.0	40.5	36.2	37.9	10.4
	1510	13.4	30.6	18.9	19.2	20.5	7.2	40.1	63.2	45.0	57.2	51.4	10.7
	1810	19.6	38.0	34.3	24.7	29.2	8.5	56.2	81.0	48.1	72.7	64.5	15.0
<b>oPMoMA-2-cl</b>	40	6.6	13.9	12.1	12.2	11.2	3.1	3.9	8.9	7.0	5.5	6.3	2.1
	400	360.7	33.9	26.9	27.7	112.3	165.6	327.9	71.1	55.3	65.1	129.8	132.2
	1000	3500.0	1846.0	1667.0	518.0	1882.8	1228.3	3170.0	1682.0	1223.0	654.2	1682.3	1077.2
	1510	10640.0	5881.0	4715.0	2382.0	5904.5	3476.0	8041.0	4819.0	3728.0	3425.0	5003.3	2111.8
	1810	13900.0	11000.0	7724.0	6681.0	9826.3	3280.5	10070.0	8450.0	5866.0	6554.0	7735.0	1901.9
<b>oPMoMA-4-cl</b>	40	14.1	19.1	17.2	11.1	15.4	3.5	8.2	9.7	9.9	7.4	8.8	1.2
	400	282.9	235.6	108.9	204.6	208.0	73.5	270.7	132.2	141.8	157.7	175.6	64.3
	1000	9895.0	13320.0	4008.0	6830.0	8513.3	4006.0	6619.0	7566.0	3267.0	4655.0	5526.8	1933.7
	1510	21510.0	28380.0	11450.0	19170.0	20127.5	6981.7	15090.0	19220.0	8772.0	12950.0	14008.0	4354.0
	1810	26200.0	33610.0	16230.0	25730.0	25442.5	7123.5	18790.0	24610.0	12400.0	17790.0	18397.5	5002.9
<b>oPMoMA-5-cl</b>	40	272.8	228.0	378.8	170.3	262.5	88.2	112.2	88.3	137.9	77.1	103.9	27.0
	400	1746.0	3455.0	969.7	673.0	1710.9	1247.6	1233.0	2641.0	686.1	533.0	1273.3	960.0
	1000	15230.0	21060.0	15570.0	14070.0	16482.5	3118.5	10780.0	14840.0	11180.0	10270.0	11767.5	2081.9
	1510	25840.0	28370.0	25370.0	27240.0	26705.0	1364.9	18640.0	20280.0	18430.0	19580.0	19232.5	858.9
	1810	31270.0	32080.0	30250.0	32620.0	31555.0	1031.8	22850.0	22980.0	22140.0	23480.0	22862.5	553.0
<b>oPMoMA-10-cl</b>	40	578.6	582.3	520.8	2307.0	997.2	873.7	301.1	372.1	266.5	1000.0	484.9	346.2
	400	711.7	782.4	446.2	2201.0	1035.3	790.5	589.7	665.6	339.9	1309.0	726.1	412.8
	1000	6787.0	5618.0	2000.0	8957.0	5840.5	2910.2	4959.0	3677.0	1474.0	5331.0	3860.3	1741.5
	1510	14850.0	11890.0	9792.0	14910.0	12860.5	2484.4	10840.0	8102.0	6471.0	8229.0	8410.5	1806.7
	1810	20150.0	15640.0	13850.0	13980.0	15905.0	2945.0	14140.0	11030.0	9394.0	7773.0	10584.3	2718.0

Complex viscosities ( $\eta^*$ ) (columns left y-axis) and loss modulus ( $G''$ ) (diamonds, right y-axis) determined at different time points from rheological data as a measure of gelation kinetics after initiation of cross-linking reaction with different MA-5 containing oligomers.

