Supporting Information

Mussel and Fish Scale-Inspired Underwater Superoleophobic Kapok Membranes for Continuous and Simultaneous Removal of Oils and Water-Soluble Dyes in Water

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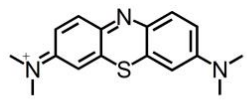
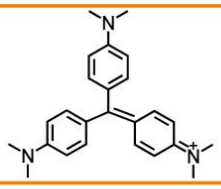
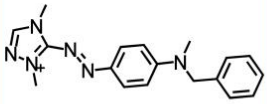
Abbr.	Structural formula	λ_{max} (nm)	Conc. (ppm)	Toxicity
MB		664		✓ damage the environment ✓ be toxic to aquatic life ✓ possess carcinogenicity
MV		591		✓ be carcinogens ✓ be toxic to mammals
CR		540		✓ be toxic to aquatic life

Fig. S1. Information of MB, MV and CR, and the photos of their aqueous solutions of different concentrations.

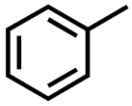
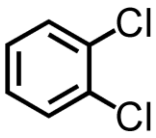
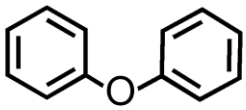
Name	Structural formula	d (g/cm^3)	λ_{max} (nm)	Toxicity
toluene		0.866	269	✓ intrauterine growth retardation ✓ premature delivery ✓ congenital malformations ✓ postnatal developmental retardation ✓ carcinogenesis
1,2-dichloro-benzene		1.30	277	✓ be highly toxic to liver, kidney and thyroid ✓ have long-term adverse effects in aquatic environment
diphenyl oxide		1.075	279	✓ be potential hazards to the aquatic environment

Fig. S2. Informations of toluene, 1, 2-dichlorobenzene and diphenyl oxide.

