

Supporting Information

Mussel-inspired hybrid network hydrogel for continuous adhesion in water

Yifu Fu, Pengfei Ren, Faming Wang, Min Liang, Wanjun Hu, Naizhen Zhou, Zuhong Lu, Tianzhu Zhang *

State Key Lab of Bioelectronics, National Demonstration Center for Experimental Biomedical Engineering Education, School of Biological Science and Medical Engineering, Southeast University, Nanjing 210096, China.

* Corresponding authors. E-mail addresses: zhangtianzhu@seu.edu.cn (T. Zhang).

Table S1. The comparison of previously reported adhesives

Number	Adhesive	Substrate	Size	Adhesion strength (kPa)	Adhesion time	Ref
1	OCMC-DA/PAM	Glass	20 mm * 25 mm	86.3 ± 7.2	More than two weeks	This work
2	PDMS-PTFE	Copper	18 mm * 6 mm	300	Not mentioned	1
3	OSA-DA/PAM	Glass	10 mm * 3 mm	10	Not mentioned	2
4	PDA-PAM	skin	25 mm * 3 mm	30	Not mentioned	3
5	CMC-DA	Porcine skin	20 mm * 10 mm	30	Not mentioned	4
6	PAM-Col-COA	Porcine skin	10 mm * 10 mm	15	Not mentioned	5
7	Aa-BA	Glass	30 mm * 20 mm	120	Not mentioned	6
8	STP	Glass	25 mm * 1 mm	45	Not mentioned	7

The OCMC-DA/PAM hydrogel cannot form when the content of OCMC-DA reached 100 mg, because excessive DA reduced the activity of ammonium persulfate, thus inhibiting the polymeric reaction of acrylamide monomers.

Table S2. The preparation formula of various hydrogels (copolymerization was initiated by APS (200 mg) with 15 μ L of TEMED as catalyst)

Hydrogels	AM (mg)	OCMC-DA (mg)	BIS (mg)
PAM5	2000	0	5
OCMC-DA1/PAM5	2000	10	5
OCMC-DA3/PAM5	2000	30	5
OCMC-DA6/PAM5	2000	60	5
OCMC-DA9/PAM5	2000	90	5
OCMC-DA1/PAM3	2000	10	3
OCMC-DA1/PAM6	2000	10	6
OCMC-DA1/PAM9	2000	10	9
OCMC-DA9/PAM3	2000	90	3
OCMC-DA9/PAM6	2000	90	6
OCMC-DA9/PAM9	2000	90	9

Degree of Substitution of Dopamine

The degree of substitution of dopamine is measured by UV-Vis Spectroscopy. Briefly, the working solution was prepared by diluting the stock solution of 10 mg/mL to a series of graded concentrations, including 0.1 mg/mL, 0.15 mg/mL, 0.2 mg/mL, 0.25 mg/mL, 0.3 mg/mL and 0.35 mg/mL, which was applied in UV-Vis Spectroscopy to determine the standard curve of dopamine, the relationship of absorbance (280 nm) and concentration of DA.

Figure S1 showed the standard curve of dopamine, which had good linearity over the range of 0.01 ~ 0.2 mg/L, the calibration equation was $y = 2.9304x + 2.2923$ with R^2 of 0.9965. UV-Vis spectrum of OCMC-DA (2 mg/mL) was shown in Figure 1. Compared with the standard curve of dopamine, the amount of DA moieties can be calculated and the degree of substitution of dopamine is 15.2%.

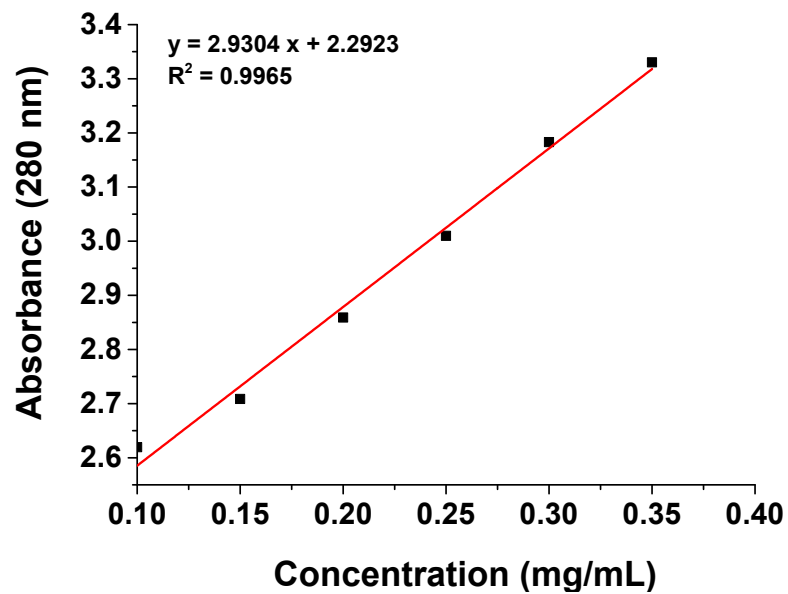


Figure S1. Standard curve of dopamine measured at a series of concentrations.



Figure S2. Optical images of OCMC-DA6/PAM5 hydrogel sample with an initial length of 20 mm (a) and a stretched length of 200 mm (b).

