1 Supporting information

2 Tough, Adhesive, Self-healable, and Antibacterial Plant-inspired

3 Hydrogel Based on Pyrogallol-Borax Dynamic Cross-linking

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- 13 The supporting information includes 4 pages and 3 Figures as well as 214 Tables.
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16 This supplement contains:

- 17 Figure S1. Macroscopic photographs of PG/Borax solutions with different molar ratios
- 18 and structures speculated of PG-borax complexes.
- 19 Figure S2. Rheological properties of hydrogels.
- 20 Figure S3. Swelling behaviors and microstructure of hydrogels.
- 21 **Table S1.** The compositions of PG/borax aqueous solution.
- 22 Table S2. The compositions of various hydrogels.



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The PG solution without addition of borax (1, Table S1) is relatively steady under acidic conditions. Visual inspection of color evolution of PG solution with addition of borax confirms its oxidization to quinone in a basic pH (2-5, Table S1). Moreover, PG/borax solution developed a light brown color with increasing concentration of borax, which is indicative of the formation of PG/borax complex and indicating that borax has inhibition effect on PG oxidation.





Figure 52. Recological properties of hydrogers.



1 behavior, with the storage modulus (G') >> loss modulus (G") over the entire frequency
2 range. With the increase of molar ratio of PG and borax, both the G' and G" of the
3 copolymers were enhanced obviously, indicating a higher cross-linking density and
4 stronger cohesion.



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Figure S3. (a) Swelling photographs of PAM hydrogel, PAM-SPI hydrogel, and PAMSPI-P/B hydrogel. (b) Swelling curves of the hydrogels. (c)-(h) The SEM images of
PAM hydrogel, PAM-SPI hydrogel, PAM-SPI-P/B (2:1) hydrogel, PAM-SPI-P/B (1:1)
hydrogel, PAM-SPI-P/B (1:2) hydrogel, and PAM-SPI-P/B (1:4) hydrogel.

In the Figure S3, the swelling ratio of PAM-SPI-P/B hydrogels decreased as the feeding ratio of PG and borax increased because of the subsequent increase in crosslinking degree. The incorporation of PG and borax had a significant effect on the microstructure of hydrogels lead to a lower swelling rate.

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PG/Borax	PG	Borax	molar ratio (PG/Borax)		
Solution	(mg/ml)	(mg/ml)			
1	1.8	0	0		
2	1.8	2.9	2		
3	1.8	5.8	1		
4	1.8	11.6	0.5		
5	1.8	23.2	0.25		

Table S1. The compositions of PG/borax aqueous solution

Table S2. The compositions of various hydrogels

Hydrogels	PG	Borax	SPI	AM	APS	BIS	TEMED	H_2O
	(mg)	(mg)	(g)	(g)	(g)	(g)	(µl)	(wt.%)
PAM	0	0	0	2.5	0.25	0.025	20	80
PAM-SPI	0	0	0.25	2.5	0.25	0.025	20	80
PAM-SPI- P/B(2:1)	18	29	0.25	2.5	0.25	0.025	20	80
PAM-SPI- P/B(1:1)	18	58	0.25	2.5	0.25	0.025	20	80
PAM-SPI- P/B(1:2)	18	116	0.25	2.5	0.25	0.025	20	80
PAM-SPI- P/B(1:4)	18	232	0.25	2.5	0.25	0.025	20	80