

Supporting Information

Jellyfish-inspired three-network crosslinked alginate composite hydrogel prepared by macro-micro double bionic strategy for efficient multifunctional water purification

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Table S1. Fabrication of hydrogels with varying SA/k-CG ratios.

	SA/k-CG ratio	SA powder	K-CG powder	PVA
	3:1	1.5 g	0.5 g	
	3:2	1.2 g	0.8 g	
2wt% CaCl ₂	1:1	1 g	1 g	3 g
	2:3	0.8 g	1.2 g	
	1:3	0.5 g	1.5 g	

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Table S2. Fabrication of hydrogels with different CaCl₂ concentrations.

	CaCl ₂ concentration	PVA
3:2 SA/k-CG	1wt%	
	2wt%	
	3wt%	3 g
	4wt%	
	5wt%	

1. Micro umbrella structure physical chart

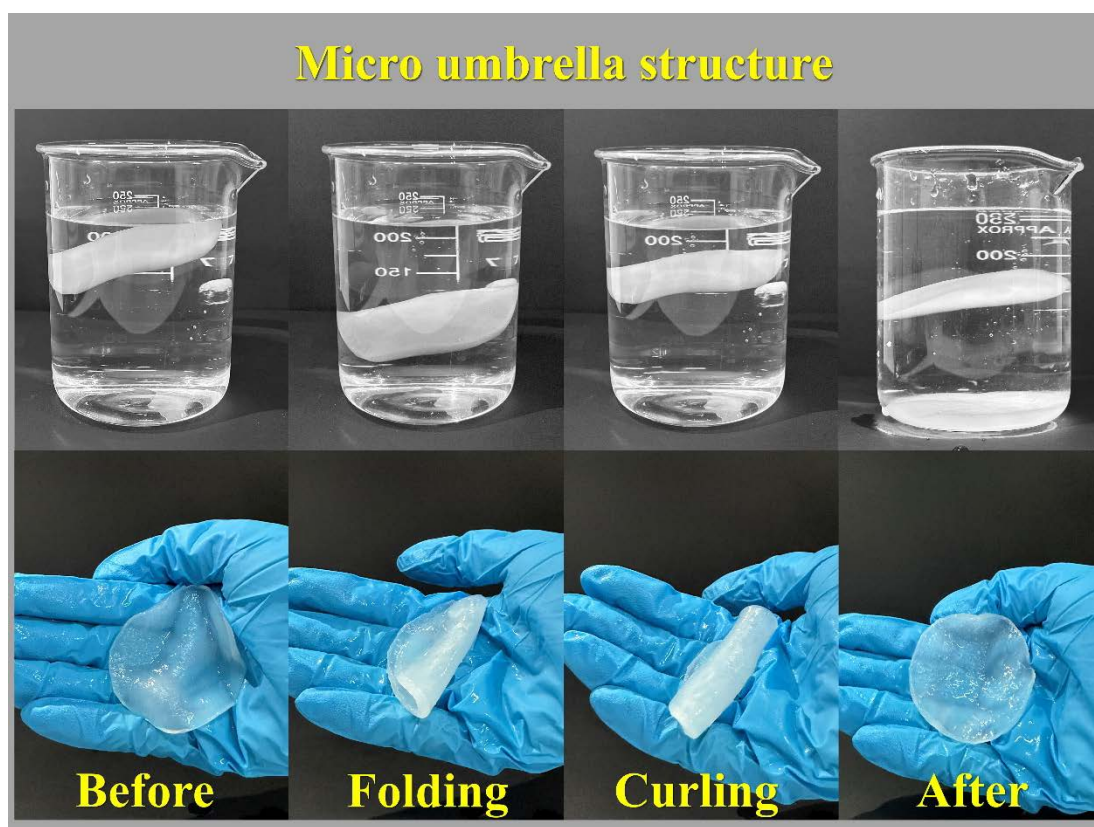


Fig. S1. Micro umbrella-shaped structure of 2%-SKP-Ca²⁺ filter.

