Supporting Information

Ultra-Stretchable and Self-Healing Anti-Freezing Strain Sensors based on Hydrophobic Associated Polyacrylic Acid Hydrogels

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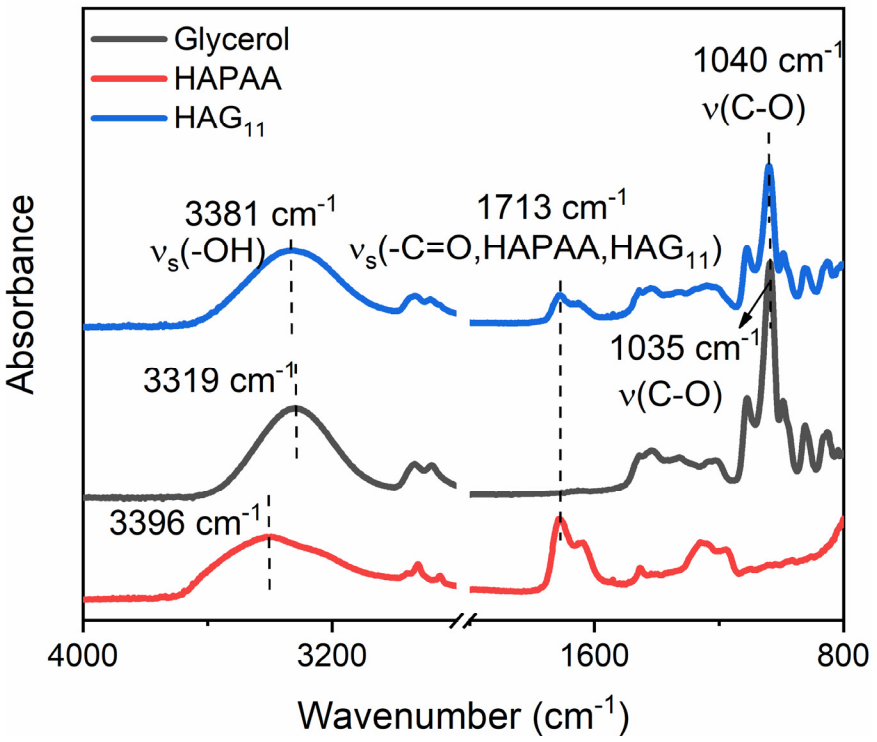

Figure S1. ATR-FTIR spectra of glycerol, HAPAA, and HAG_{11} hydrogels.

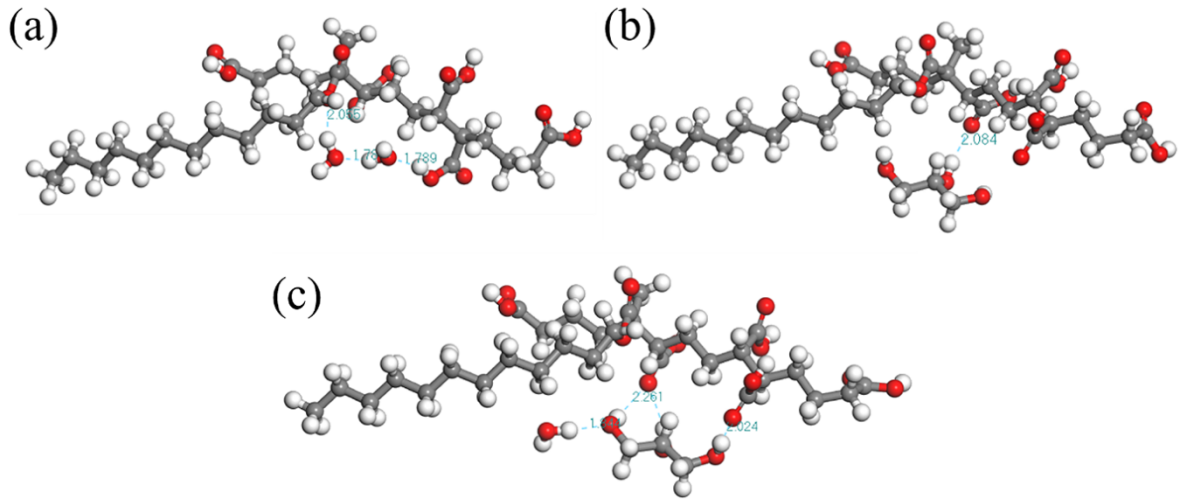

Figure S2. DFT-optimized structure of glycerol, H_2O, and HAPAA. The interaction models of **(a)** H_2O-HAPAA, **(b)** glycerol-HAPAA, and **(c)** H_2O-glycerol-HAPAA.

