Supporting Information

Robust-adhesion and high-mechanical strength hydrogel for efficient wet tissue adhesion

Chenyang Li^a, Yang Qian^a, Xueping Zhang^{a*}, and Rongwu Wang^{a*}

^aKey Laboratory of Textile Science & Technology, Ministry of Education, College of

Textiles, Donghua University, Shanghai 201620, China

Chenyang Li and Yang Qian contributed equally to this work.

*Corresponding author. E-mails: <u>xpzhang@dhu.edu.cn</u>, <u>wrw@dhu.edu.cn</u>.

This file includes:

Figures S1 to S9

Tables S1 to S4

Videos S1 to S4



Figure S1. Schematic diagram of the synthesis of C-CS.



Figure S2. FT-IR spectra of CS and C-CS.



Figure S3. UV-Vis spectra of CS and C-CS.



Figure S4. a) UV-Vis spectra of CS and different concentrations of HCA, b) standard working

curve.

Table S1. Absorbance at 286 nm for different concentrations of HCA solution.

HCA solution (mmol/L)	Absorbance
0.025	0.076
0.05	0.148
0.1	0.339
0.2	0.563
0.4	0.984

Samples	AA (wt%)	C-CS (wt%)	α-ketoglutaric acid (wt%)	AA-NHS ester (wt%)	PEGDMA (wt%)	Water (g)
PAA20-NHS1/C- CS2	20	2	0.2	1	0.05	8
PAA30-NHS1/C- CS2	30	2	0.2	1	0.05	7
PAA40-NHS1/C- CS2	40	2	0.2	1	0.05	6
PAA30/C-CS4	30	4	0.2	0	0.05	7
PAA30-NHS0.33/C- CS4	30	4	0.2	0.33	0.05	7
PAA30-NHS0.67/C- CS4	30	4	0.2	0.67	0.05	7
PAA30-NHS1	30	0	0.2	1	0.05	7
PAA30-NHS1/C- CS1	30	1	0.2	1	0.05	7
PAA30-NHS1/C- CS4	30	4	0.2	1	0.05	7
PAA30-NHS1/C- CS6	30	6	0.2	1	0.05	7

 Table S2. Composition of different hydrogels.



Figure S5. Chemical composition of the PAA-NHS/C-CS hydrogel based on PAA-NHS and

C-CS.



Figure S6. High-magnific SEM images of the freeze-dried PAA30/C-CS4, PAA30-NHS1, and

PAA30-NHS1/C-CS4 hydrogel (from left to right).



Figure S7. Stress-strain curves for the dry PAA30-NHS1/C-CS4 hydrogel.



Figure S8. Effect of AA, AA-NHS ester, C-CS, and PEGDMA contents on the tensile properties of swollen hydrogels. Tensile stress-strain curves of swollen hydrogels with different (a) AA content, (b) AA-NHS ester content, (c) C-CS content, and (d) PEGDMA content.

	Name	Tensile stress (kPa)	Strain (%)
Varied AA content	PAA20-NHS1/C-CS2	246.5±13.44	1265.73±92.33
	PAA30-NHS1/C-CS2	457.66±21.08	1847.14±86.07
	PAA40-NHS1/C-CS2	121.5±7.78	840.74±53.05
Varied AA-NHS ester content	PAA30/C-CS4	253±45.74	1271.30±18.31
	PAA30-NHS0.33/C-CS4	269.67±7.09	1535.39±85.83
	PAA30-NHS0.67/C-CS4	386.67±53.27	1585.28±42.20
	PAA30-NHS1/C-CS4	632±60.81	1974.19±96.03
Varied C-CS content	PAA30-NHS1	85±7.21	562.69±105.72
	PAA30-NHS1/C-CS1	275.33±46.17	1028.18±77.35
	PAA30-NHS1/C-CS2	457.66±21.08	1847.14±86.07
	PAA30-NHS1/C-CS4	632±60.81	1974.19±96.03
	PAA30-NHS1/C-CS6	561.33±37.91	1422.91±157.55
PAA30-NHS1/C-CS4 with varied PEGDMA content	5 mg	632±60.81	1974.19±96.03
	10 mg	349.5±22.31	1805.64±193.84
	15 mg	234.5±28.29	1100.6±118.99

 Table S3. Mechanical properties of hydrogels.