	Time [s]	loss modulus ( $G''$ ) [Pa]					Complex viscosities ( $\eta^*$ ) [Pa*s]						
		1	2	3	4	Mean	S.D.	1	2	3	4	Mean	S.D.
<b>oMoMA-5-cl</b>	40	16.3	7.3	22.1	16.1	4.3	6.1	19.5	6.9	23.1	8.1	14.4	8.1
	400	304.9	264.4	761.4	1244.0	2292.0	459.3	334.4	280.8	380.5	1202.0	549.4	437.0
	1000	14450.0	10960.0	11520.0	16400.0	17860.0	2554.2	9052.0	6689.0	6942.0	9406.0	8022.3	1404.7
	1510	26150.0	25240.0	25720.0	28740.0	26540.0	1563.2	18290.0	15850.0	17640.0	15300.0	16770.0	1423.0
	1810	31340.0	30620.0	32740.0	35610.0	30320.0	2205.0	23050.0	20650.0	24020.0	18220.0	21485.0	2597.0
<b>oPNMA-5-cl</b>	40	1151.0	737.1	799.6	594.8	820.6	236.3	558.8	396.6	396.0	321.4	418.2	100.2
	400	4307.0	1343.0	2344.0	1954.0	2487.0	1281.4	2634.0	1017.0	1519.0	1237.0	1601.8	718.2
	1000	10320.0	9133.0	17750.0	11740.0	12235.8	3827.5	6815.0	6055.0	11180.0	8018.0	8017.0	2258.2
	1510	17850.0	21730.0	28370.0	19780.0	21932.5	4574.7	11920.0	14220.0	18740.0	13690.0	14642.5	2903.3
	1810	20870.0	27690.0	34080.0	25030.0	26917.5	5538.7	13970.0	18290.0	23060.0	17460.0	18195.0	3744.7
<b>oPDMA-5-cl</b>	40	1178.0	2372.0	1135.0	1505.0	1547.5	574.0	562.0	878.4	616.2	625.4	670.5	141.4
	400	1816.0	4176.0	2143.0	2166.0	2575.3	1079.1	1122.0	2023.0	1331.0	1204.0	1420.0	411.1
	1000	3858.0	6843.0	5829.0	2717.0	4811.8	1867.1	2747.0	4223.0	4123.0	1675.0	3192.0	1215.0
	1510	8337.0	12670.0	11890.0	4566.0	9365.8	3714.2	5927.0	7636.0	8525.0	3141.0	6307.3	2370.2
	1810	11220.0	17020.0	15010.0	7775.0	12756.3	4100.1	7940.0	9858.0	10790.0	5241.0	8457.3	2450.6

Complex viscosities ( $\eta^*$ ) (columns left y-axis) and loss modulus ( $G''$ ) (diamonds, right y-axis) determined at different time points from rheological data as a measure of gelation kinetics after initiation of cross-linking reaction with different MA-10 containing oligomers.

	Time [s]	loss modulus ( $G''$ ) [Pa]					Complex viscosities ( $\eta^*$ ) [Pa*s]						
		1	2	3	4	Mean	S.D.	1	2	3	4	Mean	S.D.
<b>oMoMA-10-cl</b>	40	296.3	882.9	479.4	552.9	300.1	140.2	493.5	293.6	309.1	177.2	40	296.3
	400	304.0	842.2	420.4	522.2	283.2	237.6	475.4	286.1	333.0	125.7	400	304.0
	1000	2136.0	2064.0	2560.0	2253.3	268.0	1195.0	1370.0	1576.0	1380.3	190.7	1000	2136.0

	1510	7125.0	4176.0	4373.0	5224.7	1648.7	4989.0	2958.0	3523.0	3823.3	1048.3	1510	7125.0
	1810	9501.0	5575.0	6058.0	7044.7	2140.9	7499.0	4001.0	4778.0	5426.0	1836.8	1810	9501.0
	40	642.9	454.5	726.6	608.0	139.4	417.8	278.1	450.0	382.0	91.4	40	642.9
	400	733.5	815.5	753.9	767.6	42.7	541.5	440.7	603.0	528.4	81.9	400	733.5
<b>oPNMA-10-cl</b>	1000	3507.0	3688.0	3621.0	3605.3	91.5	1854.0	2064.0	1802.0	1906.7	138.7	1000	3507.0
	1510	6669.0	9646.0	6672.0	7662.3	1717.9	4182.0	5663.0	4342.0	4729.0	812.8	1510	6669.0
	1810	7575.0	8597.0	7592.0	7921.3	585.2	5176.0	6592.0	5360.0	5709.3	769.9	1810	7575.0
	40	928.1	1060.0	788.7	925.6	135.7	299.7	471.1	320.1	363.6	93.6	40	928.1
	400	1336.0	1646.0	968.8	1316.9	339.0	459.0	618.1	483.5	520.2	85.7	400	1336.0
<b>oPDMA-10-cl</b>	1000	6558.0	5691.0	2736.0	4995.0	2003.8	2709.0	2297.0	1385.0	2130.3	677.6	1000	6558.0
	1510	12200.0	15510.0	8446.0	12052.0	3534.3	5825.0	7432.0	3999.0	5752.0	1717.7	1510	12200.0
	1810	16390.0	18220.0	11820.0	15476.7	3296.3	8176.0	9373.0	6100.0	7883.0	1656.1	1810	16390.0

