Supporting Information

Cellulose Nanocrystal Reinforced Hydrogel with Anti-Freezing Properties for Strain Sensor

Jiawen Zheng [a], *Yong Sun*[a,b,]*, *Shuliang Yang* [a], *Zheng Li* [a], *Xing Tang* [a,b], *Xianhai Zeng* [a,b], *Lu Lin* [a,b]

[a] Xiamen Key Laboratory of Clean and High-valued Applications of Biomass, College of Energy, Xiamen University, Xiamen 361102, China.

[b] Fujian Engineering and Research Center of Clean and High-valued Technologies for Biomass, Xiamen University, Xiamen 361102, China.

* Corresponding author.

Yong Sun

Email: sunyong@xmu.edu.cn

Tel.: 00-86-592-5952786; Fax: 00-86-592-5952786

Table S1. Feed information of AA solution and PVA solution for IPN hydrogel formation

Samples	AA (g)	PVA (g)	H_2O (g)	Ethylene glycol (g)	Fe^{3+} (mmol)	CNCs (mg)	APS (mg)	MBAA (mg)
PVA/PAA	6	2	10	10	/	/	50	20
PVA/PAA/CNC-5	6	2	10	10	/	5	50	20
PVA/PAA/CNC-10	6	2	10	10	/	10	50	20
PVA/PAA/CNC-15	6	2	10	10	/	15	50	20
PVA/PAA/CNC-20	6	2	10	10	/	20	50	20
PVA/PAA/CNC-30	6	2	10	10	/	30	50	20
PVA/PAA//Fe-50	6	2	10	10	0.3	/	50	20
PVA/PAA//Fe-100	6	2	10	10	0.6	/	50	20
PVA/PAA//Fe-150	6	2	10	10	0.9	/	50	20
PVA/PAA//Fe-300	6	2	10	10	1.8	/	50	20
PVA/PAA/CNC/Fe-100	6	2	10	10	0.6	10	50	20

Table S2. Mechanical properties of various similar hydrogels

Hydrogel materials	**Elongation （%）**	**Tensile Strength（kPa）**	**Toughness（KJ/m³）**	**Elastic modulus （KPa）**	**Ionic conductivity (S/m)**	**Ref.**
PVA/PAA/CNC/Fe	756	519	179.342	0.69	1.54	This work
GO/PEDOT:PSS/PNIPAM	2512	29	N/G	N/G	0.084	[1]
PNIPAAm/PANI	290	42	N/G	N/G	0.068	[2]
MXene/PVA/PAAm	1000	35	N/G	N/G	N/G	[3]
PAAm/PEDOT:PSS	525	30	N/G	80	1.000	[4]
PAA/TA@CNC/Al	2952	256	5600	48	N/G	[5]
PEG/PAM/PAA/Fe	1350	360	458	N/G	0.006	[6]
PAA/PANI/Fe	991	35.68	N/G	N/G	N/G	[7]
PAM/PVAA/Fe	600	370	N/G	N/G	N/G	[8]
PAA/CNF/Fe	1803	1370	11.05	N/G	N/G	[9]
$_{P}$RGO/PB/TA@CNC	869	245	N/G	N/G	0.0175	[10]

Note: ‘N/G’ indicates ‘not given’ in the references.

