Supporting Information

Highly Stretchable, Self-healing and Conductive Silk Fibroin-based Dual Network Gels via Sonication-induced and Self-emulsifying Green Procedure

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Preparation of regenerated silk fibroin (RSF) solutions

RSF aqueous solution was prepared according to previously described procedure[1]. In brief, cocoons pieces were boiled twice for 30 min with 0.5 wt% Na_2CO_3 solution, then washed it three times with deionized water at 60 ℃ to remove the residual sericin. After drying at 30℃ overnight, the degummed fibers were dissolved in a 9.3 M LiBr solution at 60 ℃ for 2 h. After filtration, the silk solution was dialyzed against deionized water for 72 h with dialysis tubing (MWCO 14000 ± 8000 Da) to remove salts. Finally, the solution was concentrated by forced airflow to produce a 30 wt% RSF aqueous solution.

SF/ C18M O/W Emulsions

Silk fibroin (SF) has good amphiphilic and surface activity, we chose it to emulsify the C18M oil phase. The preliminary experimental results of dispersing C18M with SF as emulsifier are shown in Table S1. It can be seen that with the decrease of SF concentration from 5 to 2 wt%, the particle size of SF/C18M emulsions decreased from 1277 to 805.9 nm, which indicating that 2 wt% SF had a good emulsifying effect on C18M oil phase.

Table S1. The micelles size of SF/C18M emulsions with different SF concentration

SF (g/mL)	C18M quality (mM/ml)	Droplet size (nm)	PDI
5	0.0746	1277.0	0.287
4	0.0746	1178.0	0.263
3	0.0746	937.2	0.253
2	0.0746	805.9	0.220

Self-healing properties

Strain-stress curves of RSFx/S – PAAm/C18 DN gel at different RSF content before and after self-healing are shown in Fig.S1. It can be seen that with the increase of RSF content from 6.1 to 7.7(wt/v) %, the self-healing rate of stress increased from85%± 2% to 88%± 2% and the self-healing rate of strain increased from 73% ±2% to 95%± 2%； However, when the RSF content was increased to 8.9(wt/v) %, the self-healing rate of stress decreased to 87%± 2% and the self-healing rate of strain decreased to 87%± 2%. This is because when the RSF content was too high, the β-sheet conformation was excessive lead to the inter molecular hydrogen bond force in the gel was too strong, which prevented the movement of RSF chains the self-healing ability of the gel decreased instead. At the same time, as a result, the fracture stress and fracture strain of the gel are greatly reduced.

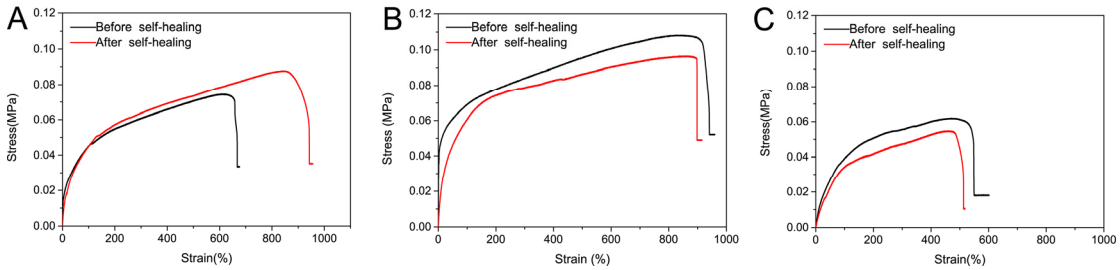

Fig. S1 Stress- strain curves of RSFx/S – PAAm/C18 DN gel at different RSF content before and after self-healing (A) RSF6.1/S – PAAm/C18 DN gel; (B) RSF7.7/S – PAAm/C18 DN gel; (C) RSF8.9/S – PAAm/C18 DN gel.

Reference

1. J. Li, J. Zhu, L. Jia, Y. Ma and H. Wu, *RSC Advances*, 2020, **10**, 323-331.