Table S5: Frequency sweep rheology instrument description and method summary. Bottom: Tables with the experimental results.

Parameter	Frequency sweep rheology
Measuring Device: (Anton Paar Rheometer)	MCR301 SN80578772
Measuring Geometry:	PP08-SN5913; [d=1 mm]
Time Setting:	21 Measurements Points Normal Force FN = 0.1 N
Measuring Profile:	Amplitude gamma = 1 % Frequency f = 0.1 ... 10 Hz log;  Slope  = 10 Pt. / dec T [-1] = 37 °C

Storage moduli ( $G'$ ) as determined from equilibrium-swollen (in culture medium) discs of iCsgel cross-linked with oPMoMA-x of increasing MA feed (left image) and with MA-5-type macromers of different comonomer composition, all at 37 °C and 1 Hz (frequency sweep rheology).

	1	2	3	4	Mean	S.D.
<b>oPMoMA-2</b>	2826	3531	3253	3884	3373.5	447.1
<b>oPMoMA-4</b>	6135	6984	6396	5609	6281.0	571.6
<b>oPMoMA-5</b>	7666	7283	7113	6770	7208.0	372.5
<b>oPMoMA-10</b>	9883	9462	8730	10100	9543.8	603.7
<b>oPMoMA-5</b>	7666	7283	7113	6770	7208.0	372.5
<b>oMoMA-5</b>	6293	6510	6702	7034	6634.8	314.3
<b>oPNMA-5</b>	11610	13070	12900	9703	11820.8	1555.1
<b>oPDMA-5</b>	25370	29920	25740	26670	26925.0	2070.2

Cross-linking degrees as determined for lyophilized discs of corresponding formulation.

	1	2	3	Mean	S.D.
<b>oPMoMA-2</b>	47.77	33.97	35.30	39.01	7.61
<b>oPMoMA-4</b>	48.35	54.20	49.50	50.68	3.10
<b>oPMoMA-5</b>	57.28	57.08	65.09	59.82	4.57
<b>oPMoMA-10</b>	76.12	80.90	78.47	78.50	2.39
<b>oPMoMA-5</b>	57.28	57.08	65.09	59.82	4.57
<b>oMoMA-5</b>	28.47	41.12	39.80	36.47	6.95
<b>oPNMA-5</b>	65.36	48.91	58.87	57.72	8.29
<b>oPDMA-5</b>	63.49	65.77	72.61	67.29	4.75

*Table S6:* Porosity of *i*Csgel cross-linked with oPMoMA-x as determined by solvent displacement method. The lyophilized discs were further immersed in absolute ethanol to measure the porosity of *i*Csgel discs by solvent displacement. Bottom: Tables with the experimental results.

	<b>Dry Weight (DW)</b>	<b>Re-hydrated weight (RW)</b>	<b>Volume (mm<sup>3</sup>)</b>
<b>oPMoMA-2 cl-gel</b>	5.78 ± 0.26	44.89 ± 6.92	56.46 ± 8.53
<b>oPMoMA-4 cl-gel</b>	6.75 ± 0.31	34.57 ± 0.64	42.77 ± 3.52
<b>oPMoMA-5 cl-gel</b>	6.71 ± 0.29	34.73 ± 0.70	45.86 ± 6.16
<b>oPMoMA-10 cl-gel</b>	6.66 ± 0.19	33.12 ± 3.30	40.86 ± 2.53

$$\text{Porosity [\%]} = \frac{RW - DW}{\rho V} \times 100$$

DW = Weight of lyophilized iCsgel

RW = Weight of rehydrated iCsgel in absolute ethanol

$$\rho(\text{absolute ethanol}) = 0.789 \frac{\text{g}}{\text{ml}} \vee \frac{\text{mg}}{\text{mm}^3}$$

$$\text{Volume [mm}^3\text{]} = \pi r^2 h$$

Porosity of *i*Csgel cross-linked with oPMoMA-x as determined by solvent displacement method.