2. Pore distribution tests

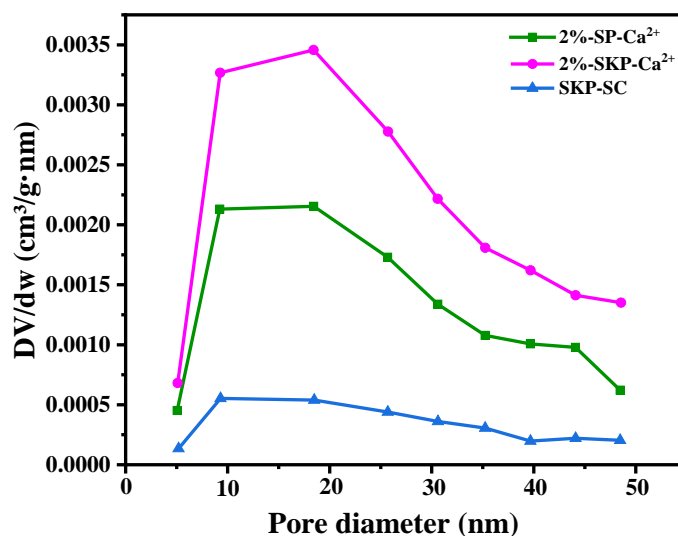


Fig. S2. Pore size distribution plots of 2%-SP-Ca²⁺, 2%-SKP-Ca²⁺, SKP-SC.

3. XRD results

Fig. S3 reveals the XRD patterns of powder samples SA, k-CG, PVA and hydrogel samples 2%-SP-Ca²⁺, SKP-SC and 2%-SKP-Ca²⁺. The diffraction pattern of PVA shows weak diffraction peak and strong diffraction peak at 2θ of 11.5° and 19.63° , respectively (Niu et al., 2020; Wang et al., 2018). The diffraction pattern of SA reveals a peak at about $2\theta=13.5^\circ$. The wide peak shape may be due to the amorphous structure of SA, resulting in poor crystallinity of the sample and easy to show a wide dispersion peak. For k-CG, it shows strong peaks at $2\theta=11.8^\circ$, 12.9° , 14.66° , 18.57° , 20.85° and 28.53° , respectively, which means that k-CG has good crystallinity. However, the addition of SA leads to the disappearance of the structural characteristic peak of PVA, which can be inferred that there is a strong hydrogen bond between SA and PVA, which may be attributed to the hydroxyl group -OH on the surface of PVA (Melby et al., 2016). Moreover, we did not observe obvious diffraction peaks in the graph of 2%-SKP-Ca²⁺ hydrogel. This is mainly attribute to the physical cross-linking between PVA and SA well as the intermolecular and intramolecular crystalline regions and hydrogen bonds of k-CG, which leads to the disappearance of the characteristic peaks of the three substances. It is worth noting that we also did not observe the characteristic peak of SKP-SC hydrogel, which is mainly due to the cross-linking of

sodium citrate and the synergistic effect with SA. However, the addition of a large amount of Na^+ to k-CG will interfere with its gelation and affect the structural characteristics of the material. Brittle changes occur, which leads to the disappearance of characteristic peaks.

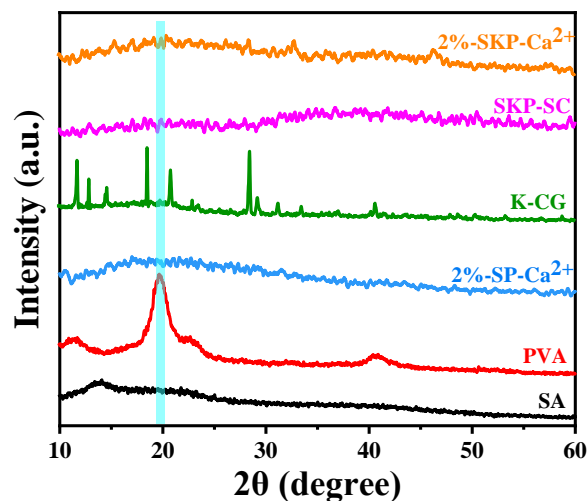


Fig. S3. XRD patterns of powder samples and hydrogel samples.

4. mechanism diagram of the triple crosslinking method

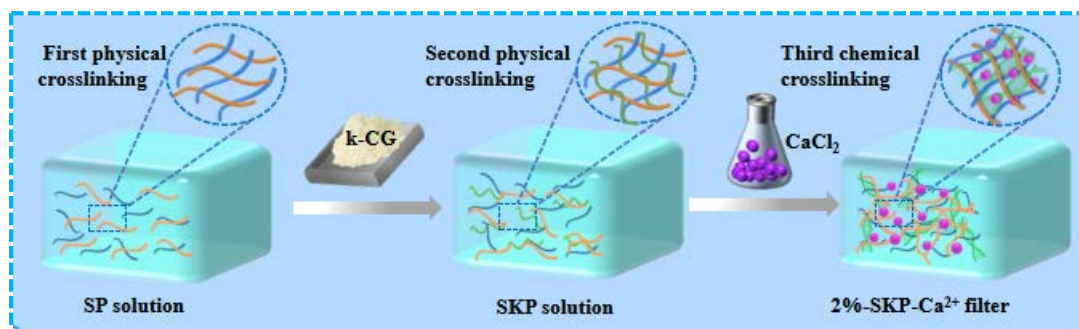


Fig. S4. mechanism diagram of the triple crosslinking method.

