Supporting Information

for

Nature-inspired semi-IPN hydrogels with tunable mechanical property and multi-responsiveness

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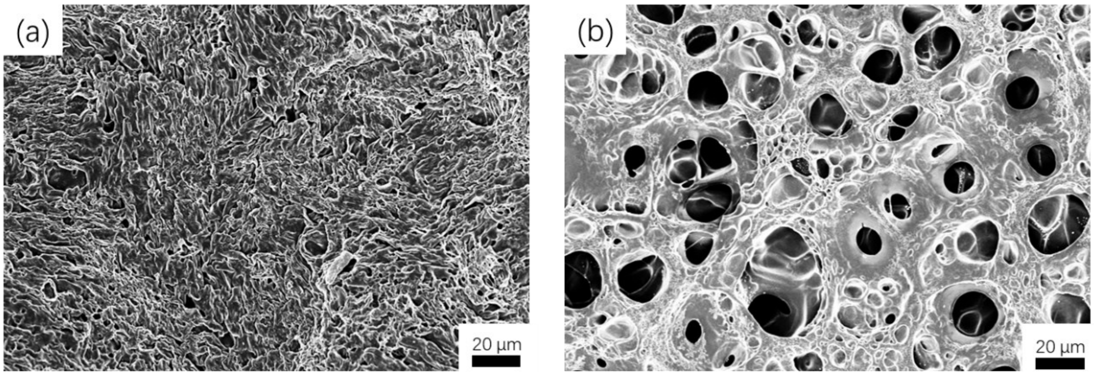

Fig. S1: SEM image of (a) PVA and (b) PAP hydrogels (scale bar = 20 μm).

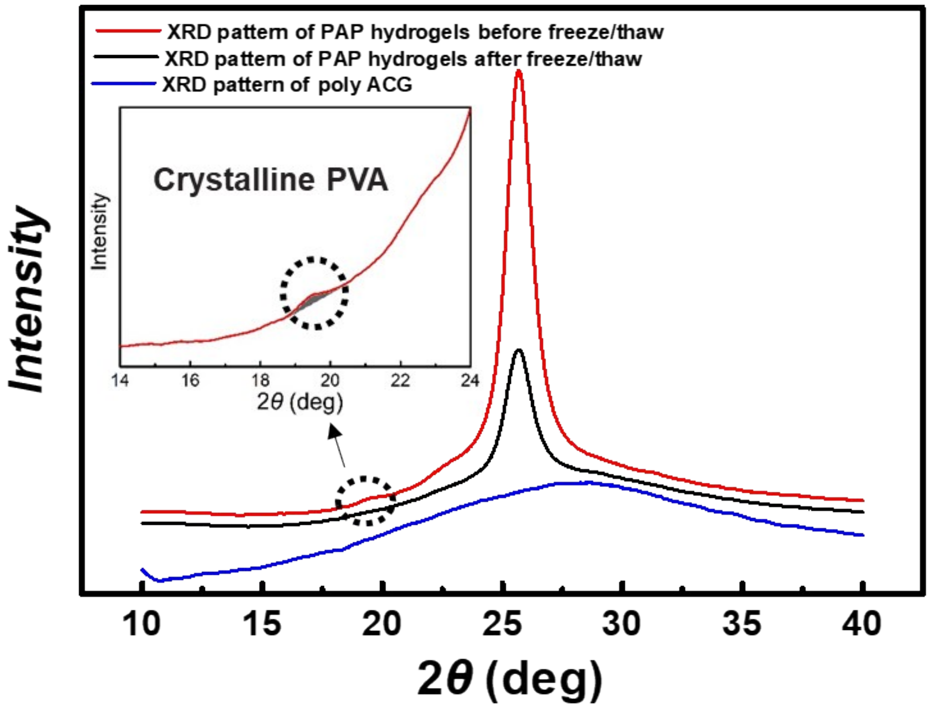

Fig. S2*:* X-ray powder diffraction profiles of poly ACG, as-prepared PAP hydrogels before and after freeze/thaw treatment.

