

Supporting Information

Superhydrophobic Graphene-coated Sponge with microcavities for High Efficiency Oil-in-Water Emulsion Separation

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1. Images of different filtration materials

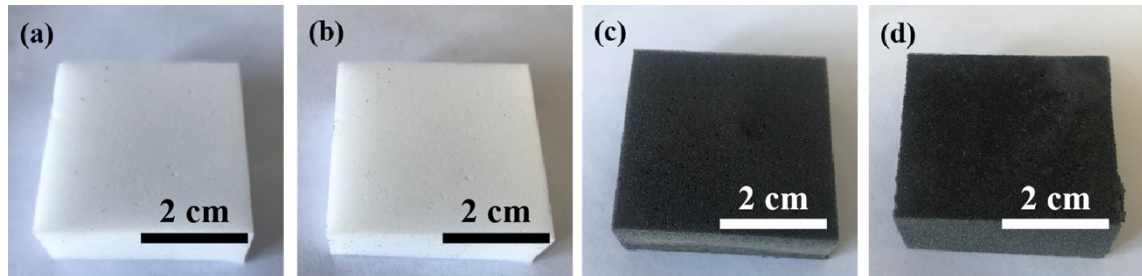


Fig. S1. Images of (a) MS, (b) P-MS, (c) G-MS, (d) PG-MS.

2. Separation of oil-in-water emulsion through the gravity.

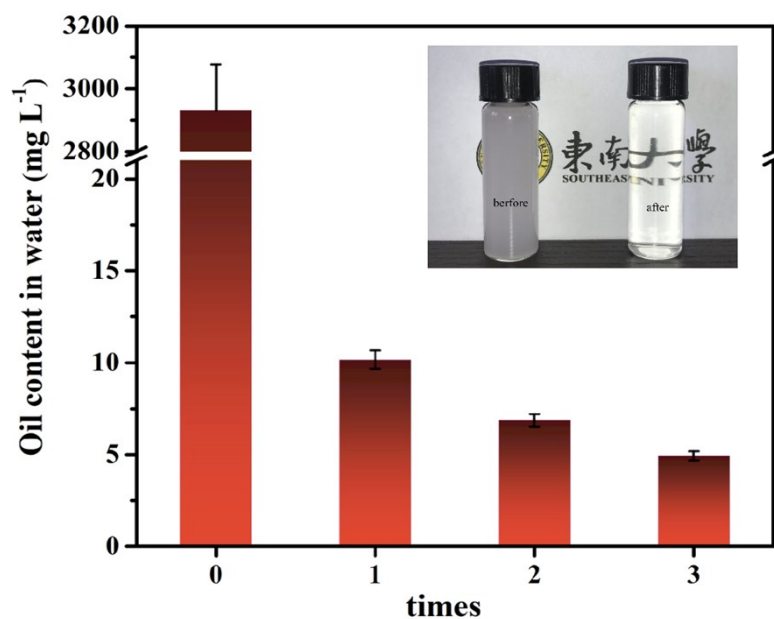


Fig. S2. The oil content of the emulsion after separation through gravity for different times and the optical photos of the emulsion before and after separation.

Fig. S2 shows the oil content and optical photo after the emulsion passes through PG-MS under gravity. In this experiment, the thickness of PG-MS was 5.4 cm, and the oil of the emulsion (with the initial oil content was 0.5%) was transmission oil. Through the photo of the water samples before and after filtration, it can be clearly seen that the light transmittance of the filtered water samples has been significantly improved. After separation for the first time by PG-MS, the oil content in the water was about 10 mg L⁻¹, and three times later, the oil content in the water sample was only 4.93 mg L⁻¹. Both of them show that the separation by PG-MS has a good separation effect for oil-in-water emulsion under gravity.