5. Surface wettability tests

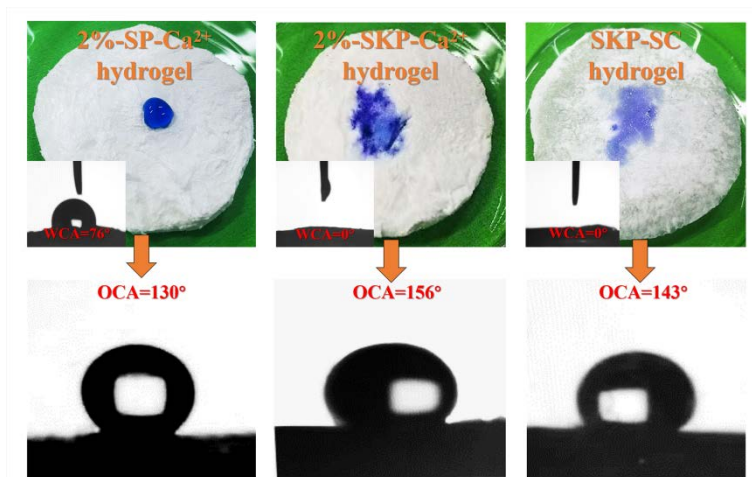


Fig. S5. Comparison diagram of water contact angle and oil contact angle of three hydrogels.

6. Application of multi-substrate anti-oil pollution coating samples in complex environment.

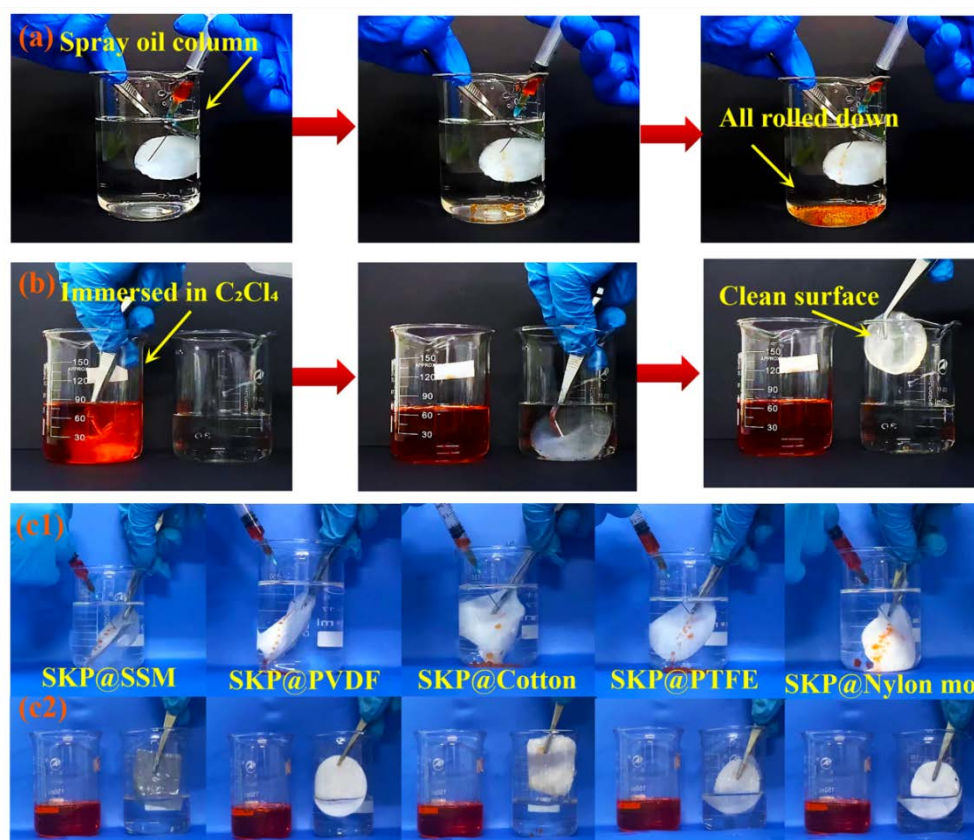


Fig. S6. (a) Oil adhesion resistance test. (b) Self-cleaning performance test. (c1, c2) Anti-oil fouling performance test of multi-base hydrogel coating hybrid membrane.