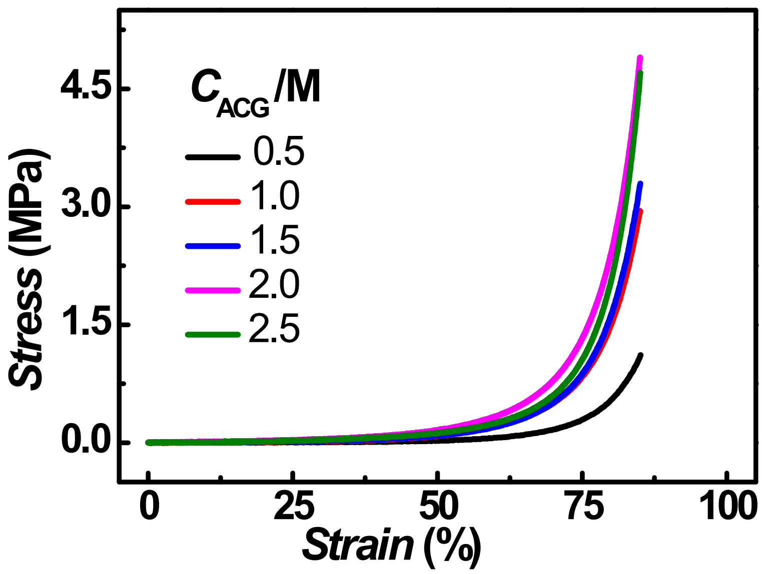

Fig. S3: Compression stress-strain curves of the PAP-*5.0*- C_{ACG} hydrogels with different ACG content, C_{ACG}.

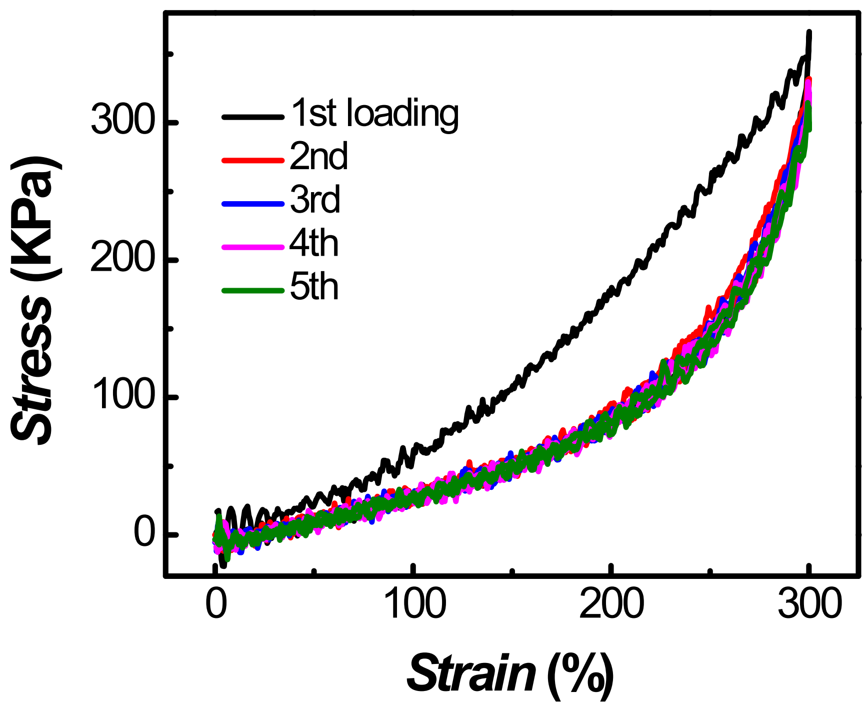

Fig. S4: Cyclic elongation stress-strain curves of PAP-*5.0-2.0* hydrogels.