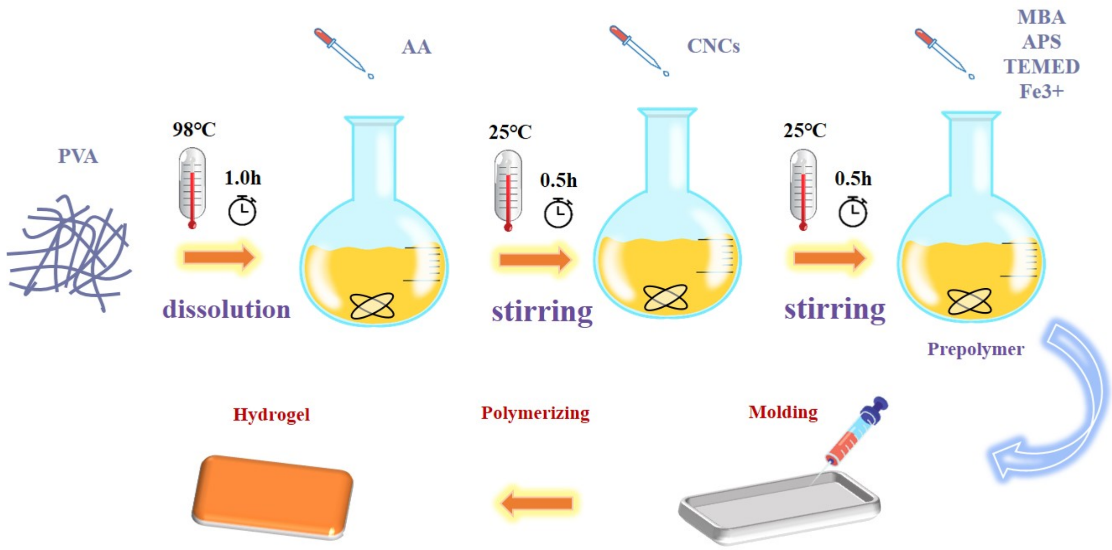

Fig. S1. Preparation of the PVA/PAA/CNC/Fe^{3+} hydrogel.

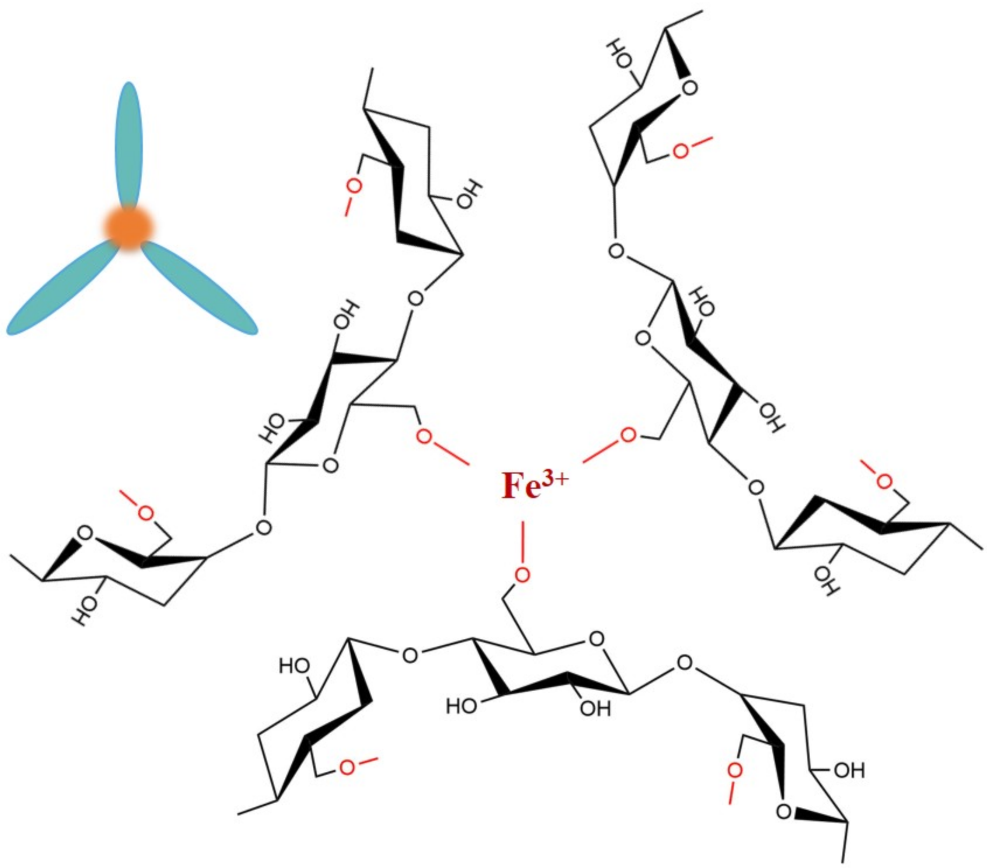

Fig. S2. CNCs−Fe^{3+} binding unit.

