Electronic Supplementary Material (ESI) for Journal of Materials Chemistry B.


# Supporting Information

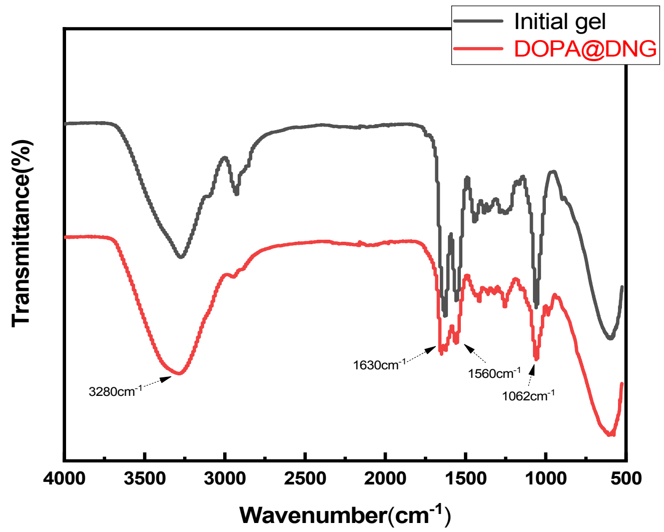


**Fig. 1.** FTIR thermogram of Initial gel and DOPA@DNG.

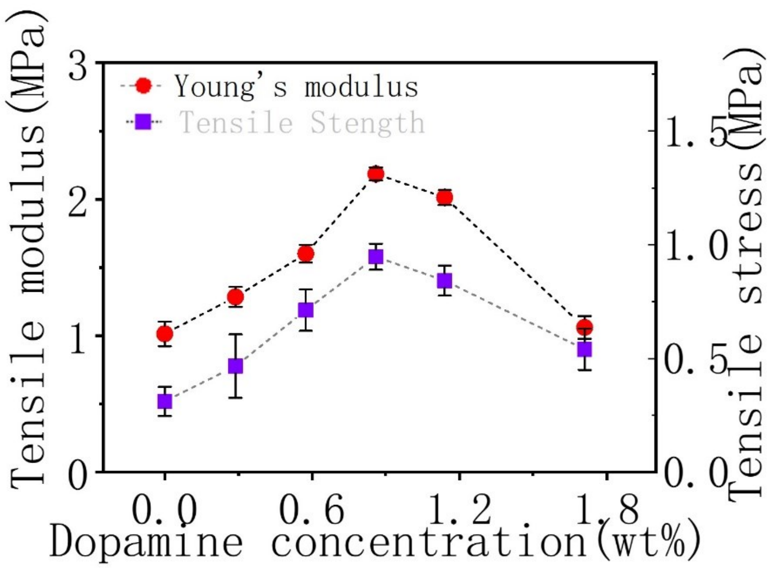


**Fig. 2.** Mechanical properties of different concentrations of DOPA@DNG.

Compared with Initial gel, the weight loss temperature of DOPA @ DNG is slightly increased. the unfolding process of the agar spiral chain was 185° C, which indicated that the mussel structure has good high temperature resistance. Therefore, the chemical structure of DOPA@DNG has not changed at room temperature and constant temperature of 37℃±2℃, which proved that its stable performance.