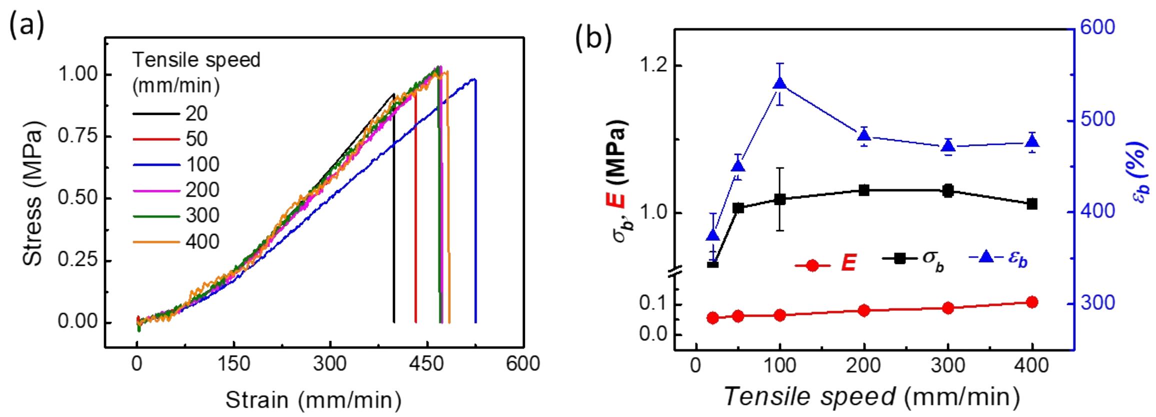

Fig. S5: (a, b) Tensile stress-strain curves (a) and corresponding mechanical properties (b) of the PAP-*5.0-2.0* hydrogels at different tensile speeds at 20 ℃.

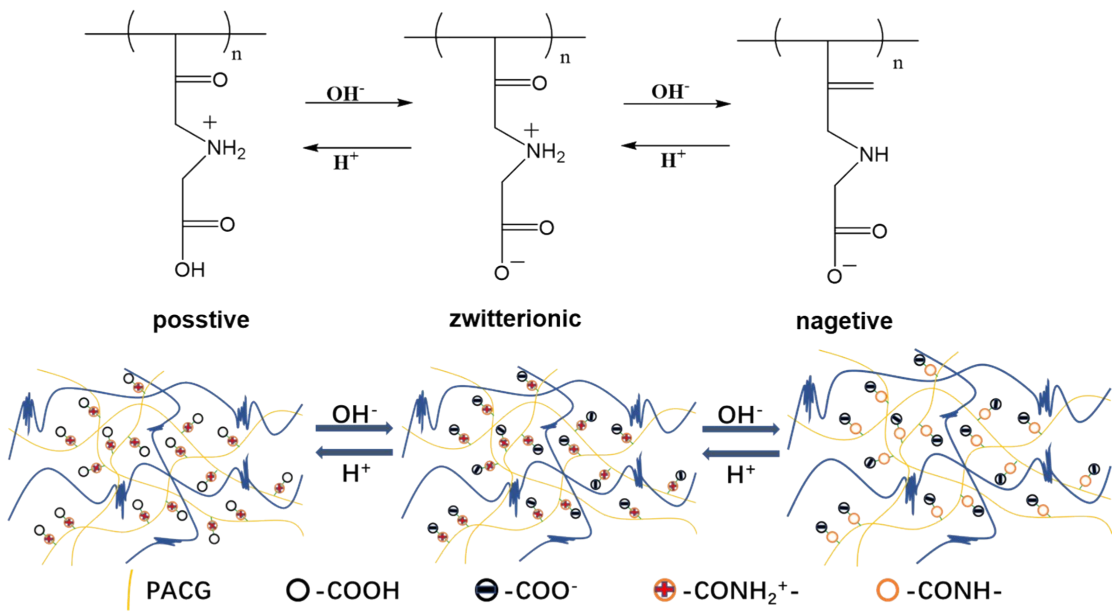

Fig. S6: Schematic structures of PAP hydrogels in solutions of different pH value.