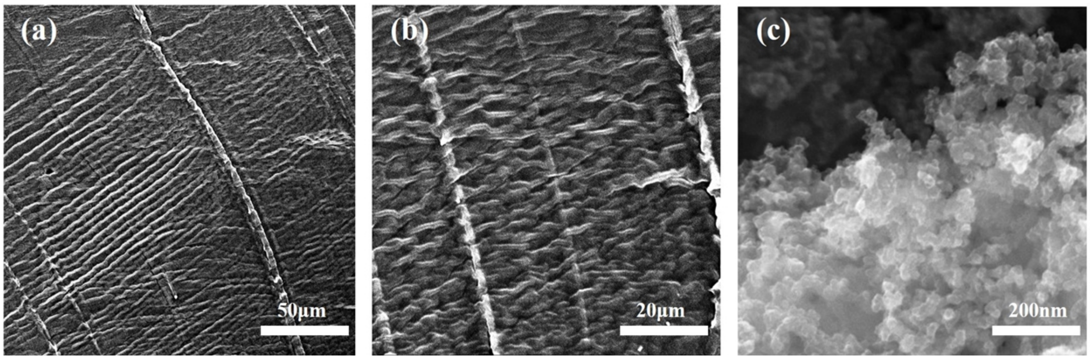

Fig. S3. SEM image of the PVA/PAA/CNC/Fe^{3+} hydrogel

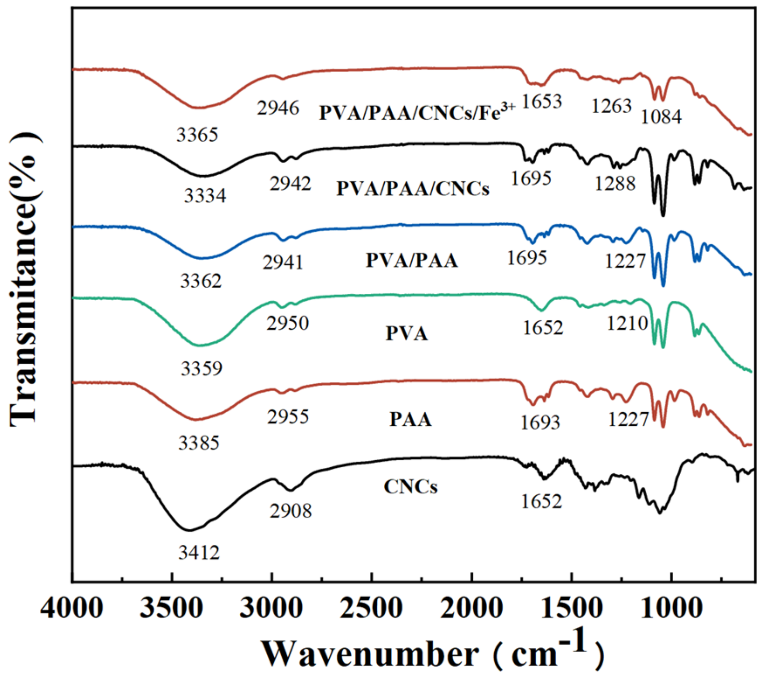

Fig. S4. FTIR spectra of the PAA, PVA, CNCs, PVA/PAA, PVA/PAA/CNC and PVA/PAA/CNC/Fe^{3+} hydrogel