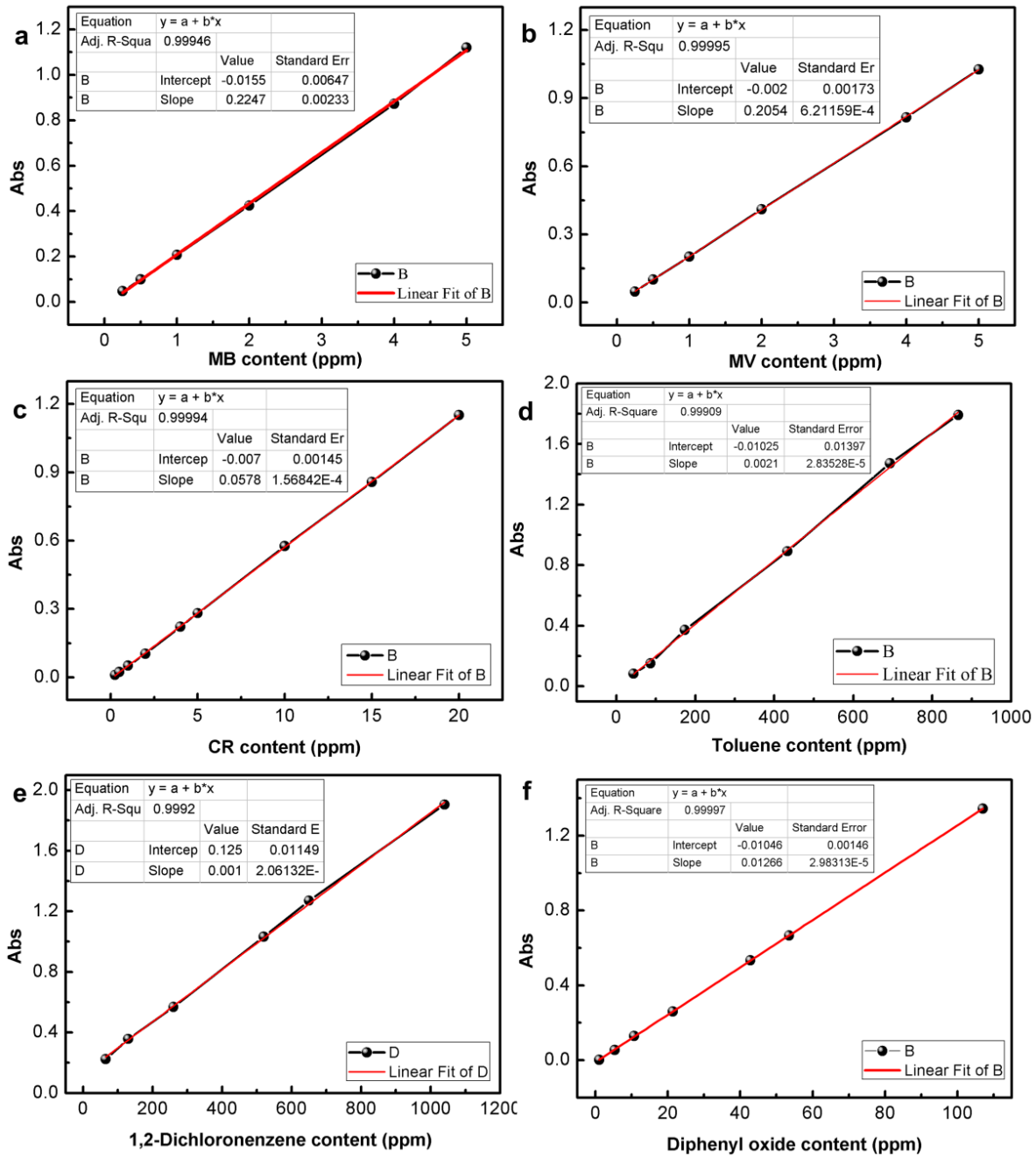


Fig. S3. Calibration curves of the model water-soluble dyes and oils. The solvent for preparing the calibration curves is methanol/water (v/v = 1/3). The correlation coefficients R^2 are more than 0.999.

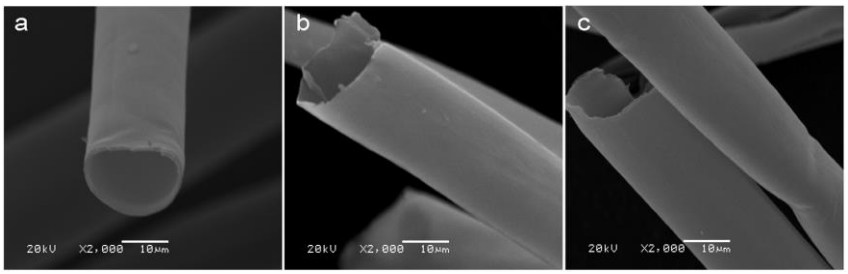


Fig. S4. SEM images of (a) KFs, (b) SC-KFs and (c) SC-KFs@PDA. KFs were treated with SC for 4 h followed by modification with PDA for 24 h.

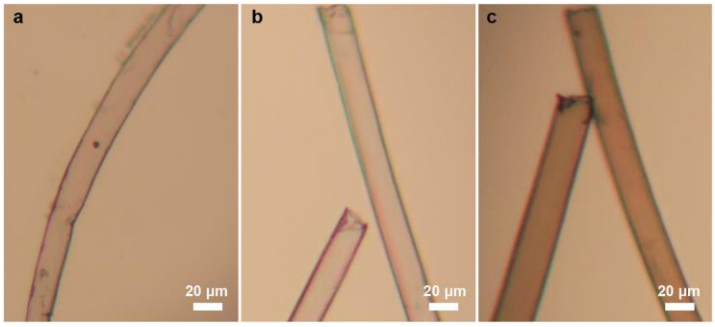


Fig. S5. Micrographs of (a) KFs, (b) SC-KFs and (c) SC-KFs@PDA. KFs were treated with SC for 4 h followed by modification with PDA for 24 h.

