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**Supporting Information** 

## Amylopectin-enabled skin-mounted hydrogel wearable sensor

Lingshu Kong, Zijian Gao, Xinyao Li, Guanghui Gao\*

Polymeric and Soft Materials Laboratory, School of Chemical Engineering and Advanced Institute of Materials Science, Changchun University of Technology, Changchun 130012, China Corresponding authors: Guanghui Gao

E-mail: ghgao@ccut.edu.cn

Sample name	AAm	AAc	Amy	MBA	Borax	NaOH	NaCl	KPS	H <sub>2</sub> O
	(g)	(g)	(g)	(µL)	(g)	(mol)	(g)	(g)	(mL)
Amy-PAAm-PAAc	2	7	1	100	0.05	0.06	0.5	0.02	30
m(Amy):m(AAm)=0:4	2	7	0	100	0.05	0.06	0.5	0.02	30
m(Amy):m(AAm)=1:4	2	7	0.5	100	0.05	0.06	0.5	0.02	30
m(Amy):m(AAm)=2:4	2	7	1	100	0.05	0.06	0.5	0.02	30
m(Amy):m(AAm)=3:4	2	7	1.5	100	0.05	0.06	0.5	0.02	30
m(Amy):m(AAm)=4:4	2	7	2	100	0.05	0.06	0.5	0.02	30
m(AAc):m(AAm)=5:2	2	5	1	100	0.05	0.06	0.5	0.02	30
m(AAc):m(AAm)=6:2	2	6	1	100	0.05	0.06	0.5	0.02	30
m(AAc):m(AAm)=7:2	2	7	1	100	0.05	0.06	0.5	0.02	30
m(AAc):m(AAm)=8:2	2	8	1	100	0.05	0.06	0.5	0.02	30
M(MBA)/M(AAm)=0 mol%	2	7	1	0	0.05	0.06	0.5	0.02	30
M(MBA)/M(AAm)=0.023 mol%	2	7	1	50	0.05	0.06	0.5	0.02	30
M(MBA)/M(AAm)=0.046 mol%	2	7	1	100	0.05	0.06	0.5	0.02	30
M(MBA)/M(AAm)=0.069 mol%	2	7	1	150	0.05	0.06	0.5	0.02	30
M(Borax)/M(AAm)=0 mol%	2	7	1	100	0	0.06	0.5	0.02	30
M(Borax)/M(AAm)=0.466 mol%	2	7	1	100	0 05	0.06	0.5	0.02	30
M(Borax)/M(AAm)=0.932  mol%	2	, 7	1	100	0.00	0.06	0.5	0.02	30
M(Borax)/M(AAm)=1.398 mol%	2	, 7	1	100	0.15	0.06	0.5	0.02	30

Table S1 Recipes for all hydrogel samples.



**Figure S1** Peeling strength curve of the hydrogel under the five times repeated peeling tests on Al substrate surfaces.



**Figure S2** Peeling strength curve of the hydrogel under the five times repeated peeling tests on Ti substrate surfaces.



**Figure S3** Peeling strength curve of the hydrogel under the five times repeated peeling tests on PMMA substrate surfaces.



**Figure S4** Peeling strength curve of the hydrogel under the five times repeated peeling tests on rubber substrate surfaces.



**Figure S5** Peeling strength curve of the hydrogel under the five times repeated peeling tests on pigskin substrate surfaces.



**Figure S6** Peeling strength curve of the hydrogel under the five times repeated peeling tests on glass substrate surfaces.



**Figure S7** Tensile stress-strain curves of hydrogel for different contents of Borax.



Figure S8 Corresponding elastic modulus and toughness from Figure S7.

As shown in Figure S1, the increase of borax cross-linking agent would increase the boron ester bond dynamic cross-linking point in the Amy network, which would enhance the mechanical properties of the hydrogel. And it could be seen that the toughness and elastic modulus of the hydrogel increased with the borax content was increased shown in Figure S2.



**Figure S9** Tensile stress-strain curves of hydrogel for different contents of MBA.



**Figure S10** Corresponding elastic modulus and toughness from Figure S9. As shown in Figure S3, the increase of MBA cross-linking agent would increase the cross-linking point in the hydrogel network, which would enhance the mechanical properties. However, the too much MBA would make the hydrogel brittle and the elongation at break decreased.

Therefore, considering with the adhesive data and individual tensile propertie, the Amy-P(AAm/AAc) hydrogel with a molar ratio of Borax to AAm of 0.466 mol% and a molar ratio of MBA to AAm of 0.046 mol% was determined as the optimal recipe for the entire experimental exploration.



**Figure S11** Storage modulus (G') and loss modulus (G") in frequency sweep ( $\omega$ =10 rad/s) curves of Amy/P(AAm-AAc) hydrogel with the different crosslinking agent content.



**Figure S12** Storage modulus (G') and loss modulus (G") in strain ( $\gamma$ =0.5 %) sweep curves of Amy/P(AAm-AAc) hydrogel with the different crosslinking agent content.



**Figure S13** SEM images of the Amy/P(AAm-AAc) hydrogels. (a) The Amy/P(AAm-AAc) hydrogel using borax crosslinking agent alone; (b) The Amy/P(AAm-AAc) hydrogel using MBA crosslinking agent alone; (c) he Amy/P(AAm-AAc) hydrogel using both borax and MBA crosslinking agent. The images showed the hydrogels have a porous

structure due to the loss of water in the process of freeze-drying. And, the significant difference of the hole size was observed. Compared with the hydrogel using only one crosslinking agent, the hydrogel using both borax and MBA crosslinking agents at the same time exhibited an obviously denser network structure, and the hole size dramatically decreased, indicating the enhancement of mechanical properties.



**Figure S14** Electrical conductivity of the hydrogel with different AAc content. The electrical conductivity of the hydrogels was decreased with the increase of AAc content, proving the introduction of more AAc would result in a denser hydrogel network and restrict the movement of free ions.



**Figure S15** Comparison of amylopectin solution and casein solution placed at room temperature. (a) The first day; (b) After being left for four days; (c) After being left for two weeks. We put the casein solution that could be used to prepare the hydrogel (Polym. Chem. 2018, 9, 2617-2624) and the amylopectin solution in this experiment in the same environment at room temperature. As shown in figure S15, the casein solution was obviously deteriorated after being left for four days, the specific manifestations of the solution were the color had been darker, the mixture had been stratified and turbid, and the smell had become fishy. Furthermore, after being left for two weeks, the casein solution had more obvious layering and deterioration phenomenon. In contrast, the amylopectin solution could be stored stably for more than two weeks.