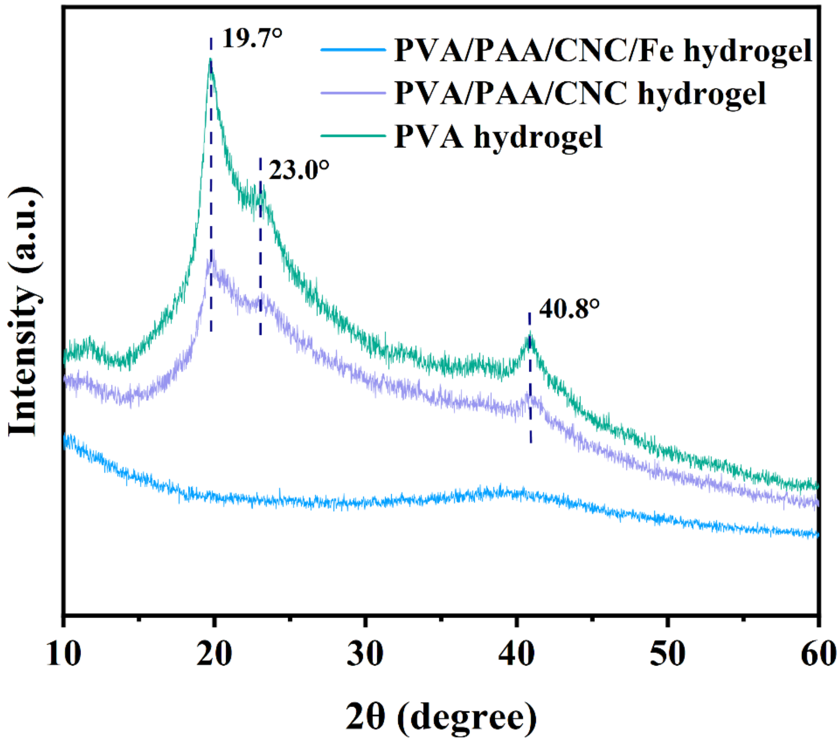

Fig. S5. X-ray diffraction profiles of the PVA, PVA/PAA/CNC, and PVA/PAA/CNC/Fe^{3+} hydrogels.

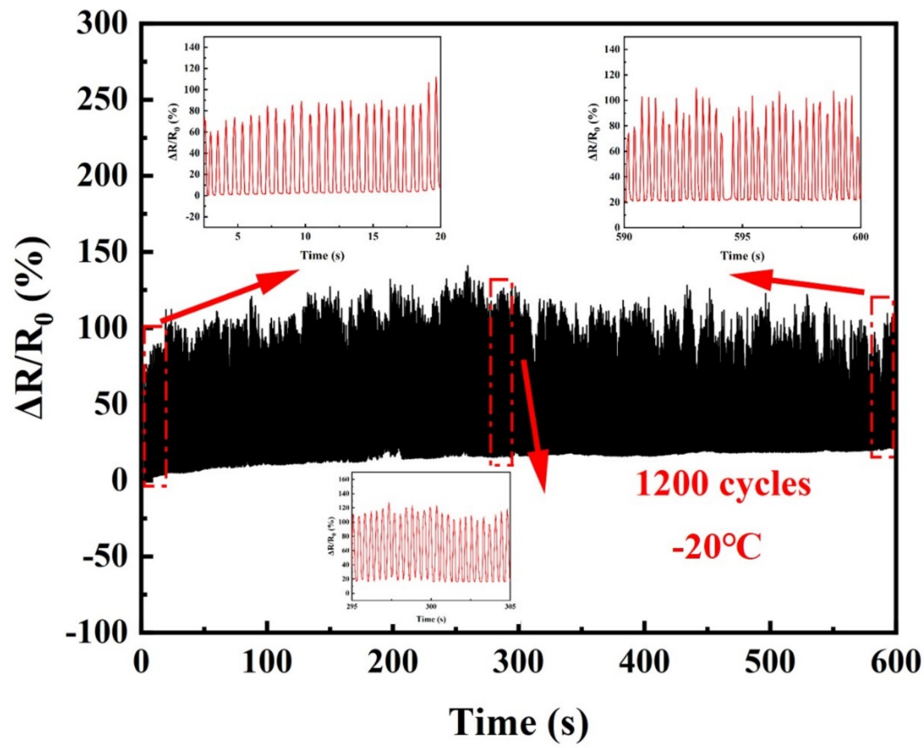

Fig. S6. The PVA/PAA/CNC/Fe^{3+} hydrogel was subjected to 1200 cycles of the tensile test at low temperature.