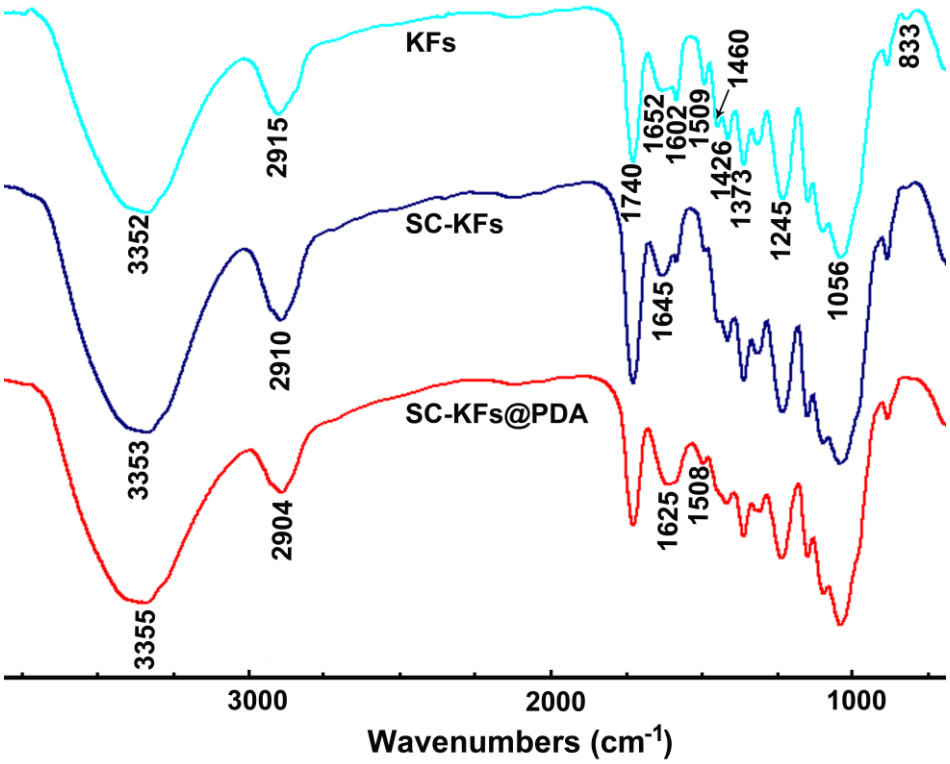


Fig. S6. FTIR spectra of KFs, SC-KFs and SC-KFs@PDA. KFs were treated with SC for 4 h followed by modification with PDA for 24 h.

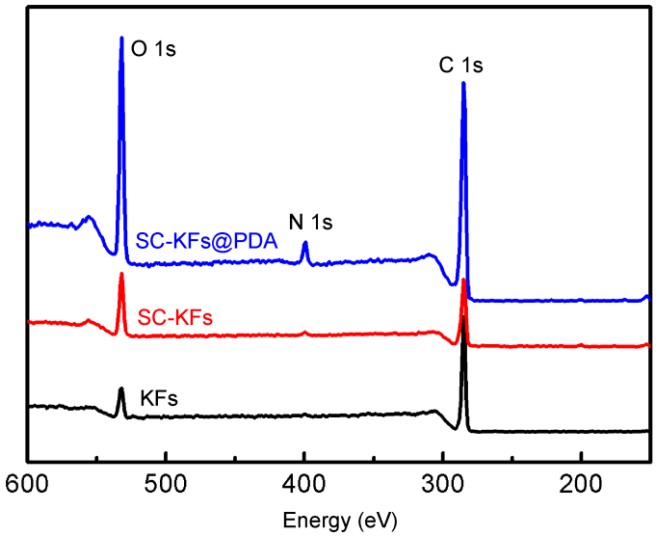


Fig. S7. (a) XPS survey spectra of KFs, SC-KFs and SC-KFs@PDA. KFs were treated with SC for 4 h followed by modification with PDA for 24 h.