	1	2	3	4	Mean	S.D.
<b>oPMoMA-2 cl-gel</b>	90.30	82.69	92.48	85.07	87.63	4.53
<b>oPMoMA-4 cl-gel</b>	87.28	74.53	85.67	83.71	82.79	5.70
<b>oPMoMA-5 cl-gel</b>	71.21	66.23	86.85	90.16	78.61	11.68
<b>oPMoMA-10 cl-gel</b>	73.79	67.82	92.68	96.14	82.61	13.92

Elastic moduli of *i*Csgel (using the general estimation of  $E = 3G'$ ) cross-linked with oPMoMA-x.

	1	2	3	4	Mean	S.D.
<b>oPMoMA-2 cl-gel</b>	8.48	10.59	9.76	11.65	10.12	1.34
<b>oPMoMA-4 cl-gel</b>	18.41	20.95	19.19	16.83	18.84	1.71
<b>oPMoMA-5 cl-gel</b>	23.00	21.85	21.34	20.31	21.62	1.12
<b>oPMoMA-10 cl-gel</b>	29.65	28.39	26.19	30.30	28.63	1.81

Table S7: Thickness-normalized fluorescence intensities of *i*Csgel: control and oPMoMA-x ( $MA_{eq}$  1.0) labeled cross-linked gels.

oPMoMA-x cross-linked chitosan gels (control)	Fluorescence Intensity/height [a.u./mm]				
	1	2	3	Mean	S.D.
<b>oPMoMA-4</b>	389.4	390.6	374.9	385.0	8.7
<b>oPMoMA-5</b>	460.6	427.3	409.6	432.5	25.9
<b>oPMoMA-10</b>	307.5	448.6	488.3	414.8	95.0
<b>oPMoMA-x<sup>+6-AF</sup> cross-linked chitosan gels</b>					
<b>oPMoMA-4<sup>+6-AF</sup> <math>MA_{eq}</math>1.0</b>	4468.5	6306.4	4744.9	5173.3	991.0
<b>oPMoMA-5<sup>+6-AF</sup> <math>MA_{eq}</math>1.0</b>	6549.9	6020.9	5149.9	5906.9	706.9
<b>oPMoMA-10<sup>+6-AF</sup> <math>MA_{eq}</math>1.0</b>	9714.0	6872.2	10502.4	9029.5	1909.4

Labeled *i*Csgel cross-linked with oPMoMA-5 modified with different  $MA_{eq}$  of 6-AF

oPMoMA-5 <sup>+6-AF</sup> cross-linked chitosan gels	Fluorescence Intensity/height [a.u./mm]				
	1	2	3	Mean	S.D.
<b>oPMoMA-5<sup>+6-AF</sup> <math>MA_{eq}</math>0.5</b>	4101.5	4396.8	4195.8	4231.4	150.8
<b>oPMoMA-5<sup>+6-AF</sup> <math>MA_{eq}</math>1.0</b>	6549.9	6020.9	5149.9	5906.9	706.9
<b>oPMoMA-5<sup>+6-AF</sup> <math>MA_{eq}</math>1.5</b>	6063.6	6278.5	6304.0	6215.4	132.0
<b>oPMoMA-5 <math>MA_{eq}</math>2.0</b>	5597.5	7344.3	8102.8	7014.9	1284.7

Table S8: Cytocompatibility testing of encapsulated L929 fibroblast cells in *i*Csgel upon cross-linking with anhydride containing macromers.