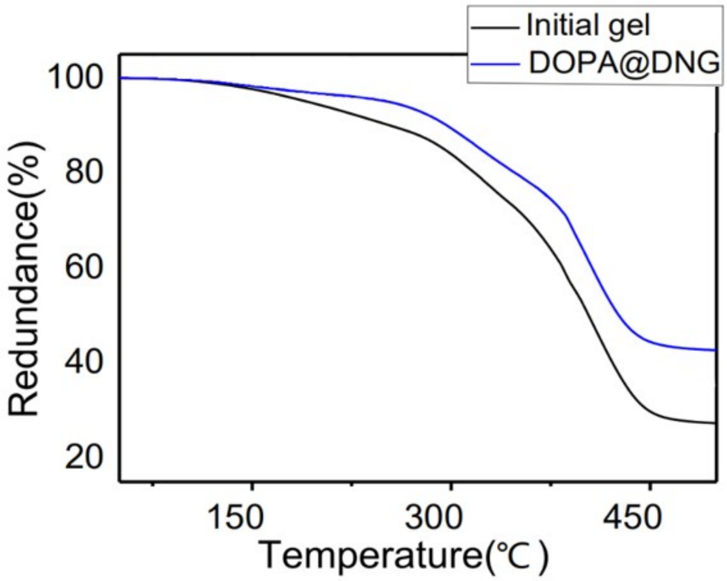


**Fig. 3.** TGA thermogram of Initial gel and DOPA@DNG.

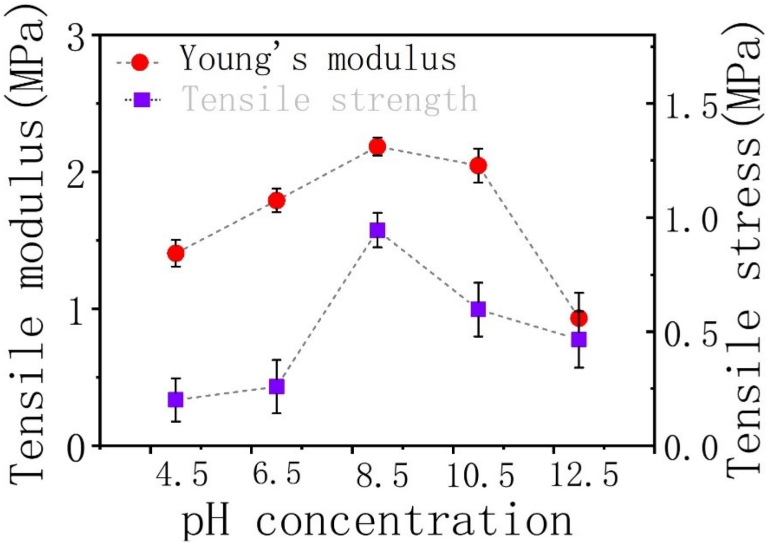


**Fig. 4.** The influence of pH on the mechanical properties of DOPA@DNG.

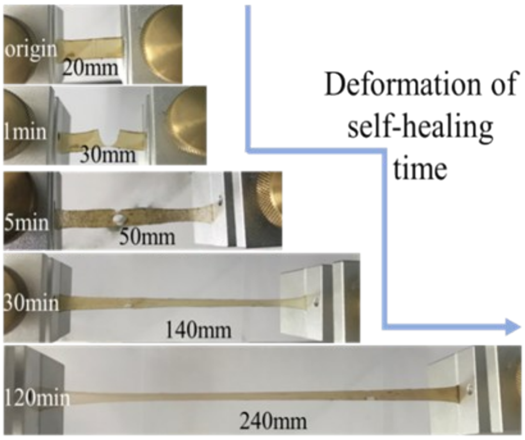


**Fig. 5.** DOPA@DNG deformation of self-healing time.

The FTIR spectra showed the characteristic -NH and -OH peak at 3280 $cm^{-1}$ from ZnO NG were enhanced indicate that H-bond was formed by the reaction between the hydroxyl groups on the surface of ZnO NPs and the functional groups in the polymer, which made the gel network tighter. And the characteristic peak of ZnO NG (1062 $cm^{-1}$, 1560 $cm^{-1}$, 1630 $cm^{-1}$, 3280 $cm^{-1}$) is shifted from that of the initial gel, which indicated that the ZnO NPs are successfully recombined in the gel network.