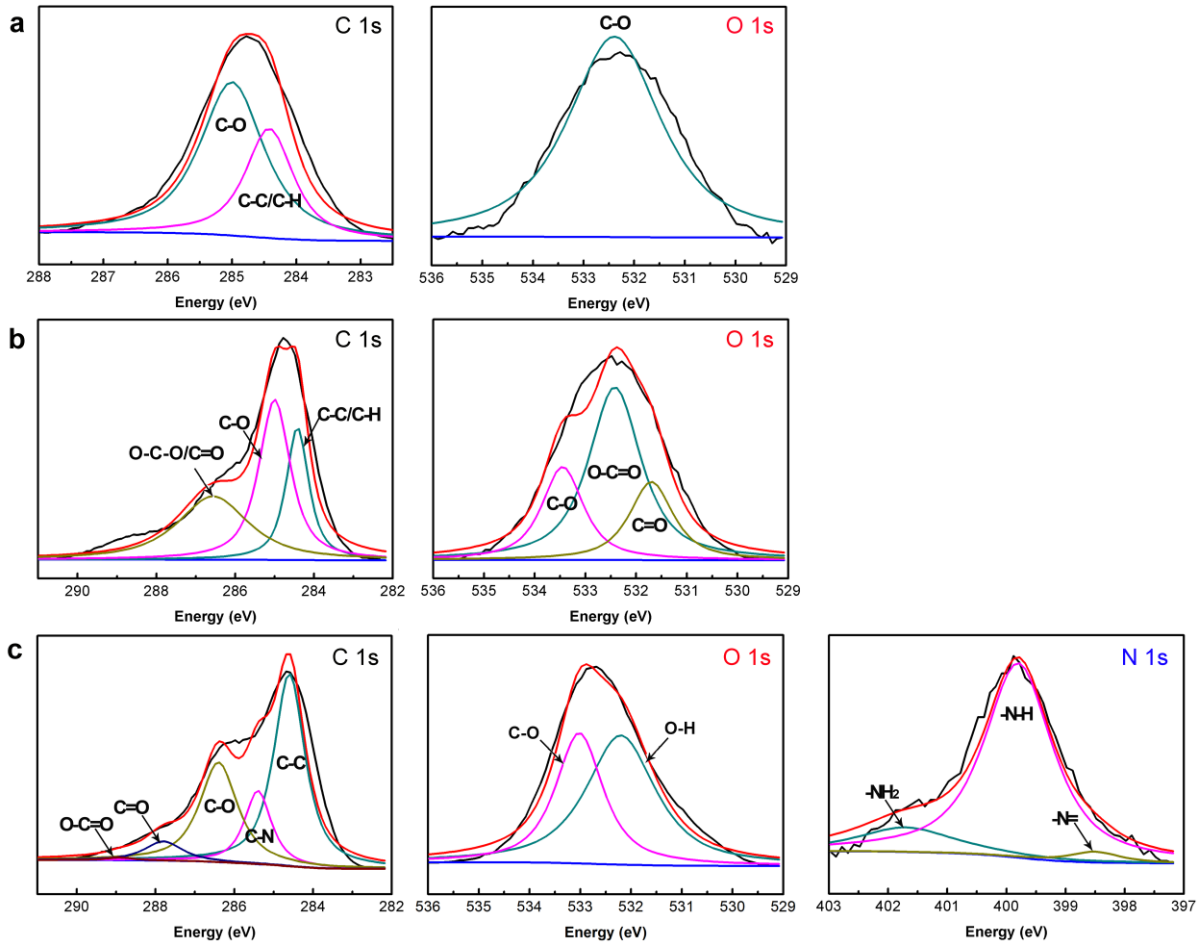


Fig. S8. High-resolution XPS spectra of (a) C 1s and O 1s of KFs, (b) C 1s and O 1s of SC-KFs, and (c) C 1s, O 1s and N 1s of SC-KFs@PDA. KFs were treated with SC for 4 h followed by modification with PDA for 24 h.

