Tough, ultrastretchable and tear-resistant hydrogels enabled by linear macro-crosslinker

Zhenxing Cao,[‡]^a Yi Wang,[‡]^a Hao Wang,^a Changshu Ma,^a Heng Li,^a Jing Zheng,^a

Jinrong Wu*a and Guangsu Huang*a

^aState Key Laboratory of Polymer Materials Engineering, College of Polymer Science and

Engineering, Sichuan University, Chengdu 610065, P. R. China

‡ These authors contributed equally to this paper.

Corresponding author contact:

Prof. Jinrong Wu; Prof. Guangsu Huang

College of Polymer Science and Engineering

Sichuan University

Chengdu 610065, China

Email: <u>wujinrong@scu.edu.cn; guangsu-huang@hotmail.com;</u>

Phone: 86-28-85463433

Fax: 86-28-85463433

Supporting Tables & Figures

Table S1. Different yield values of CA macro-crosslinker.

Yield of CA	20.32%
Yield of AGE	13.99%

Table S2. The swelling properties of the MC hydrogels and the calculation of crosslinking density.

Sample	Hydrogel	Polymer	Swollen	Swelling	Crosslinking
	Weight	Weight	weight	ratio	density
	(g)	(g)	(g)	(Q)	(mol•m ⁻³)
MC _{0.5-30}	1.81	0.44	62.46	139.97	0.48
MC _{1.0-30}	2.28	0.56	53.31	94.35	0.93
MC _{2.0-30}	1.84	0.45	23.70	51.67	2.63
MC _{3.0-30}	1.83	0.45	20.69	45.18	3.32

Sample	Hydrogel	Polymer	Swollen	Swelling	Crosslinkin
	Weight	Weight	weight	ratio	g
	(g)	(g)	(g)	(Q)	density
					(mol•m ⁻³)
H-MC	2.11	0.53	67.98	128.14	0.57
M-MC	1.95	0.49	61.12	124.53	0.58
L-MC	2.13	0.53	67.50	125.48	0.58

Table S3. The swelling properties of the H-MC, M-MC and L-MC.



Fig. S1. FT-IR spectra of CS powder and CA macro-crosslinker.



Fig. S2. ¹H NMR spectra for CA macro-crosslinker reaction solution during different reaction

stages. CS and AGE were dissolved in CD_3COOD/D_2O and then reacted, we took samples at different reaction time and cooled them rapidly to stop the reaction. Because the addition of AGE is much larger than that of CS, the peak of H on CS is not obvious, and it is covered by AGE.



Fig. S3. Young's modulus of MC_{2.0-y} hydrogels.



Fig. S4. Photos of the MC hydrogel before and after compression, respectively. The black fonts represent the compressive strain, and the red fonts represent the strain recovery ratio.



Fig. S5. Locally enlarged stress–strain curves of MC_{x-30} hydrogels.



Fig. S6. Yield stress and neck elongation of MC_{x-30} hydrogels.



Fig. S7. Typical stress–strain curves of $MC_{1.0-30}$ and $MC_{6.0-30}$ hydrogels.



Fig. S8. (a) Ensemble-averaged correlation function $\langle g_T^{(2)}(\tau)-1 \rangle$ and (b) relaxation-time distribution function $G(\Gamma)$ for the SC hydrogel.