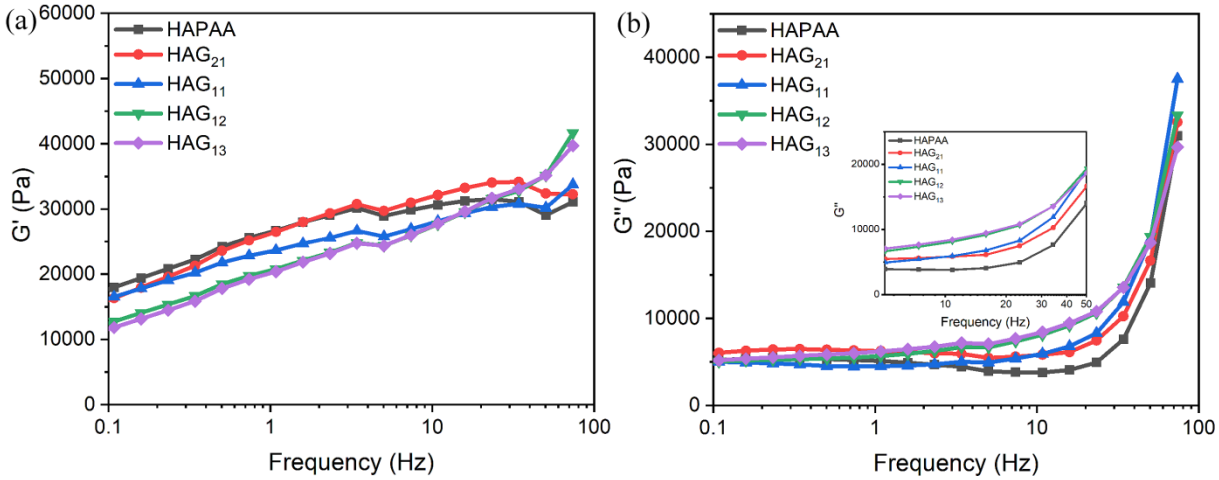

Figure S3. (a) Storage modulus (G'), and **(b)** the loss modulus (G'') of the HAPAA and HAG_x hydrogels.

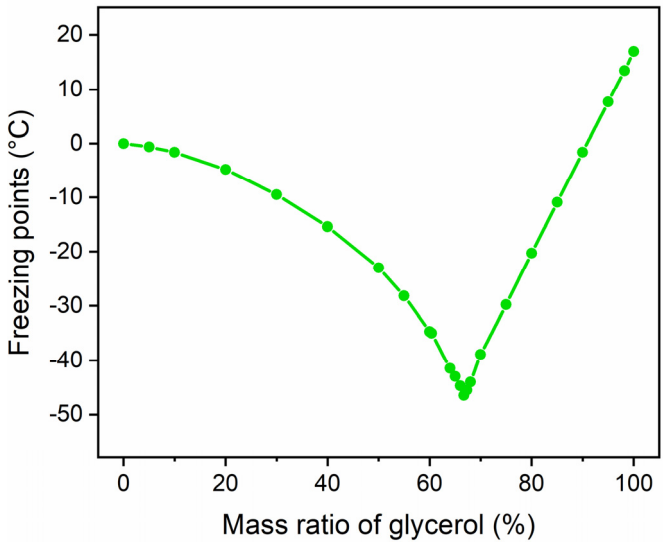

Figure S4. Relationship between the freezing points and the glycerol mass ratio in the glycerol-water mixed solutions.[1]

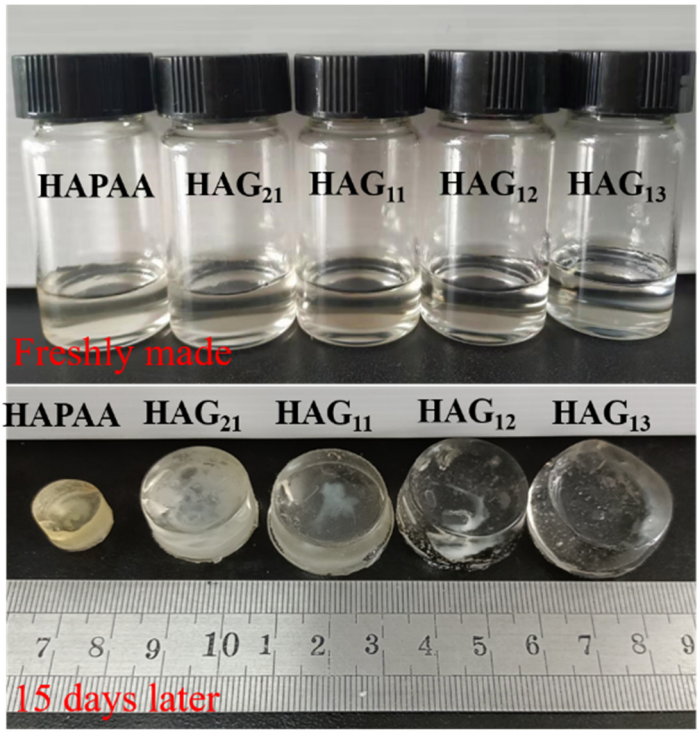

Figure S5. Comparison of the freshly prepared HAPAA and HAGx hydrogels (top) and after a storage time of 15 days (bottom).