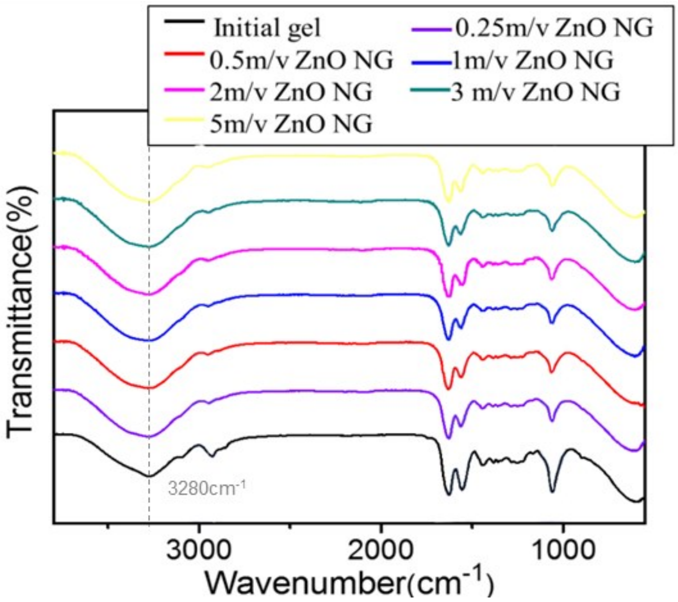


**Fig. 6.** FTIR of different concentrations of ZnO@NG.

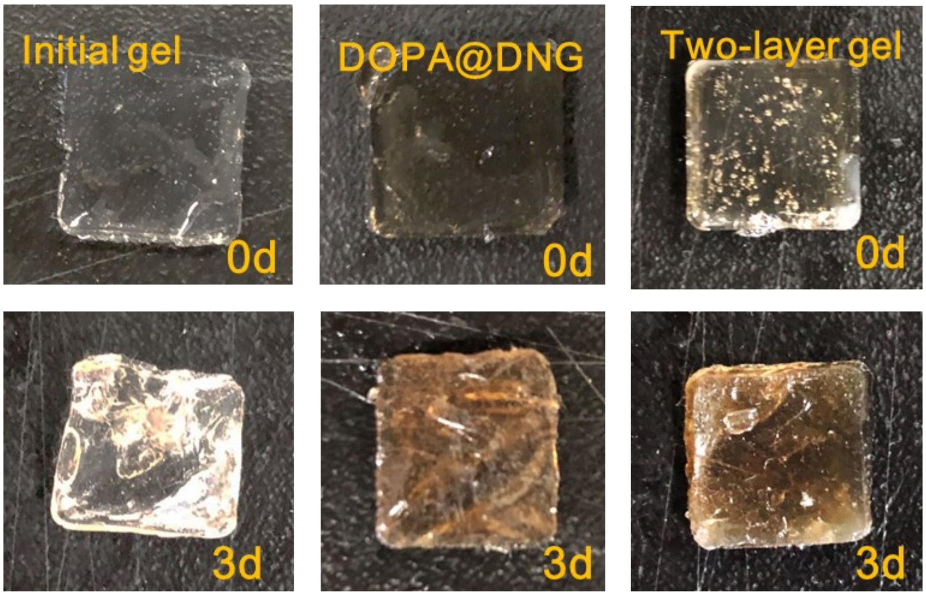


**Fig. 7.** Images of Initial gel, DOPA@DNG, Two-layer gel stored at 28 ℃ and 56% RH for 0 and 3 days.

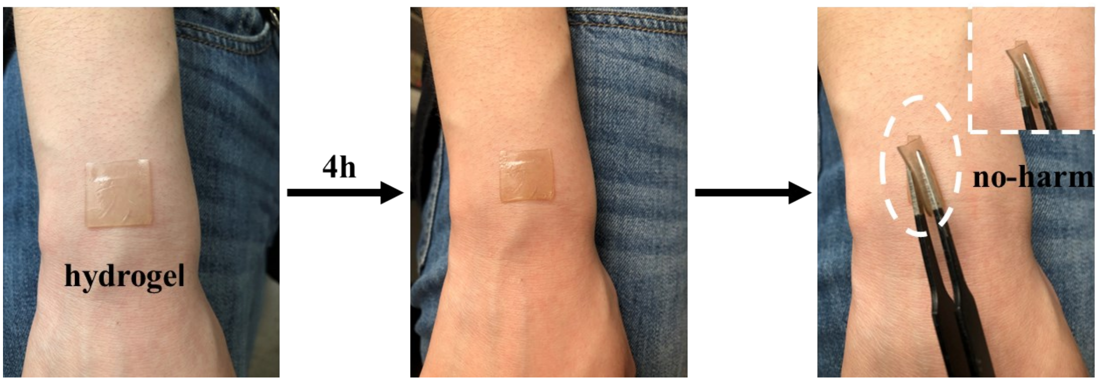


**Fig. 8.** The long-term adhesion of Two-layer gel to skin.

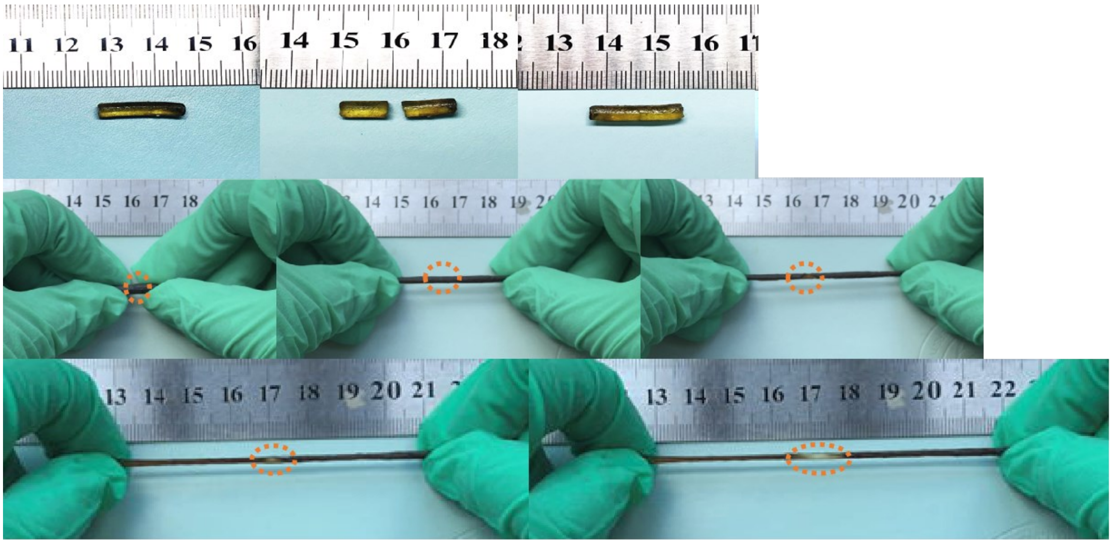

**Fig. 9.** Self-healing properties of Two-layer gel.

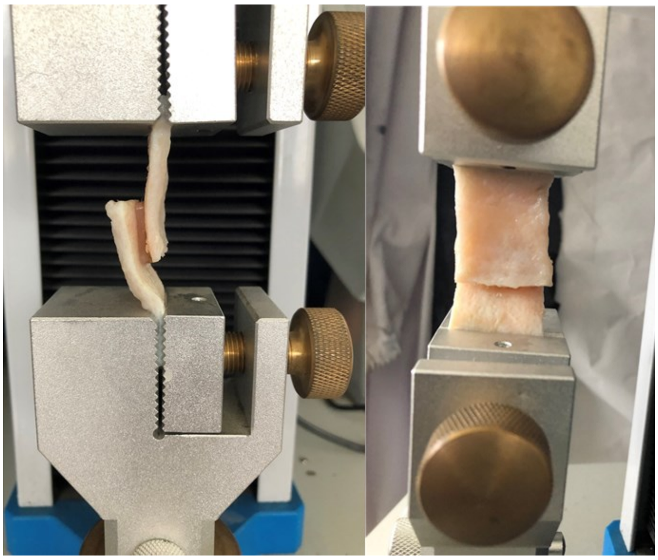

**Fig. 10.** Experimental setup on Two-layer gel adhered between the two porcine skins.

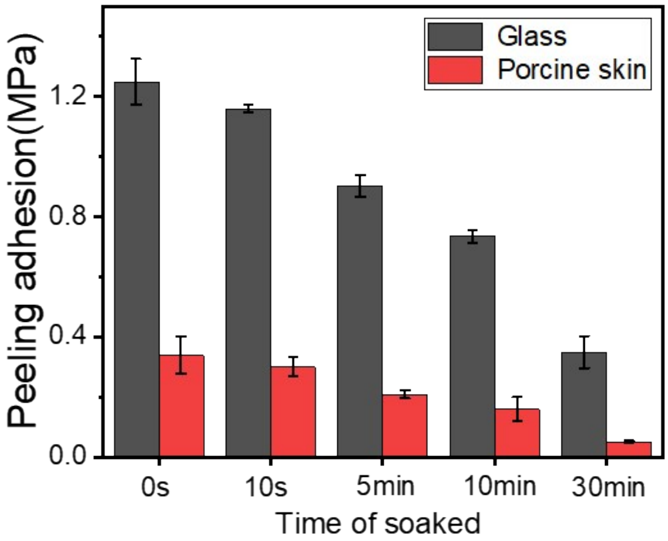


**Fig. 11.** The adhesion properties of Two-layer gel with different times of soaked in oil

**Table S1.