Cytocompatibility Test	oPMoMA-2, -4, -5, -10 cl-gel	MA-5 cl-gels
Calcein-AM and EthD-1 (Live/Dead® assay)	+	+

AlamarBlue™ assay	+	+
Consecutive AlamarBlue™ assay	+	-
WST-8 (Rotitest® Vital) assay	-	+
SEM Analysis of Cell-Laden <i>i</i> Csgel	+	-
Osmolarity [mOsm]	+	-

Metabolic activity as a measure of fluorescence emission of encapsulated L929 fibroblast cells and AlamarBlue™ reagent in *i*Csgel at day 1, 3 and 7

Cell-laden <i>i</i> Csgel	Day	Fluorescence emission a.u.					
		1	2	3	4	Mean	S.D.
oPMoMA-2 cross-linked	Day 1	43098	44505	44478	44319	44100.0	673.0
	Day 3	46162	47267	51462	49958	48712.3	2429.4
	Day 7	49384	50573	54735	59475	53541.8	4572.7
oPMoMA-4 cross-linked	Day 1	47122	43590	45226	44127	45016.3	1560.2
	Day 3	49506	45429	48529	51653	48779.3	2586.7
	Day 7	67535	61253	51476	61653	60479.3	6653.8
oPMoMA-5 cross-linked	Day 1	48976	48745	40904	40738	44840.8	4643.1
	Day 3	50702	51205	43447	49891	48811.3	3616.9
	Day 7	62724	64355	62971	64176	63556.5	828.1
oPMoMA-10 cross-linked	Day 1	43639	43065	41362	34535	40650.3	4189.9
	Day 3	53232	43486	45984	47160	47465.5	4138.3
	Day 7	50584	50860	62151	54826	54605.3	5390.9

Osmolarity of primary extracts (at day 1) and secondary extracts (at day 3) from cell-laden *i*Csgel as well as osmolarity of PBS, plain medium (DMEM-low glucose) and serum-containing medium (DMEM-low glucose 89% + FBS 10% + P/S 1%).

Cell-laden <i>i</i> Csgel	Day	Osmolarity [mOsm]					
		1	2	3	4	Mean	S.D.
PBS		287	289	286	287	287.3	1.3
DMEM-Ig	1	329	326	327	328	327.5	1.3
cond. DMEM-Ig		333	334	332	336	333.8	1.7
89%DMEM,10%FBS,1%P/S		420	400	420	407	411.8	9.9
oPMoMA-2 cross-linked	1	362	384	367	374	371.8	9.5
	3	401	393	383	398	393.8	7.9
oPMoMA-4 cross-linked	1	367	363	373	382	371.3	8.3
	3	389	401	382	392	391.0	7.9
oPMoMA-5 cross-linked	1	335	338	354	333	340.0	9.6
	3	387	379	365	376	376.8	9.1
oPMoMA-10 cross-linked	1	349	340	333	336	339.5	7.0
	3						

Consecutive AlamarBlue™ assay. Metabolic activity as a measure of fluorescence emission of encapsulated L929 fibroblast cells and AlamarBlue™ reagent in *i*Csgel at days 1, 2 and 3.

Cell-laden <i>i</i> Csgel	Day	Fluorescence emission a.u.					
		1	2	3	4	Mean	S.D.
oPMoMA-2 cross-linked	Day 1	38732	42392	41575	39681	40595.0	1921.2

	<b>Day 2</b>	38858	45450	42572	42530	42352.5	3304.8
	<b>Day 3</b>	41113	47586	41498	48893	44772.5	3631.2
<b>oPMoMA-4 cross-linked</b>	<b>Day 1</b>	40505	40194	39996	39662	40089.3	256.6
	<b>Day 2</b>	42849	41895	41928	41784	42114.0	541.5
	<b>Day 3</b>	42493	44882	45377	41988	43685.0	1542.2
<b>oPMoMA-5 cross-linked</b>	<b>Day 1</b>	39669	38697	39629	40844	39709.8	550.0
	<b>Day 2</b>	45014	39847	39606	42214	41670.3	3055.1
	<b>Day 3</b>	41481	48322	45630	42329	44440.5	3446.3
<b>oPMoMA-10 cross-linked</b>	<b>Day 1</b>	40756	38691	39027	38316	39197.5	1108.0
	<b>Day 2</b>	41800	37165	38265	37459	38672.3	2421.8
	<b>Day 3</b>	34761	31971	33492	32629	33213.3	1396.9

*Table S9:* Degradation characteristics of oPMoMA-x cross-linked iCsgel in comparison to physically gelled control in PBS at 37 °C.

