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## Supporting informaiton

## A transparent, tough self-healing hydrogel based on dual physically

and chemically triple crosslinked network

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Fig. S1 The transparent film of PVA/B TN hydrogel.



Fig. S2 Temperature dependence of modulus and tan delta for different freeze-thaw cycles hydrogels. (a) 0 cycle, (b) 1 cycle, (c) 2 cycles (d) 3 cycles. All hydrogels were freeze-dried for 24h to the DMA measurement.



Fig. S3 The glass-transition temperature (Tg) of different freeze-thaw cycles hydrogels



Fig. S4 The tensile strain-stress test was performed on the hydrogel samples.



Fig. S5 Tensile stress–strain curves for different PVA mass concentration without freezing/thawing cycles. The "x"in the figure represents unfracture, which was difficult to accurately measure the magnitude of the stress and strain at low mass concentrations (10wt% and 15wt%).



Fig. S6 The PVA/B TN hydrogel cyclic tensile loading-unloading curves for five successive stretching to 400% strain.



Fig. S7 Cyclic tensile loading-unloading curves at 400% strain with different resting times.



Fig. S8 Tensile stress-strain curves of hydrogels with 20wt% PVA mass concentration after three freeze-thaw cycles at different healing times.



Fig S9. G' and G" for the original and healed 3 freeze-thaw cycles hydrogels measured as a function of oscillatory strain amplitude,  $\gamma_0$  ( $\omega = 6.28$  rad s<sup>-1</sup>).

Table S2 A review on transparent, tough and self-healing hydrogels (RT: room	n
temperature, -: No figures were given)	

Ref.	Ingredient	transparent	Stretchability/	Healing	healing	Voor	
			tough	conditions	efficiencies	real	
1	PAA/Fe <sup>3+</sup>	-	650%	RT/6h	99%	2013	
2	Catechol/	-	5 MPa	5 MDo DT	DT/21h	800/	2016
	$Mg^{2+},Zn^{2+}$			K1/2411	8070	2010	
3	PVA-PAM	92%	200 kPa	-	-	2018	
4	PDMAEA	90%	4 MPa	4 MDa	DT/19h		2010
	-Q/PAA			K1/4811	-	2019	
5	PAM/carra	-	950%	85°C/20min		2010	
	geenan			930%	85 C/30mm	-	2019
	PVA/B	82%	528%/123kDa	DT/5min	08 50/	This	
			52070/125KFa		90.370	work	

## **References:**

[1] Z. Wei, J. He, T. Liang, H. Oh, J. Athas, Z. Tong, C. Wang, Z. Nie, Polym. Chem. 2013,4, 4601-4605.

[2] J. Li, H. Ejima, N. Yoshie. ACS Appl. Mater. Interfaces, 2016, 8, 29, 19047-19053.

[3] G. Ge, Y. Zhang, J. Shao, W. Wang, W. Si, W. Huang, X. Dong, 2018, 28, 32, 1802576-1802584.

[4] Z. Lei, P. Wu. Mater. Horiz., 2019,6, 538-545.

[5] J. Wu, Z. Wu, X. Lu, S. Han, B. Yang, X. Gui, K. Tao, J. Miao, C. Liu. ACS Appl. Mater. Interfaces20191199405-9414.