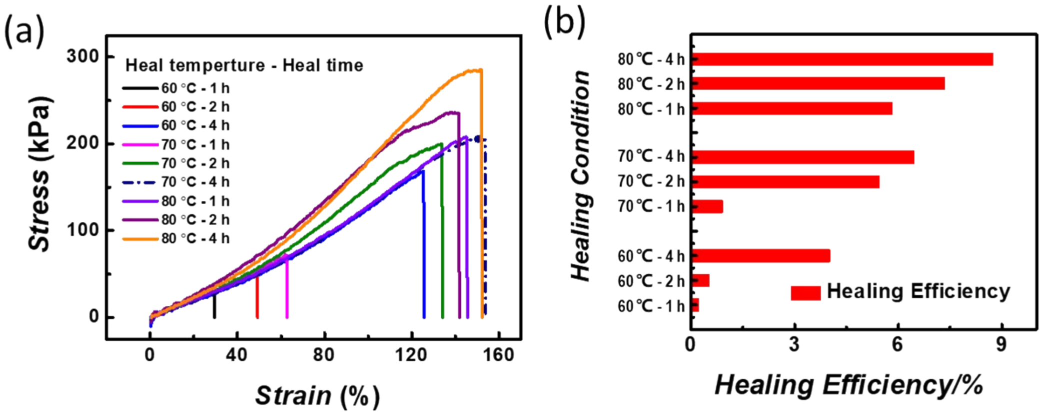

Fig. S7: (a, b) PAP-*5.0-2.0* hydrogels tensile stress-strain curves (a) and healing efficiency (b) after healed under different healing conditions.

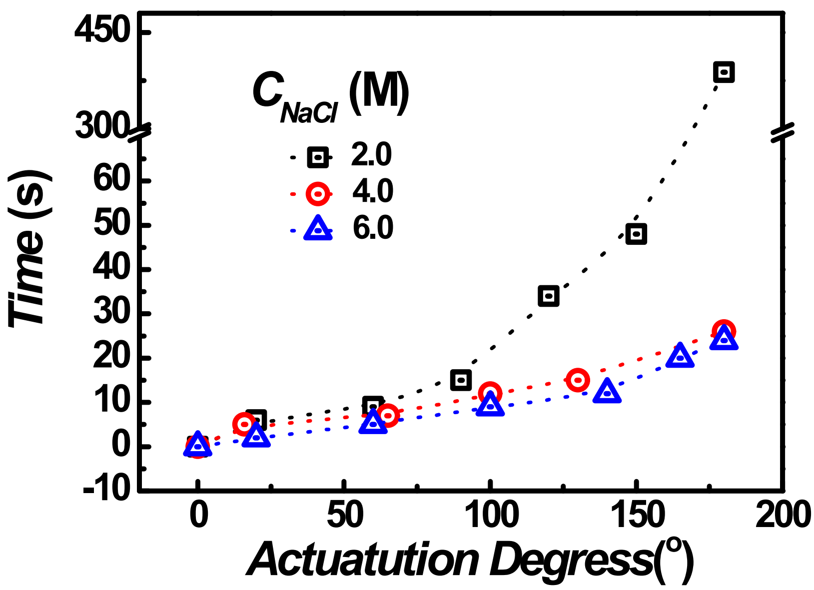

Fig. S8: Actuation degree of the bilayer hydrogels in NaCl solution with different concentrations.

The influence of NaCl concentration on self-rolling speed has been investigated (Fig. S8). The bilayer hydrogels was nonresponsive to the aqueous solution with C_{NaCl} less than 2.0 M. When C_{NaCl} increases from 2.0 M to 6.0 M, the rolling speed increases with the increase of salt concentration generally. The rolling speed at 4.0 M salt solution is comparable with that of 6.0 M salt solution, which is consistent with the size variations in **Fig.6b**. This self-rolling behavior is mainly due to osmotic pressure. As the salt concentration increases, the salting-out effect caused by osmotic pressure acted as the driving force and phase separation occurred, leading to the volume shrinking of the PAP layer, then the bilayer hydrogel bend to the PAP layer side [1, 2].