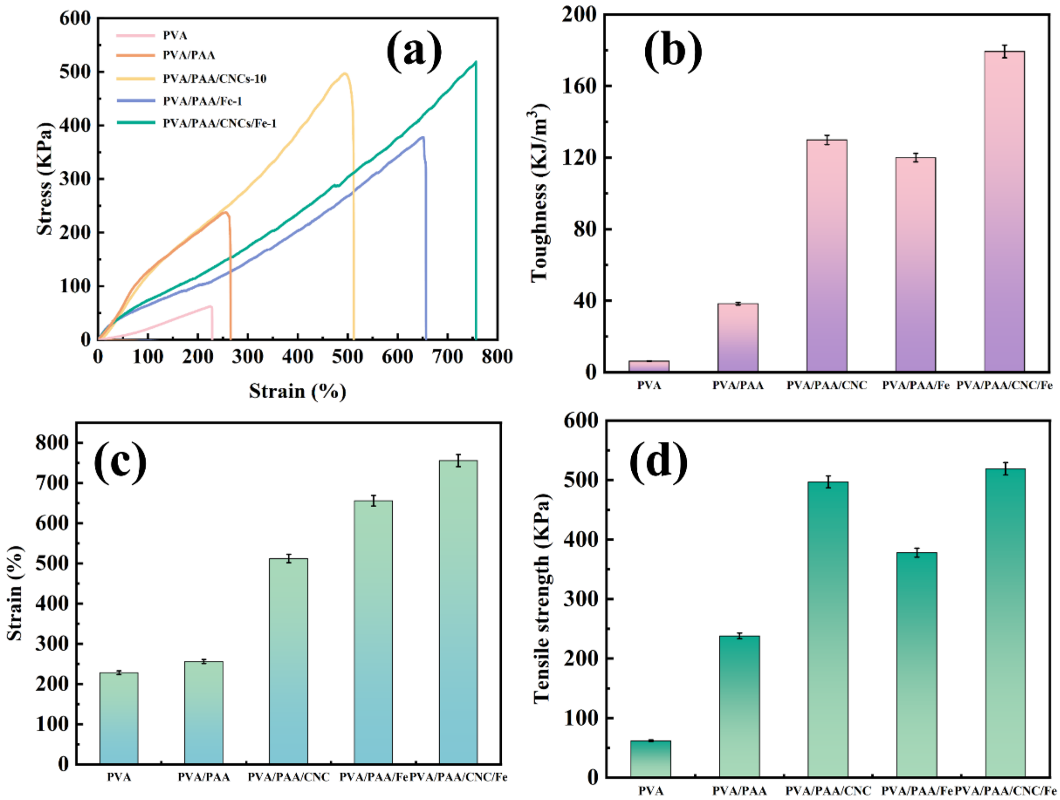

Fig. S7. Tensile tests of IPN hydrogels: (a) Typical stress-train curves of different hydrogels; Toughness (b), Strain (c), and Tensile strength (d) of different hydrogels.

When only Fe^{3+} was added, the tensile length of PVA/PAA/Fe increased significantly from 256% to 656%, which was higher than 512% of PVA/PAA/CNC, but the tensile strength and toughness were only 378 KPa and 119 KJ /m^3, which was lower than 497 KPa and 129 KJ/m^3 of PVA/PAA/CNC, these changes demonstrated that hydrogen bonds and ionic coordinate bonds could improve the mechanical properties of hydrogels together.