	d0	d1	d7	d14	d21	d28	d35	d42
Physically gelled control	Stereomicroscopy, Remaining dry weight, Leachables [%], Medium pH and Swelling ratio				dissolved (n.d.) <sup>a</sup>			
oPMoMA-2 cl-gel	Stereomicroscopy, Remaining dry weight, Leachables [%], Medium pH and Swelling ratio				dissolved (n.d.)			
oPMoMA-4 cl-gel	Stereomicroscopy, Remaining dry weight, Leachables [%], Medium pH and Swelling ratio							
oPMoMA-5 cl-gel								
oPMoMA-10 cl-gel								

<sup>a</sup>n.d. (not determined): missing data due to complete degradation.

Remaining dry weight, leachables [%], swelling ratio and medium pH at selected degradation time points

iCsgel degradation	Days	Remaining dry weight [%]					
		1	2	3	4	Mean	S.D.
Physically gelled control		74.28	60.70	58.76	40.04	58.44	14.08
oPMoMA-2 cl-gel		117.69	98.38	95.73	86.09	99.47	13.24
oPMoMA-4 cl-gel	d7	100.21	110.60	77.35	110.70	99.71	15.70
oPMoMA-5 cl-gel		100.24	95.77	90.28	85.51	92.95	6.42
oPMoMA-10 cl-gel		78.93	101.29	82.24	93.66	89.03	10.33
Physically gelled control		20.47	6.07	79.32	55.72	40.40	33.29
oPMoMA-2 cl-gel		90.61	81.77	70.25	89.40	83.01	9.36
oPMoMA-4 cl-gel	d14	83.69	92.29	104.82	81.63	90.61	10.54
oPMoMA-5 cl-gel		94.04	90.11	76.62	83.81	86.14	7.62
oPMoMA-10 cl-gel		92.80	88.77	86.30	86.82	88.67	2.95
Physically gelled control	d21	0.00	0.00	0.00	0.00	0.00	0.00
oPMoMA-2 cl-gel		34.73	37.86	34.28	52.83	39.92	8.75

oPMoMA-4 cl-gel		90.04	102.61	95.03	109.74	99.36	8.64
oPMoMA-5 cl-gel		107.94	86.35	100.40	74.24	92.23	14.97
oPMoMA-10 cl-gel		80.10	83.39	82.46	90.27	84.06	4.37
Physically gelled control							
oPMoMA-2 cl-gel							
oPMoMA-4 cl-gel	d28	77.45	84.02	58.86	88.66	77.25	13.09
oPMoMA-5 cl-gel		81.57	71.96	94.92	73.00	80.36	10.62
oPMoMA-10 cl-gel		66.65	69.26	77.19	101.34	78.61	15.80
Physically gelled control							
oPMoMA-2 cl-gel							
oPMoMA-4 cl-gel	d35	72.12	67.43	67.28	73.09	69.98	3.05
oPMoMA-5 cl-gel		72.92	70.07	80.11	82.73	76.46	5.95
oPMoMA-10 cl-gel		76.33	84.38	84.44	74.94	80.02	5.10
Physically gelled control							
oPMoMA-2 cl-gel							
oPMoMA-4 cl-gel	d42	57.77	81.96	59.81	57.02	64.14	11.94
oPMoMA-5 cl-gel		43.95	74.51	76.40	86.29	70.29	18.30
oPMoMA-10 cl-gel		71.76	62.70	61.40	73.03	67.22	6.02