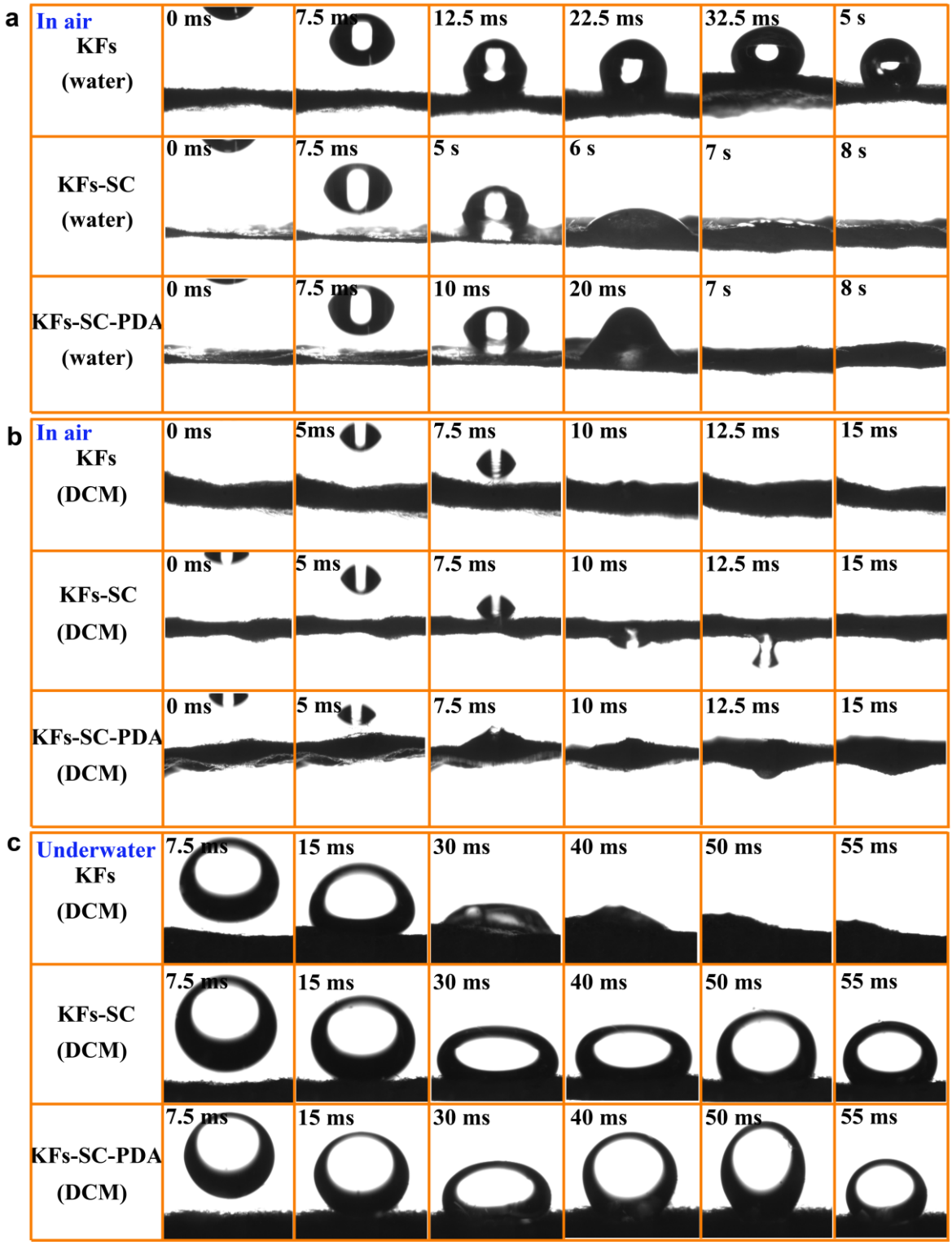


Fig. S9. Snapshots of (a) 7 μL water drops in air, (c) 7 μL DCM drops in air and (d) 7 μL DCM drops underwater on the KFs, SC-KFs and SC-KFs@PDA membranes taken using Contact Angle System OCA 20. KFs were treated with SC for 4 h followed by modification with PDA for 24 h.

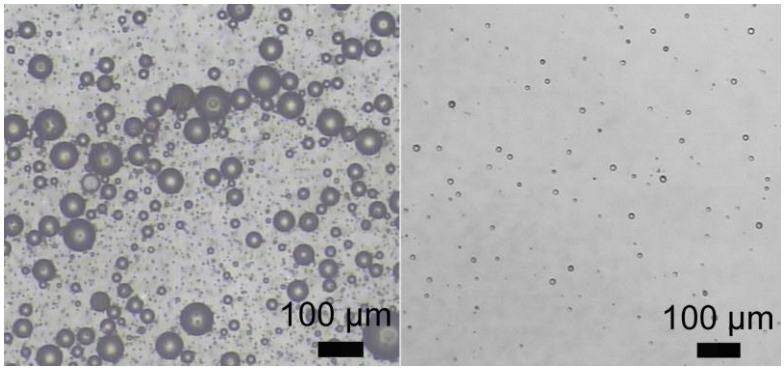


Fig. S10. Micrographs of the (a) surfactant free toluene-in-water emulsion and (b) tween 80 stabilized toluene-in-water emulsion.