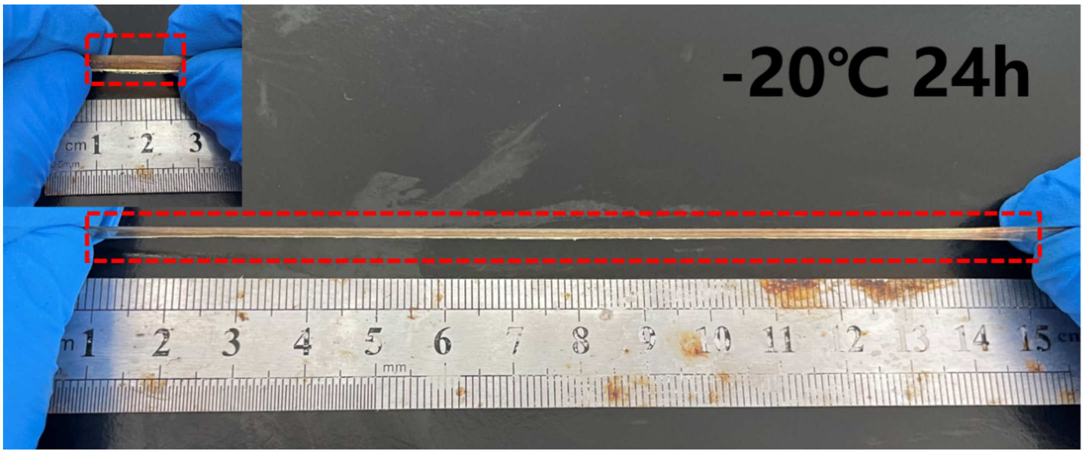

Fig. S8. Highly stretched after 24 h of freezing.

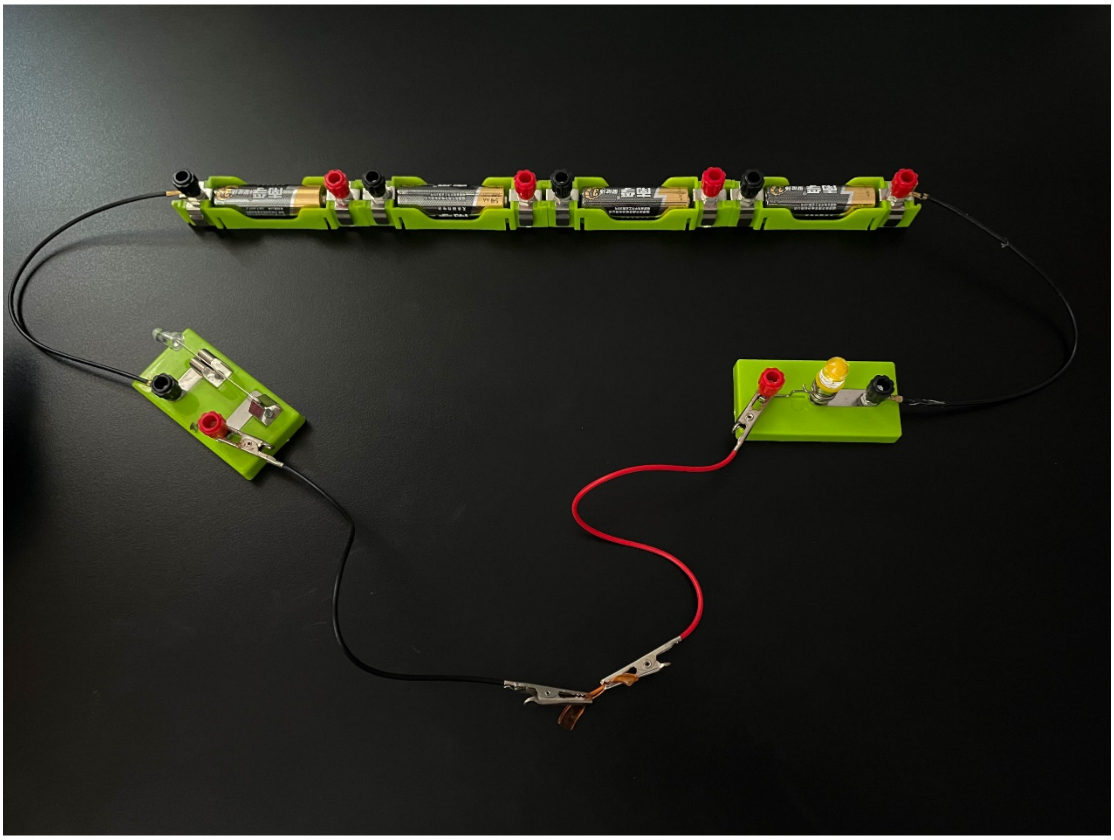

Fig. S9. The PVA/PAA/CNC/Fe^{3+} hydrogel could be electrically conductive at -20 °C.