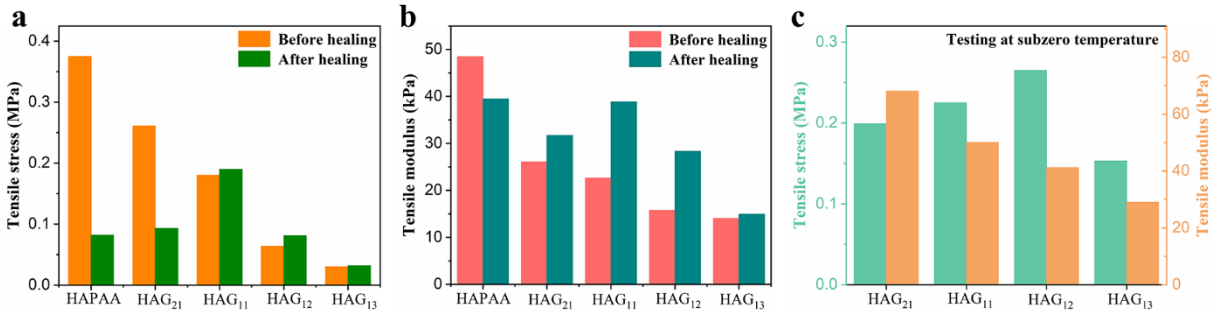

Figure S6. (a-b) The tensile stress and (b) modulus of HAPAA and HAG$_x$ hydrogels before and after self-healing (RT, 24h). **(c)** The tensile stress and modulus of HAPAA and HAG$_x$ hydrogels at subzero temperature.

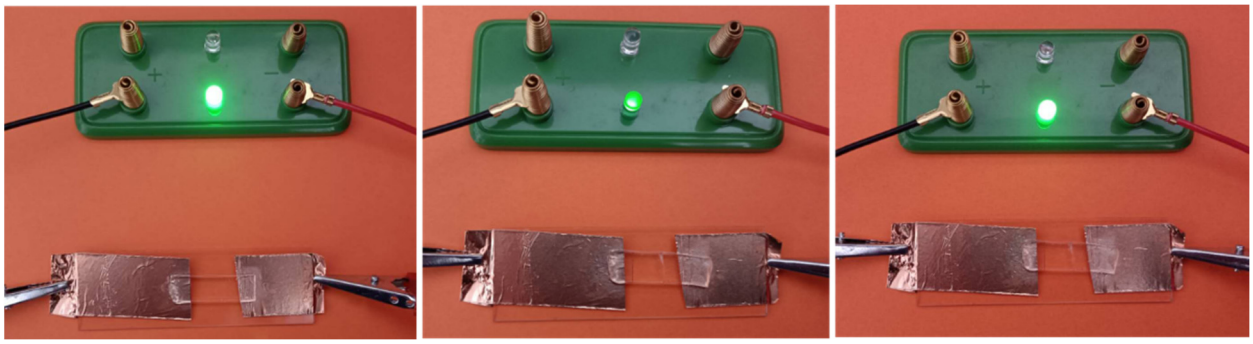

Figure S7. Circuits comprised of the HAG$_{11}$ hydrogel and a green LED light: **(a)** original, **(b)** cutting off and being contacted immediately, **(c)** self-healing for 6 h.

Table S1. Compositions of the HAPAA hydrogel and HAG_x hydrogels

Samples	AA(g)	LMA(g)	CTAB(g)	H_2O(g)	Glycerol(g)	APS(mg)	H_2O/Glycerol (wt/wt)
HAPAA	2.5	0.2	0.3	7.00	0	1	--
HAG_{21}	2.5	0.2	0.3	4.67	2.33	1	2:1
HAG_{11}	2.5	0.2	0.3	3.50	3.50	1	1:1
HAG_{12}	2.5	0.2	0.3	2.33	4.67	1	1:2
HAG_{13}	2.5	0.2	0.3	1.75	5.25	1	1:3

Table S2. The comparison of the HAG_{11} hydrogel with other antifreeze stretchable hydrogels in mechanical strength

Materials	Antifreeze temperature (℃)	Stretchability (%)	Tensile Strength (MPa)	Self-healing	Refs.
PVA (EG)	-40	1000	1.5	Yes (freeze thawing)	[2]
PAMPS/PAAm (EG/LiCl)	-80	225	0.57	No	[3]
PAAm ($CaCl_2$)	-54	450	0.12	No	[4]
PAAm/PVA (EG)	-40	--	--	No	[5]
PAAm/casein (LiCl)	-20	1450	0.16	No	[6]
PVA-TA@talc (EG)	-30	700	0.6	No	[7]
PVA-PAA (EG)	-25	550	0.025	No	[8]
PAA-PANI (Glycerol)	-26	1000	0.035	Yes	[9]
PAA-PAAm (Glycerol)	-20	600	0.075	No	[10]
HAG_{11} (Glycerol)	-70	4000	0.18	Yes	This work

Table S3. Interaction energies in H_2O-HAPAA, glycerol-HAPAA, and glycerol-H_2O-HAPAA by DFT calculations.

Interaction pair	Interaction energy (Ha)
H_2O-HAPAA	-0.02013
Glycerol-HAPAA	-0.00994
Glycerol-H_2O-HAPAA	-0.02651

References

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