** Comparison of the Recovery rate of tensile strain after healing and self-healing temperature in the reported literature

| Copolymers | Self-healing temperature(°C) | Self-healing time | Recovery rate of tensile strain after healing | Refs |
|---|---|---|---|---|
| Agar/pHEAA DN hydrogels | 95 | 12h | 12.5% | [1] |
| Agar/pHEAA-AAc-Fe3+ DN gels | 80 | 12h | 4.36% | [2] |
| Agar/pHEAA@ MPS-CSNPs gels | 95 | 12h | 13.3% | [3] |
| Agar/acrylamide hydrogels | 95 | 6h | 14% | [4] |
| agar-PAM/GO DN gel | 95 | 1h | 31.9% | [5] |
| DOPA@DNG | 100 | 2h | 67% | This work |
| PLL/ Aga DN hydrogel | 120 | 10min | 61% | [6] |
| Agarose/PAM/Laponite hydrogels | 95 | 20min | 80% | [7] |
| Agar/pAAEE DN Gels. | 80 | 30min | 92% | [8] |

References:


[1] H. Chen, Y. L. Liu, B. Q. Ren, Y. X. Zhang, J. Ma, L. J. Xu, Q. Chen, and J. Zheng, Advanced Functional Materials, 2017, 27.

[2] D. Zhang, F. Y. Yang, J. He, L. J. Xu, T. Wang, Z. Q. Feng, Y. Chang, X. Gong, G. Zhang, and J. Zheng, ACS Appl. Polym. Materials, 2020, 2, 1031-1041.

[3] S. W. Xie, B. P. Ren, G. Gong, D. Zhang, Y. Chen, L. J. Xu, C. F. Zhang, J. X. Xu, and J. Zheng, ACS Appl. Nano Mater, 2020, 3, 2774−2786.

[4] D. D. Wei, J. Yang, L. Zhu, F. Chen, Z. Q. Tang, G. Qin and Qi. Chen, Polymer Testing, 2018,

69, 167-174.
[5] P. Zhu, M. Hu, Y. H. Deng and C. Y. Wang, Adv. Eng Mater, 2016, 18.
[6] K. Lei, K. Q. Wang, Y. L. Sun, Z. Zheng, and X. L. Wang, Advanced Functional Materials, 2021, 31.
[7] J. H. Guo, R. R. Zhang and L. N. Zhang, ACS Macro Lett, 2018, 7, 442.
[8] Y. X. Zhang, B. P. Ren, S. W. Xie, Y. Q. Cai, T. Wang, Z. Q. Feng, J. X. Tang, Q. Chen, J. X. Xu, L. J. Xu, and J. Zheng, ACS Appl. Polym. Mater. 2019, 1, 701−713.