Supporting Information

Mechanically robust and stretchable silk/hyaluronic acid hydrogels

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Table S1. Tensile mechanical properties of the hydrogels. Standard deviations are shown in the parentheses.

DM %	MeHA w/v%	SF w/v%	σ_f / kPa	ε_f %	W / kJ m^{-3}
4	1	2.5	65 (5)	4.4 (0.5)	170 (40)
14	1	2.5	57 (2)	2.9 (0.3)	83 (16)
25	1	2.5	52 (2)	2.1 (0.1)	38 (8)
4	1	0	3 (0.5)	5.4 (0.5)	9 (2)
14	1	0	4 (0.6)	3.9 (0.1)	8 (0.8)
25	1	0	6 (0.7)	3.2 (0.3)	8 (2)

Table S2. Toughness (W) of the hydrogels. Standard deviations are shown in the parentheses.

DM %	MeHA w/v%	SF w/v%	W / kJ m^{-3}	
			As-prepared state	After equilibrium swelling in water
4	1	2.5	320 (6)	42 (2)
14	1	2.5	250 (30)	37 (1)
25	1	2.5	200 (30)	40 (3)
4	2	2.5	166 (17)	26 (6)
14	2	2.5	97 (20)	25 (4)
25	2	2.5	76 (10)	15 (1)
4	1	0	19 (2)	6
14	1	0	17 (2)	4
25	1	0	20 (4)	3 (1)
4	2	0	43 (4)	3
14	2	0	32 (5)	7
25	2	0	24 (1)	6

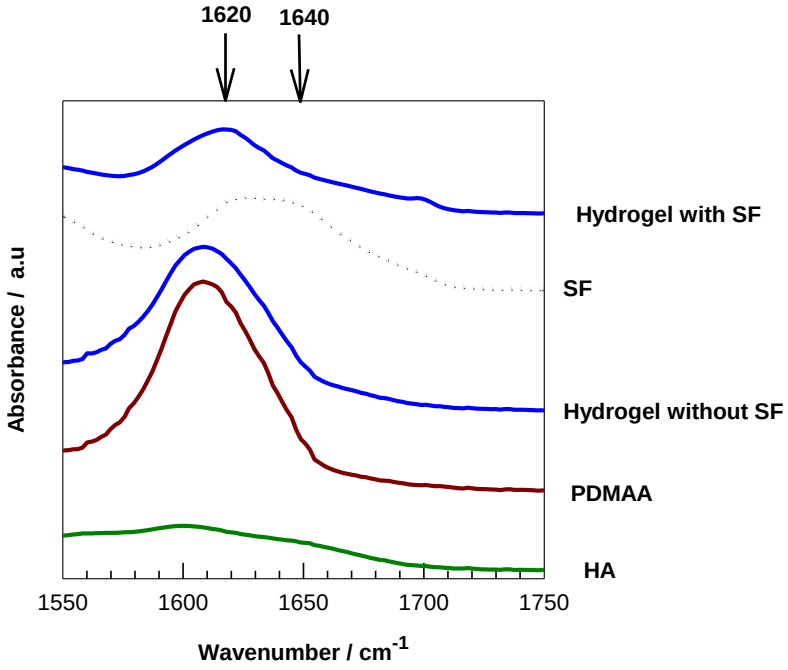

Figure S1: Amide-I region of FTIR spectra of freeze-dried SF solution (dotted curve), HA, PDMAA, and hydrogels with and without SF.