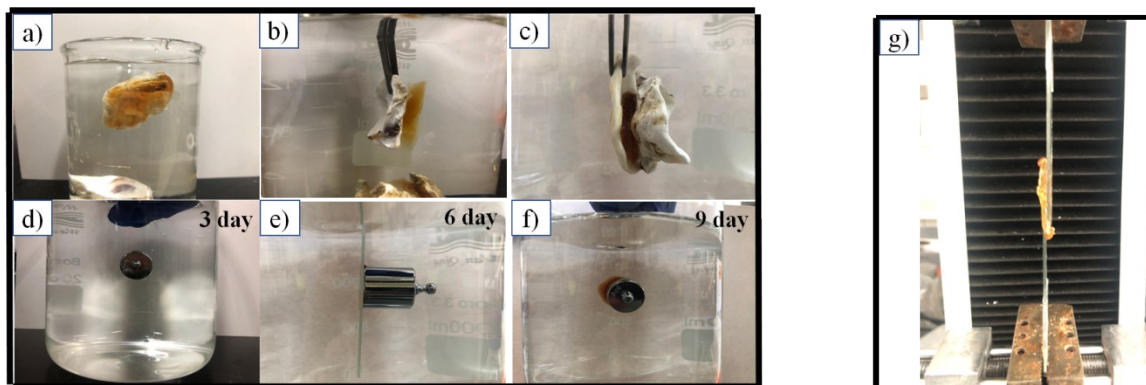


Figure S3. Adhesion ability of OCMC-DA6/PAM5 hydrogel. (a) and (b) OCMC-DA6/PAM5 hydrogel adhered the shell to the beaker wall under water and fell off after fifteen days; (c) OCMC-DA6/PAM5 hydrogel adhered two shells under water; (d)-(f) The OCMC-DA6/PAM5 hydrogel adhered a 50 g weight to the glass for 9 days.

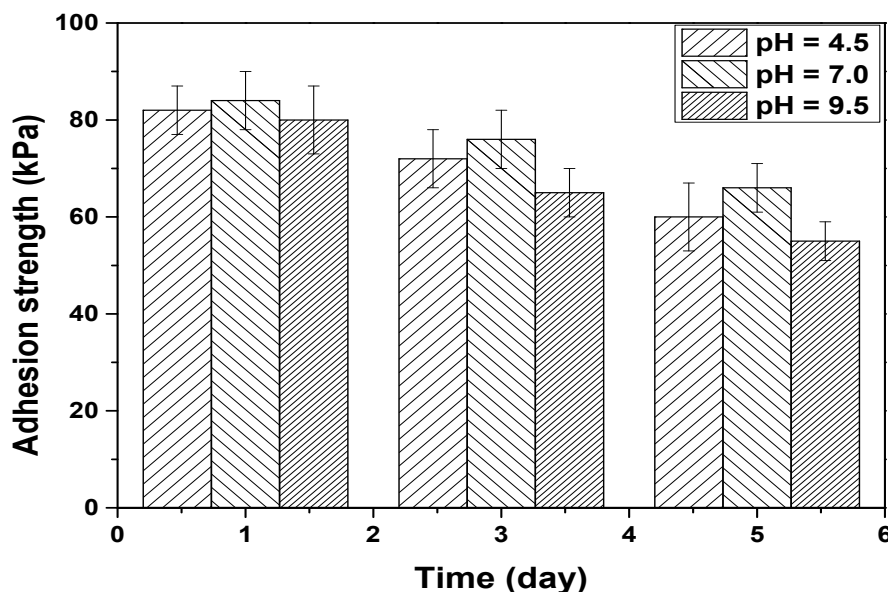


Figure S4. The influence of pH on adhesion strength of OCMC-DA6/PAM5.

References

1. A. C. Chipara, T. Tsafack, P. S. Owuor, J. Yeon, C. E. Junkermeier, A. C. T. van Duin, S. Bhowmick, S. A. S. Asif, S. Radhakrishnan, J. H. Park, G. Brunetto, B. A. Kaiparettu, D. S. Galvão, M. Chipara, J. Lou, H. H. Tsang, M. Dubey, R. Vajtai, C. S. Tiwary and P. M. Ajayan, *Materials Today Chemistry*, 2018, **9**, 149-157.
2. T. Chen, Y. Chen, H. U. Rehman, Z. Chen, Z. Yang, M. Wang, H. Li and H. Liu, *ACS Appl Mater Interfaces*, 2018, **10**, 33523-33531.
3. L. Han, L. Yan, K. Wang, L. Fang, H. Zhang, Y. Tang, Y. Ding, L.-T. Weng, J. Xu, J. Weng, Y. Liu, F. Ren and X. Lu, *NPG Asia Materials*, 2017, **9**, e372-e372.
4. Y. Zhong, J. Wang, Z. Yuan, Y. Wang, Z. Xi, L. Li, Z. Liu and X. Guo, *Colloids Surf B Biointerfaces*, 2019, **179**, 462-469.
5. Z. Bai, W. Dan, G. Yu, Y. Wang, Y. Chen, Y. Huang, C. Yang and N. Dan, *RSC Advances*, 2018, **8**, 42123-42132.
6. X. Liu, Q. Zhang, L. Duan and G. Gao, *ACS Applied Materials & Interfaces*, 2019, **11**, 6644-6651.
7. H. Qiao, P. Qi, X. Zhang, L. Wan3g, Y. Tan, Z. Luan, Y. Xia, Y. Li and K. Sui, *ACS Applied Materials & Interfaces*, 2019, **11**, 7755-7763.