7. Durability tests

Considering the actual complex oily wastewater environment, it is necessary to evaluate the corrosion resistance of the separation material. The salt tolerance and acid/alkali resistance of 2%-SKP-Ca²⁺ filter is displayed in Fig. S7. After soaking in NaCl and MgSO₄ solutions with different mass fractions for 24 h, the surface super-wettability and separation efficiency of the 2%-SKP-Ca²⁺ filter are highly stable, as shown in Fig. S7a and b. The underwater oil contact angle is still greater than 152.6°, and the oil-water separation efficiency is greater than 98.2%, indicating that the filter material has good salt corrosion resistance. The filter was placed in different pH solutions for 1.0 d and its stability was studied. From Fig. S7c, it is evident that the contact angle of underwater oil exhibits minimal decrease in highly acidic or alkaline environments. Over the entire pH range, all underwater oil contact angles are higher than 151.8°. Otherwise, the effect of 50 times of cleaning on water flux was also studied, and the overall water flux remained above 11000 L m⁻² h⁻¹ (Fig. S7d). The above results confirm that the 2%-SKP-Ca²⁺ filter has good chemical stability (salt/acid/alkali resistance) and can be used as a multi-substrate hydrogel coating material in complex oily wastewater treatment.

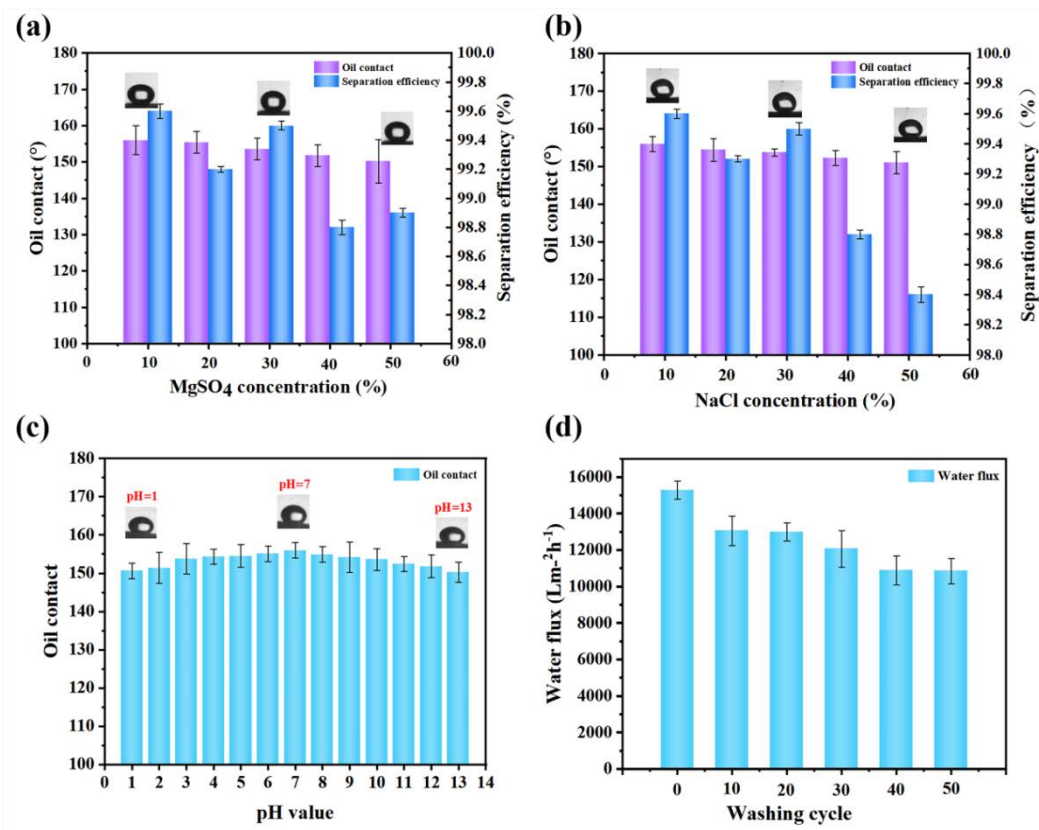


Fig. S7. (a, b) Effects of MgSO₄ and NaCl salt solution on the underwater oil contact angle and separation efficiency of 2%-SKP-Ca²⁺ filter was studied. (c) Effects of different pH value solutions on the contact angle and separation efficiency of 2%-SKP-Ca²⁺ filter underwater oil. (d) Effect of cleaning 50 times on water flux.