 Table S4. Comparison of wet tissue adhesive properties of PAA30-NHS1/C-CS4 hydrogel with

 other reported hydrogel adhesives.

Name	Adhesion strength test Method	Wet adhesion strength to porcine skin	Reference
PDA-Silicate-CG	Shear	21.3 kPa	1
TA/PVA/PAA	Shear	31 kPa	2
CMC-DA/TA	Shear	45.9 kPa	3
PEG-SG/TA	Shear	62 kPa	4
PVA/PAAc-N	Shear	63.1 kPa	5
Fe-PAM-C-M	Shear	77.5 kPa	6
P (AA-co-UCAT5) –CS3	Shear	187.1 kPa	7
G0.6-T0.6-U0.12	Shear	152.9 kPa	8
PAA30-NHS1/C-CS4	Shear	163 kPa	This work



Figure S9. a) The N1s spectrum of untreated porcine skin. The peak at the binding energy of 399.9 eV could be attributed to C-N, demonstrating the presence of the -NH₂ group in the porcine skin. b) The C1s spectrum of untreated porcine skin. The peaks at the binding energy of 284.8, 286.6, 287.6, and 289.0 eV could be assigned to C-C, C-O, N-C=O, and O-C=O, respectively. c) and d) The N1s and C1s spectrum of porcine skin after 30-minute adhesion of PAA30-NHS1/C-CS hydrogel.

Supporting Videos

Video S1. The overall process of adhesion formation between wet porcine skins by the PAA30-NHS1/C-CS4 hydrogel.

Video S2. The PAA30-NHS1/C-CS4 hydrogel adhered for 5 min is peeled off from the porcine skin.

Video S3. The repeatable underwater adhesion of the PAA30-NHS1/C-CS4 hydrogel to porcine skin.

Video S4. Different adhesive properties of the PAA30-NHS1/C-CS4 hydrogel to underwater glass and porcine skin.

Supporting References

- 1. Y. Chen, Y. Qiu, Q. Wang, D. Li, T. Hussain, H. Ke and Q. Wei, *Chemical Engineering Journal*, 2020, **399**, 125668.
- J. Park, T. Y. Kim, Y. Kim, S. An, K. S. Kim, M. Kang, S. A. Kim, J. Kim, J. Lee, S.-W. Cho and J. Seo, *Advanced Science*, 2023, 10, 2303651.
- 3. H. Xie, G. Shi, R. Wang, X. Jiang, Q. Chen, A. Yu and A. Lu, *Carbohydrate Polymers*, 2024, **334**, 122014.
- 4. F. Sun, Y. Bu, Y. Chen, F. Yang, J. Yu and D. Wu, *ACS Applied Materials & Interfaces*, 2020, **12**, 9132-9140.
- 5. P. Ma, W. Liang, R. Huang, B. Zheng, K. Feng, W. He, Z. Huang, H. Shen, H. Wang and D. Wu, *Advanced Materials*, 2024, **36**, 2305400.
- 6. L. Han, M. Wang, L. O. Prieto-López, X. Deng and J. Cui, *Advanced Functional Materials*, 2020, **30**, 1907064.
- 7. X. Fan, Y. Fang, W. Zhou, L. Yan, Y. Xu, H. Zhu and H. Liu, *Materials Horizons*, 2021, **8**, 997-1007.
- 8. X. Su, W. Xie, P. Wang, Z. Tian, H. Wang, Z. Yuan, X. Liu and J. Huang, *Materials Horizons*, 2021, **8**, 2199-2207.