3. Turbidity analysis of water samples after separation with different thickness of filtration materials and flow rates

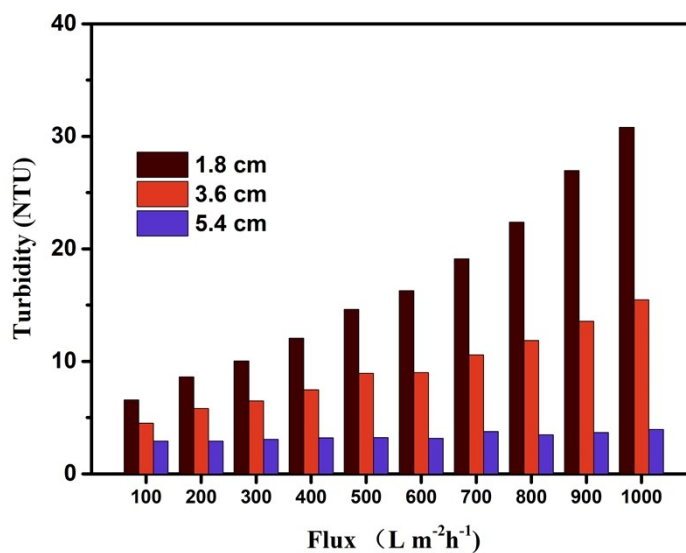


Fig. S3. The turbidity of the emulsion separation with different thickness of filtration materials and flow rates.

At a filtration flux of 100 L m⁻² h⁻¹, the turbidity of the filtered water is 6.58, 4.51, and 2.91 NTU, when the PG-MS thickness is 1.8 cm, 3.6 cm, and 5.4 cm, respectively. When the thickness of the filtration layer is constant, the turbidity of the water sample increases with the increase of the flow rate, that is, the separation efficiency decreases continuously. When the flux is 1000 L m⁻² h⁻¹, and the thickness of the PG-MS is 1.8 cm, 3.6 cm, and 5.4 cm, respectively, the turbidity of the filtered water is 30.8 NTU, 15.47 NTU, and 3.96 NTU.

4. Analysis of separation efficiency under different initial oil contents and different filtration flux.



Fig. S4. Images of emulsions with different initial oil content before and after separation using PG-MS.

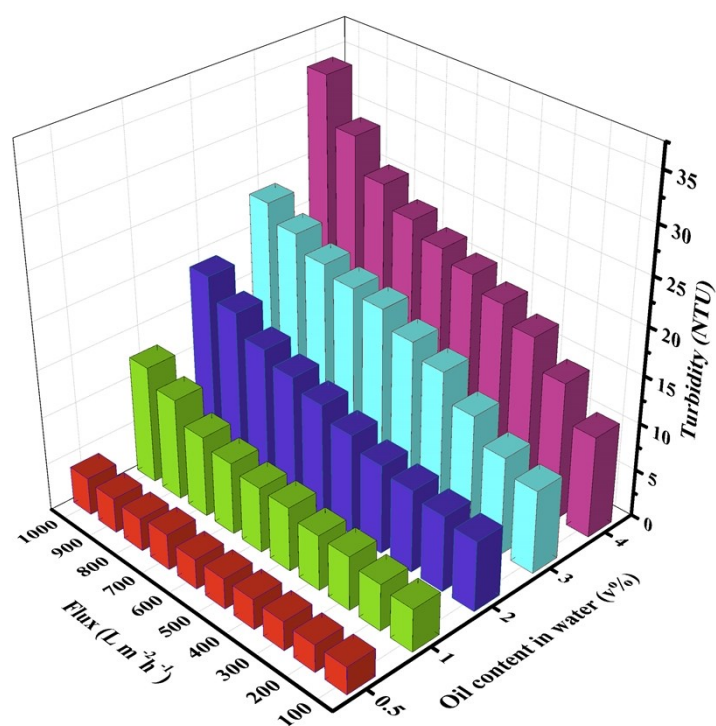


Fig. S5. The turbidities of filtrate after PG-MS filtration of emulsion with different oil content is in different flux.