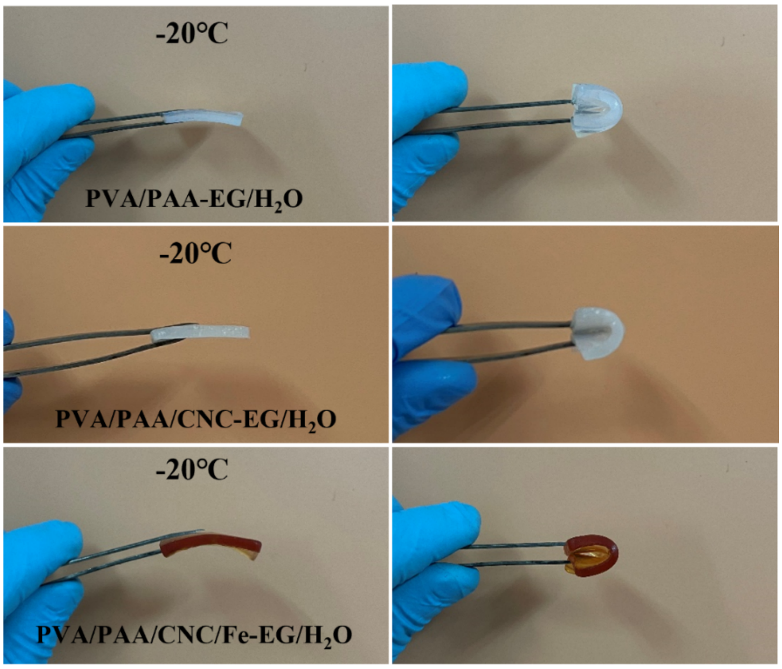

Fig. S10. PVA/PAA, PVA/PAA/CNC, and PVA/PAA/CNC/Fe hydrogels can be bent after being frozen at -20℃ for 24 hours.

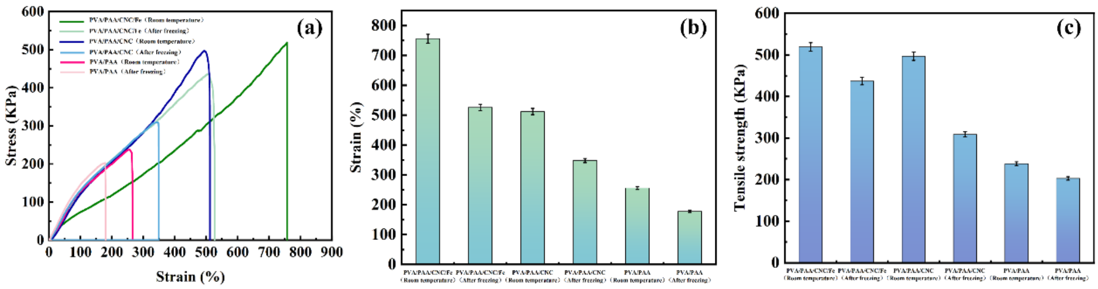

Fig. S11. Tensile tests of PVA/PAA, PVA/PAA/CNC, and PVA/PAA/CNC/Fe hydrogels at room temperature and after freezing: (a) Typical stress-train curves of different hydrogels; Strain (b), Tensile strength (c) of different hydrogels.

The tensile strain and tensile strength of different hydrogels decreased to different degrees after 24 hours of freezing at -20℃. The tensile length of PVA/PAA/CNC/Fe^{3+}， PVA/PAA/CNC， and PVA/PAA hydrogels decreased to 69.5%, 67.9%, and 69.5% of that at room temperature, respectively. It can be seen that the addition of CNC and Fe^{3+} had little effect on the tensile length at low temperatures. At the same time, the tensile strength decreased to 84.2%, 77.0%, and 85.2% of that at room temperature. When only CNC was added, the tensile strength of hydrogel decreased more at low temperature.

Reference

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