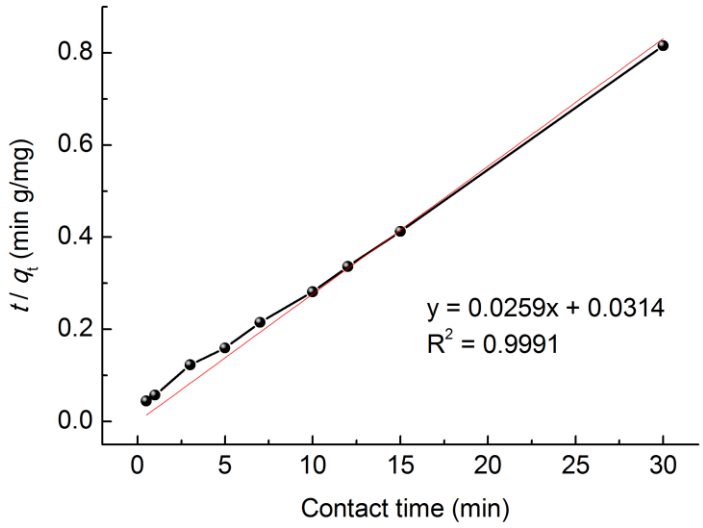


Fig. S11. Pseudo-second-order model for the adsorption of MB by SC_{4h}-KFs@PDA_{24h}.

The pseudo-second-order rate equation is given as [1]:

$$\frac{t}{q_t} = \frac{1}{kq_e^2} + \frac{t}{q_e} \tag{S3}$$

where q_e and q_t are the adsorption capacity (mg/g) at equilibrium and at time t (min), and k (g/(mg·min)) is the adsorption rate constant of pseudo-second-order adsorption rate.

Fig. S8 shows the plot of t/q_t versus t. The value of q_e is obtained from the slope of the plot and k is obtained from the intercept. The $q_{e,exp}$ (36.80 mg/g) and the $q_{e,cal}$ (38.61 mg/g) values from the pseudo-second-order model are very close to each other. In addition, the linear R^2 coefficient is 0.9991. These results indicate that the adsorption of MB by SC-KFs@PDA follows the pseudo-second-order model.

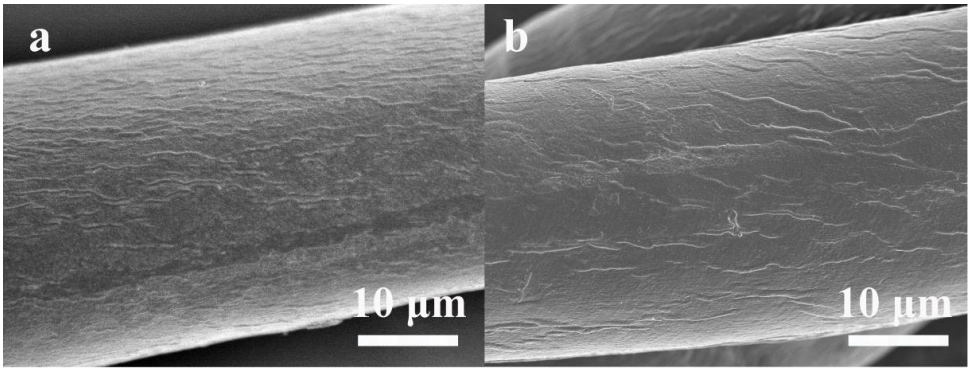


Fig. S12. SEM images of (a) SC_{4h}-KFs@PDA_{24h} and (b) after five adsorption-desorption cycles.

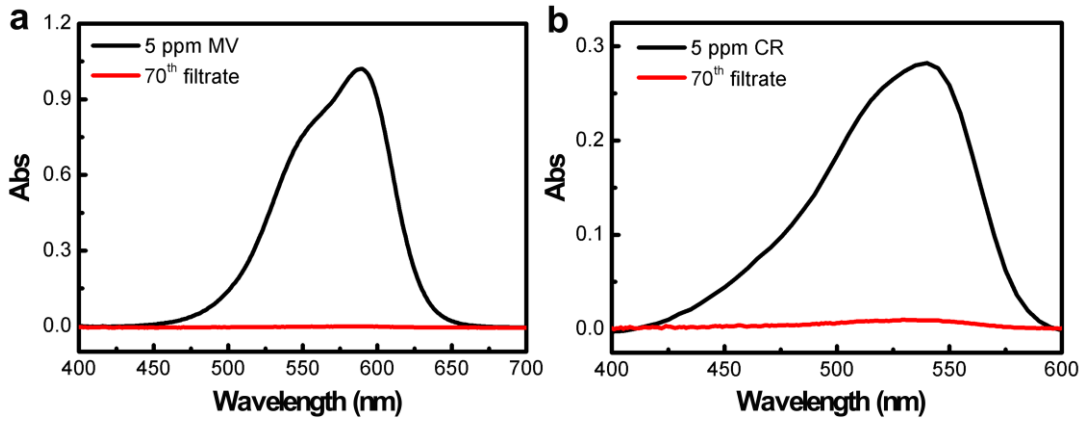


Fig. S13. UV-Vis spectra of the (a) 5 ppm MV solution and the 70th filtrate, and (b) 5 ppm CR solution and the 70th filtrate.