Tab. S1 Gel fraction of PAP hydrogels.

Type	C_{PVA}/%	C_{ACG}/M	*Designed* W_{gf}/%	W_{gf}/%
PAP-5.0-0	5.0	0	5.0	4.3
PAP-5.0-0.5	5.0	0.5	11.8	11.2
PAP-5.0-1.0	5.0	1.0	16.8	16.1
PAP-5.0-1.5	5.0	1.0	21.2	20.5
PAP-5.0-2.0	5.0	2.0	25.2	24.3
PAP-5.0-2.5	5.0	2.5	29.4	28.5
PAP-2.5-2.0	2.5	2.0	23.3	22.6
PAP-7.5-2.0	7.5	2.0	27.2	26.3

The gel fraction of as-prepared hydrogels was determined by the weight change upon drying using an oven with the following steps. First, the weight of as-prepared hydrogel was recorded as W_{ap} which was measured by an electronic balance. Then the sample was transferred to the oven (110 °C) and heated to a constant weight. The weight of the dried hydrogel was denoted as W_d. The gel fraction W_{gf} is defined as the ratio in percentage of the weight of the dried hydrogel to the total weight of the as-prepared hydrogel as shown in Equation S1.

$$W_{gf}=(W_d/W_{ap})\times 100 \qquad \text{Equation S1}$$

Tab. S2 Comparisons of mechanical properties of hydrogels

Composition	σ_b (kPa)	ε_b (%)	E (kPa)	W_b (kJ/m^3)	Reference
PAM/PACG	1.38×10^3	530	106	308	This work
PVA/SA/Gly	1.25×10^3	516	340	/	(Oudong Hu et al., 2020)
PVA/ARC	35.3	/	/	/	(Lei Ding et al., 2020)
PVA/GSP	220	254	35	227.2	(Chunhui Luo et al., 2020)
PAM/PVA	270	500	/	/	(Zhang et al., 2016)
PVA/PSBMA	596	275	/	/	(Zhenwu Wang., 2016)
PVA-Borax	123.26	528	/	/	(Shengping Dai et al., 2019)
PVA/TTSBI	1.23×10^3	774	1.60×10^3	/	(Xiao Zhang et al., 2019)
TA-PVA/BSA	~3×10^3	410	/	/	(Rongnian Xu et al., 2018)
PVA/AAm/AAc	1.2×10^3	500	/	1.2×10^3	(Zhengyu Gong et al., 2016)
PVA/PAA/Fe^{3+}	1.0×10^3	1370	/	6.37×10^3	(Yongzhi Liang et al., 2020)
PVA/Glycerol/Polyaniline	477	472	/	/	(Chengxin Hu et al., 2018)
PVA/NaCl/glycerol	570	575	/	/	(Shuijiao Peng et al., 2018)
PVA/TA/CNC	~100	~900	/	/	(Changyou Shao et al., 2019)
PVA/NaCl	1.6×10^3	631	/	/	(Yujun Sun et al., 2017)
PVA/PA/NH_2-POSS	361	363	/	/	(Liang Shao et al., 2020)
PAM/PVA	183.8	488	87	/	(Gang Ge et al., 2018)
PVA/cellulose	37.3	747	/	/	(Yang Wang et al., 2020)

Note: '/' indicates 'not shown' in the references.

Movie S1: Reversible self-rolling deformation of the bilayer hydrogel strip in saline solution and DI water-3X SPEED

References

1. X. Jv, X. Zhao, H. Ge, J. Sun, H. Li, Q. Wang and H. Lu, *Journal of Chemical & Engineering Data*, 2019, **64**, 1228-1236.
2. S. Zheng, Y. Tian, X. Zhang, M. Du, Y. Song, Z. Wu and Q. Zheng, *Soft Matter*, 2018, **14**, 5888-5897.