8. Optical microscope images of emulsified oil separation at different multiples

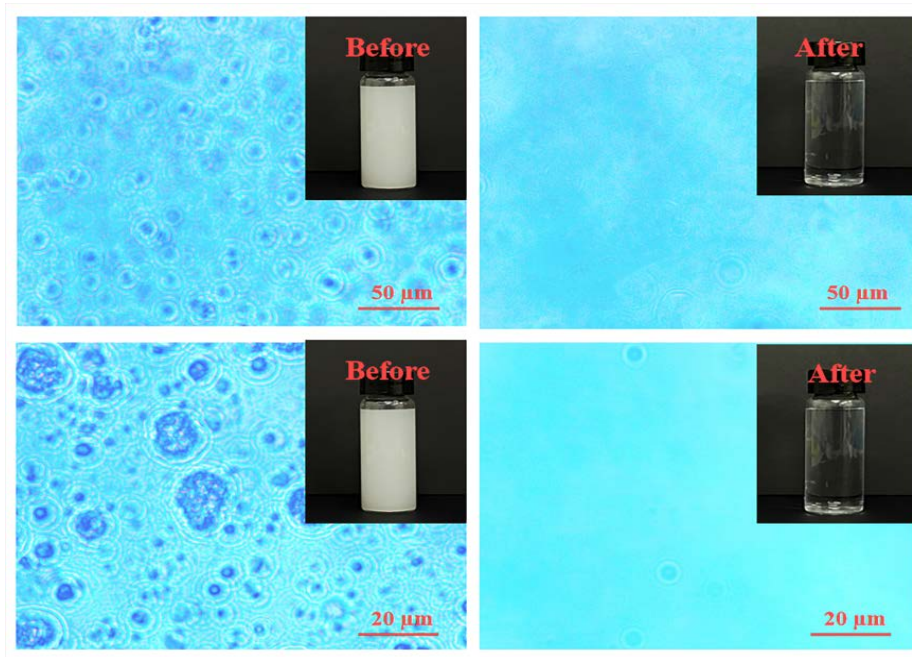


Fig. S8. Optical microscopy of emulsified oil separation at high magnification.

9. Separation efficiency and water flux of MB-containing emulsified oil

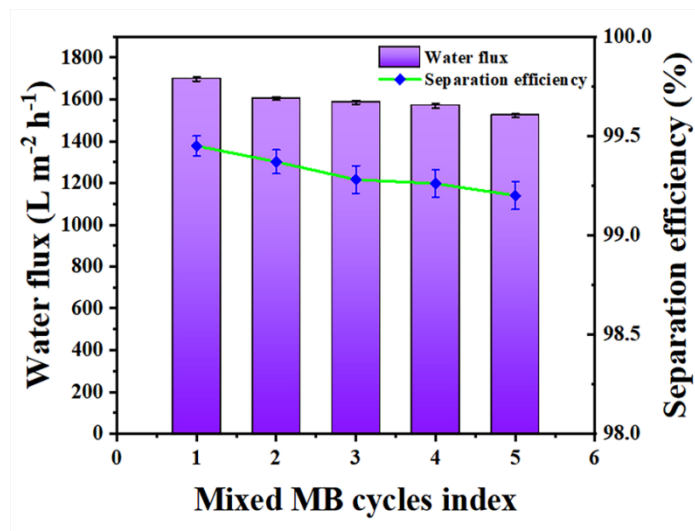


Fig. S9. Separation efficiency and water flux of emulsified oil containing MB.

Table S3. Comparison of the purification performance of the 2%-SKP-Ca²⁺ filter and other advanced membranes for wastewater.

Type	Environmental hazard	Functionality	Water flux (L m ⁻² h ⁻¹)	Ref.
MXene composite Membrane	Middle	99.5% (Oil/Water emulsion)	500.4	(Hu, 2021)
PVDF@TP membrane	None	>99% (Oil/Water emulsion)	955-1672	(Xu, 2023)
Composite MnO fly ash	Middle	>95% (MB degradability)	/	(Anggraini et al., 2023)
Pulp/CNF-CeO₂-STA	Small	>80% (Oil/Water emulsion) 94.3% (MB degradability)	60	(Yin et al., 2023)
Co-ZnO@SSM	Middle	>90% (Oil/Water emulsion) >75% (MB degradability)	/	(Wu et al., 2023)
2%-SKP-Ca²⁺ filter	None	>99.6% (Oil/Water emulsion) >80% (MB degradability)	1698	This work

"/" means not mentioned in the article.

Video

S1 2%-SP-Ca²⁺ hydrogel underwater oil adhesion test

S2 2%-SKP-Ca²⁺ hydrogel filter underwater oil adhesion test

S3 SKP-SC hydrogel underwater oil adhesion test

S4 Oil-water separation performance of 2%-SKP-Ca²⁺ hydrogel filter

References

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