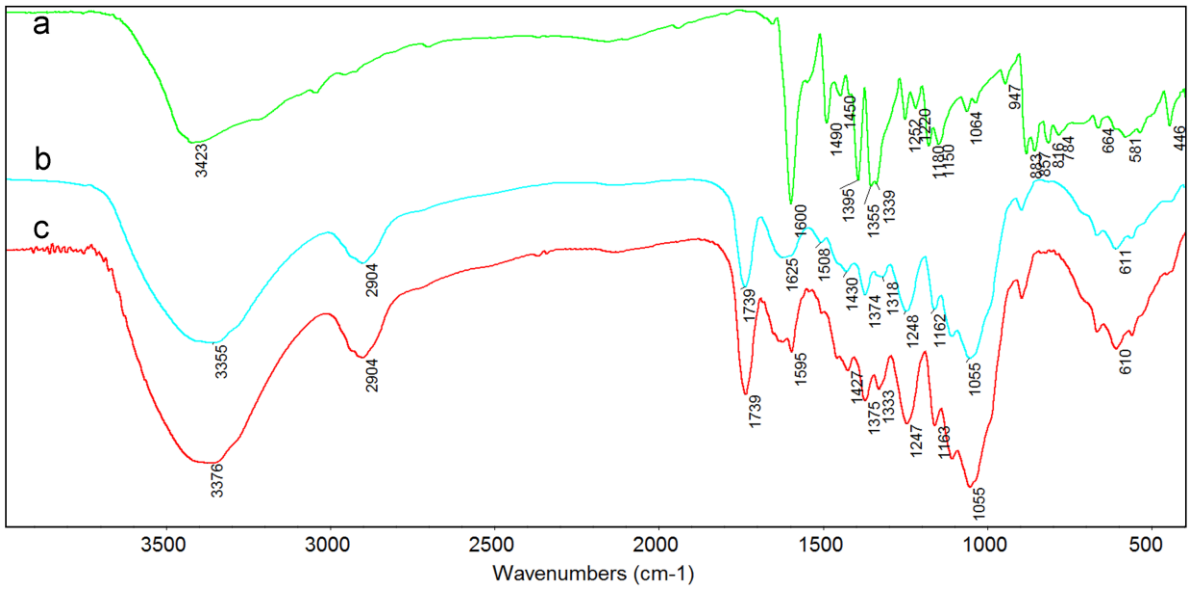


Fig. S14. FTIR spectra of (a) MB, (b) SC_{4h}-KFs@PDA_{24h} and (c) SC_{4h}-KFs@PDA_{24h} with adsorbed MB.

Table S1. Surface chemical composition and atomic ratios of KFs, SC-KFs and SC-KFs@PDA. KFs were treated with SC for 4 h followed by modification with PDA for 24 h.

Samples	Surface chemical composition (at.%)		
	C	O	N
KFs	89.3	10.7	-
SC-KFs	76.4	23.6	-
SC-KFs@PDA	70.8	25.5	3.7

Table S2. The zeta potentials of KFs, SC_{4h}-KFs@PDA_{24h} and SC_{4h}-KFs@PDA_{24h} with adsorbed MB.

Samples	KF	SC_{4h}-KFs@PDA_{24h}	SC_{4h}-KFs@PDA_{24h} with adsorbed MB
Zeta potential / mV	-30.5	-44.3	-17.9

Movie S1. Underwater wettability of the 10 º titled KFs, SC-KFs and SC-KFs@PDA membranes by DCM. KFs were treated with SC for 4 h followed by modification with PDA for 24 h. DCM was colored with Oil Red O. This video highlights the obviously different underwater wettability of the membranes towards oil.

Movie S2. Interaction of DCM droplets (7 μL) with the SC-KFs membrane (Part 1) and the SC-KFs@PDA membrane (Part 2) in water. KFs were treated with SC for 4 h followed by modification with PDA for 24 h. This video demonstrates the very weak interaction between the DCM droplet and the SC-KFs@PDA membrane in water.

References

[1] Y. S. Ho, G. McKay, *Process Biochem.* **1999**, *34*, 451.