Figure S5 shows the trend of turbidity change caused by the increase of filtration flow rates (from $100 \text{ L m}^{-2} \text{ h}^{-1}$ to $1000 \text{ L m}^{-2} \text{ h}^{-1}$) for emulsion with different initial oil contents. When the initial oil content is constant, turbidity increases with the increase of filtration flux. When the initial oil content is 0.5%, the turbidity of the filtered water basically stays between 2.91 and 3.94 NTU. However, when the initial oil content was 4%, the turbidity of the filtered water sample changed significantly, from 10.71 to 34.26 NTU.

Comparison of the separation materials for oil-in-water emulsion

Table S1. Comparison of the separation materials for oil-in-water emulsion

	Membranes or 3D materials	Super-hydrophobic	hydrophilic	Pressure (Pa)	Flux (L m ⁻² h ⁻¹)	separation efficiency	References
1	3D material	√		gravity	7 500	>95%	1
2	3D material		√	2 771	1 100	>99.2%	2
3	3D material	√		static adsorption	-	~93.8	3
4	3D material	√	√	static adsorption	-	>98%	4
5	Membrane		√	10 000	3 500	>99%	5
6	Membrane		√	40 000	~280	~99%	6
7	Membrane		√	9 000	405	>99%	7
8	Membrane		√	175 000	767	>99%	8
9	Membrane		√	100 000	2 270	>99%	9
10	3D material	√		gravity	10 000	>99%	This work

Supplementary Movies:

Mov. S1 A dynamic demonstration of droplets moving quickly through PG-MS with formed water channels inside.. The thickness of PG-MS is 1.8 cm.

Mov. S2 A dynamic demonstration of droplets moving quickly through PG-MS with formed water channels inside. The thickness of PG-MS is 5.4 cm.

Mov. S3 A dynamic demonstration of oil-in-water emulsion separation, in which the oil is transmission oil, the initial oil content is 0.5 % (volume ratio), the filter layer thickness is 5.4cm, the flux is $200 \text{ L m}^{-2} \text{ h}^{-1}$.

Mov. S4 A dynamic demonstration of oil-in-water emulsion separation, in which the oil is transmission oil, the initial oil content is 5 % (volume ratio), the filter layer thickness is about 20 cm, the flux is $10\,000 \text{ L m}^{-2} \text{ h}^{-1}$.

Mov. S5 A dynamic demonstration of desorption of oil adsorbed by PG-MS with the increase of filtration time, in which the oil is dodecane (stained with Sudan red 5B) , the initial oil content is 2 % (volume ratio), the filter layer thickness is 5.4 cm, the flux is $400 \text{ L m}^{-2} \text{ h}^{-1}$.

Supplementary References

1. R. Z. Xing, B. H. Yang, R. L. Huang, W. Qi, R. X. Su, B. P. Binks, and Z. M. He, *Langmuir*, 2019, **35**, 12799.
2. J. J. Yun, F. A. Khan and S. Baik, *ACS Appl. Mater. Interfaces*, 2017, **9**, 16694.
3. J. T. Wang, H. F. W, G. H. Geng, *Marine Pollution Bulletin*, 2018, 127, 108.
4. G. Y. Chen, Y. R. Cao, L. Ke, X. X. Ye, X. Huang, and B. Shi, *Ind. Eng. Chem. Res.*, 2018, **57**, 16442.
5. G. L. Cao, Y. G. Wang, C. Y. Wang and S. H. Ho, *J. Mater. Chem. A*, 2019, **7**, 11305
6. J. P. Chaudhary, N. Vadodariya, S. K. Nataraj, and R. Meena, *ACS Appl. Mater. Interfaces*, 2015, **7**, 24957.
7. J. W. Sun, H. C. Bi, S. Su, H. Y. Jia, X. Xie, L. T. Sun, *J. Membrane Sci.*, 2018, **553**, 131.
8. J. A. Prince, S. Bhuvana, V. Anbharasi, N. Ayyanar, K. V. K. Boodhoo, G. Singh, *Water Research*, 2016, **103**, 311.
9. F. Zhang, S. J. Gao, Y. Z. Zhu, J. Jin, *J. Membrane Sci.*, 2016, **513**, 67.