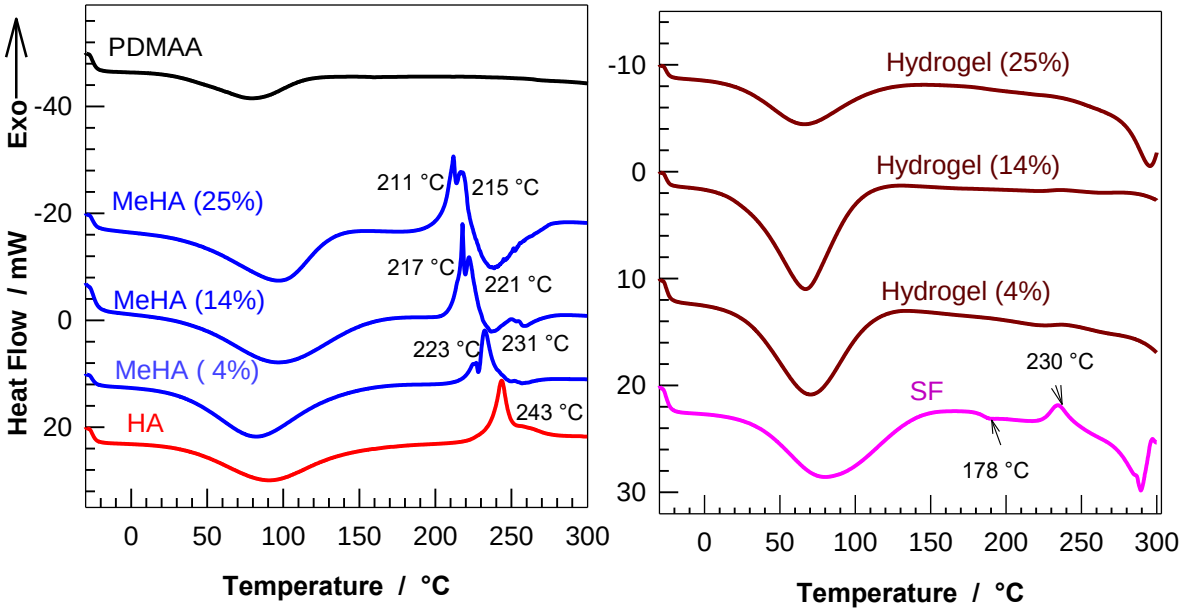

Figure S2: DSC scans of freeze-dried gel components and composite hydrogels. The numbers in parentheses are the methacrylation degrees of MeHA.

All samples show broad endothermic peaks at around 100 °C due to the loss of moisture remaining after freeze-drying. SF shows typical peaks at 178 and 230 °C corresponding to the glass transition and

non-isothermal crystallization, respectively, followed by the appearance of an endothermic peak at around 300 °C due to degradation. HA shows a sharp exothermic peak at 243 °C due to thermal degradation, whereas MeHA macromers show two exothermic peaks at 211-223 and 215-243 °C representing conversion of HA into a less-ordered state and thermal degradation, respectively. In composite hydrogels, these characteristic peaks do not appear and they all start to degrade at around 300 °C.

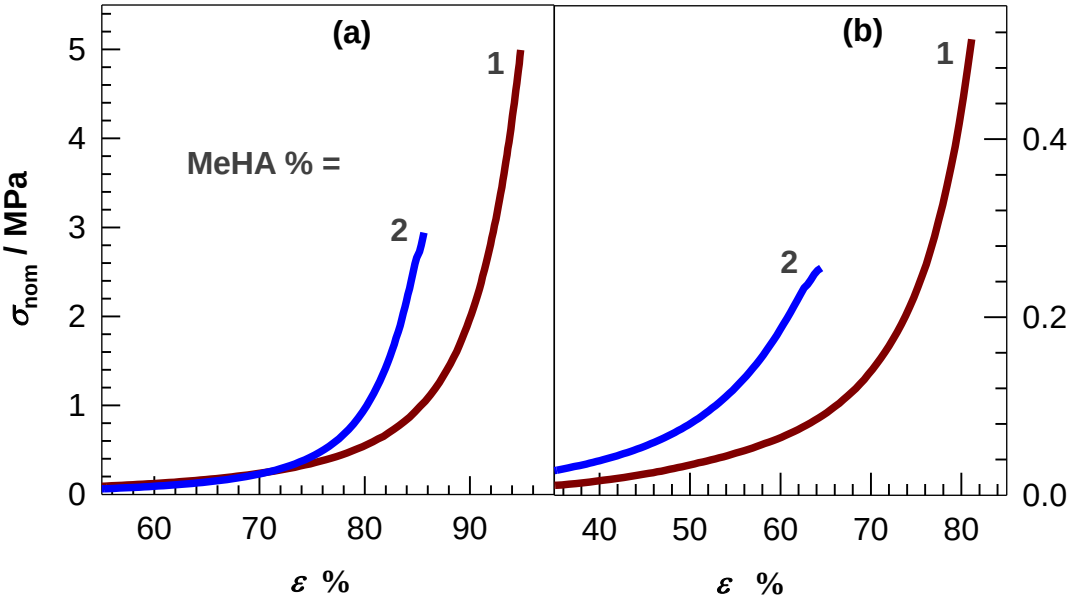

Figure S3. Compressive stress-strain curves of the hydrogels with 1 and 2 w/v % MeHA in as-prepared (a) and water-swollen states (b). DM = 4%. SF = 2.5 w/v %.