<i>i</i> Csgel degradation	Days	Leachables [%]					
		1	2	3	4	Mean	S.D.
Physically gelled control		31.65	37.11	11.43	23.16	25.84	11.19
oPMoMA-2 cl-gel		11.72	12.38	7.78	12.38	11.06	2.21
oPMoMA-4 cl-gel	d0	9.69	13.64	7.27	8.66	9.82	2.74
oPMoMA-5 cl-gel		9.87	8.25	7.67	12.04	9.46	1.96
oPMoMA-10 cl-gel		6.54	12.15	-2.49	7.04	5.81	6.08
Physically gelled control		49.23	61.83	47.96	69.23	57.06	10.25
oPMoMA-2 cl-gel		-3.90	13.80	11.72	24.56	11.55	11.73
oPMoMA-4 cl-gel	d7	9.49	4.50	28.28	-1.11	10.29	12.75
oPMoMA-5 cl-gel		9.66	12.13	16.64	24.78	15.80	6.65
oPMoMA-10 cl-gel		26.23	11.01	15.71	12.93	16.47	6.79
Physically gelled control		86.01	96.18	29.74	57.18	67.28	29.98
oPMoMA-2 cl-gel		20.01	28.35	35.22	21.66	26.31	6.95
oPMoMA-4 cl-gel	d14	24.41	20.30	2.81	25.44	18.24	10.53
oPMoMA-5 cl-gel		15.25	17.33	29.26	26.28	22.03	6.79
oPMoMA-10 cl-gel		13.28	22.01	11.55	19.30	16.53	4.93
Physically gelled control		69.34	66.83	68.39	53.71	64.57	7.31
oPMoMA-2 cl-gel		18.68	11.39	11.88	-0.23	10.43	7.85
oPMoMA-4 cl-gel	d21	2.71	20.77	7.30	34.70	16.37	14.42
oPMoMA-5 cl-gel		25.14	26.73	15.49	16.08	20.86	5.90
oPMoMA-10 cl-gel							
Physically gelled control							
oPMoMA-2 cl-gel							
oPMoMA-4 cl-gel	d28	30.05	27.45	45.43	19.02	30.49	11.02
oPMoMA-5 cl-gel		26.49	33.98	12.36	35.79	27.15	10.65
oPMoMA-10 cl-gel		37.71	39.15	20.89	5.80	25.89	15.75

Physically gelled control							
oPMoMA-2 cl-gel							
oPMoMA-4 cl-gel	d35	34.87	41.77	37.61	33.24	36.87	3.73
oPMoMA-5 cl-gel		34.28	35.71	26.03	27.23	30.81	4.89
oPMoMA-10 cl-gel		28.66	25.87	13.46	30.34	24.58	7.64
Physically gelled control							
oPMoMA-2 cl-gel							
oPMoMA-4 cl-gel	d42	47.82	29.23	44.54	47.92	42.38	8.91
oPMoMA-5 cl-gel		60.39	31.64	29.46	24.10	36.39	16.31
oPMoMA-10 cl-gel		32.93	44.92	37.07	32.11	36.76	5.86

<i>i</i> Csgel degradation	Days	Swelling ratio					
		1	2	3	4	Mean	S.D.
Physically gelled control		16.42	27.75	24.07	32.04	25.07	6.62
oPMoMA-2 cl-gel		10.34	13.87	10.95	10.41	11.39	1.67
oPMoMA-4 cl-gel	d0	11.98	10.98	10.55	10.92	11.11	0.61
oPMoMA-5 cl-gel		11.50	10.01	11.78	14.09	11.85	1.69
oPMoMA-10 cl-gel		10.26	8.84	9.35	8.20	9.16	0.87
Physically gelled control		11.58	40.94	23.75	37.23	28.38	13.41
oPMoMA-2 cl-gel		9.84	11.98	12.80	12.45	11.77	1.33
oPMoMA-4 cl-gel	d7	12.82	12.65	12.85	10.88	12.30	0.95
oPMoMA-5 cl-gel		10.82	13.17	13.16	17.25	13.60	2.67
oPMoMA-10 cl-gel		15.93	10.93	12.95	11.49	12.82	2.24
Physically gelled control		40.26	103.05	30.61	58.29	58.05	32.12
oPMoMA-2 cl-gel		12.02	13.85	12.86	13.21	12.98	0.76
oPMoMA-4 cl-gel	d14	13.42	12.74	13.47	14.89	13.63	0.90
oPMoMA-5 cl-gel		14.21	16.15	15.53	15.88	15.44	0.86
oPMoMA-10 cl-gel		13.91	16.24	13.02	15.30	14.62	1.43
Physically gelled control		24.20	20.12	22.90	29.51	24.18	3.94
oPMoMA-2 cl-gel		14.46	15.72	14.26	10.28	13.68	2.36
oPMoMA-4 cl-gel	d21	13.12	13.85	12.99	19.03	14.75	2.88
oPMoMA-5 cl-gel		17.15	15.88	15.70	14.96	15.92	0.91
oPMoMA-10 cl-gel							
Physically gelled control							
oPMoMA-2 cl-gel							
oPMoMA-4 cl-gel	d28	13.77	11.20	13.97	11.07	12.50	1.58
oPMoMA-5 cl-gel		13.60	15.48	15.47	19.66	16.05	2.56
oPMoMA-10 cl-gel		17.12	15.76	16.05	14.24	15.79	1.19
Physically gelled control							
oPMoMA-2 cl-gel							
oPMoMA-4 cl-gel	d35	12.25	19.01	8.73	14.81	13.70	4.33
oPMoMA-5 cl-gel		16.33	18.25	14.33	15.14	16.01	1.70
oPMoMA-10 cl-gel		13.16	15.53	12.06	14.03	13.70	1.47
Physically gelled control							
oPMoMA-2 cl-gel	d42						
oPMoMA-4 cl-gel		19.37	14.50	13.78	20.76	17.10	3.48

oPMoMA-5 cl-gel	22.49	14.13	14.27	14.69	16.39	4.07
oPMoMA-10 cl-gel	14.64	13.69	15.53	14.79	14.66	0.76

	Day	Medium pH				Mean	S.D.
		1	2	3	4		
Physically gelled control		7.36	7.30	7.33	7.32	7.33	0.03
oPMoMA-2 cl-gel		7.20	7.28	7.23	7.18	7.22	0.04
oPMoMA-4 cl-gel	d0	7.11	7.14	7.15	7.05	7.11	0.05
oPMoMA-5 cl-gel		6.98	7.13	7.11	7.14	7.09	0.07
oPMoMA-10 cl-gel		6.99	7.02	7.01	6.96	7.00	0.03
Physically gelled control		7.36	7.41	7.41	7.39	7.39	0.02
oPMoMA-2 cl-gel		7.24	7.28	7.23	7.30	7.26	0.03
oPMoMA-4 cl-gel	d7	7.19	7.25	7.15	7.27	7.22	0.06
oPMoMA-5 cl-gel		7.23	7.29	7.21	7.24	7.24	0.03
oPMoMA-10 cl-gel		7.14	7.23	7.19	7.34	7.23	0.09
Physically gelled control		7.31	7.39	7.36	7.42	7.37	0.05
oPMoMA-2 cl-gel		7.31	7.38	7.41	7.38	7.37	0.04
oPMoMA-4 cl-gel	d14	7.33	7.39	7.35	7.28	7.34	0.05
oPMoMA-5 cl-gel		7.39	7.41	7.39	7.40	7.40	0.01
oPMoMA-10 cl-gel		7.33	7.37	7.32	7.41	7.36	0.04
Physically gelled control				7.46	7.51	7.49	0.04
oPMoMA-2 cl-gel		7.42	7.44	7.39	7.37	7.41	0.03
oPMoMA-4 cl-gel	d21	7.49	7.42	7.43	7.48	7.46	0.04
oPMoMA-5 cl-gel		7.41	7.43	7.43	7.32	7.40	0.05
oPMoMA-10 cl-gel		7.49	7.39	7.41	7.38	7.42	0.05
Physically gelled control							
oPMoMA-2 cl-gel							
oPMoMA-4 cl-gel	d28	7.37	7.42	7.46	7.37	7.40	0.05
oPMoMA-5 cl-gel		7.43	7.41	7.37	7.42	7.41	0.03
oPMoMA-10 cl-gel		7.36	7.41	7.47	7.41	7.41	0.04
Physically gelled control							
oPMoMA-2 cl-gel							
oPMoMA-4 cl-gel	d35	7.38	7.39	7.43	7.43	7.41	0.03
oPMoMA-5 cl-gel		7.43	7.44	7.41	7.43	7.43	0.01
oPMoMA-10 cl-gel		7.38	7.42	7.38	7.41	7.40	0.02
Physically gelled control							
oPMoMA-2 cl-gel							
oPMoMA-4 cl-gel	d42	7.39	7.42	7.35	7.46	7.41	0.05
oPMoMA-5 cl-gel		7.43	7.38	7.36	7.46	7.41	0.05
oPMoMA-10 cl-gel		7.42	7.44	7.39	7.37	7.41	0.03