

Supporting Information

Advanced Hybrid Nanosheets Membranes with Stable Nanochannels for Ultrafast Molecular Separation

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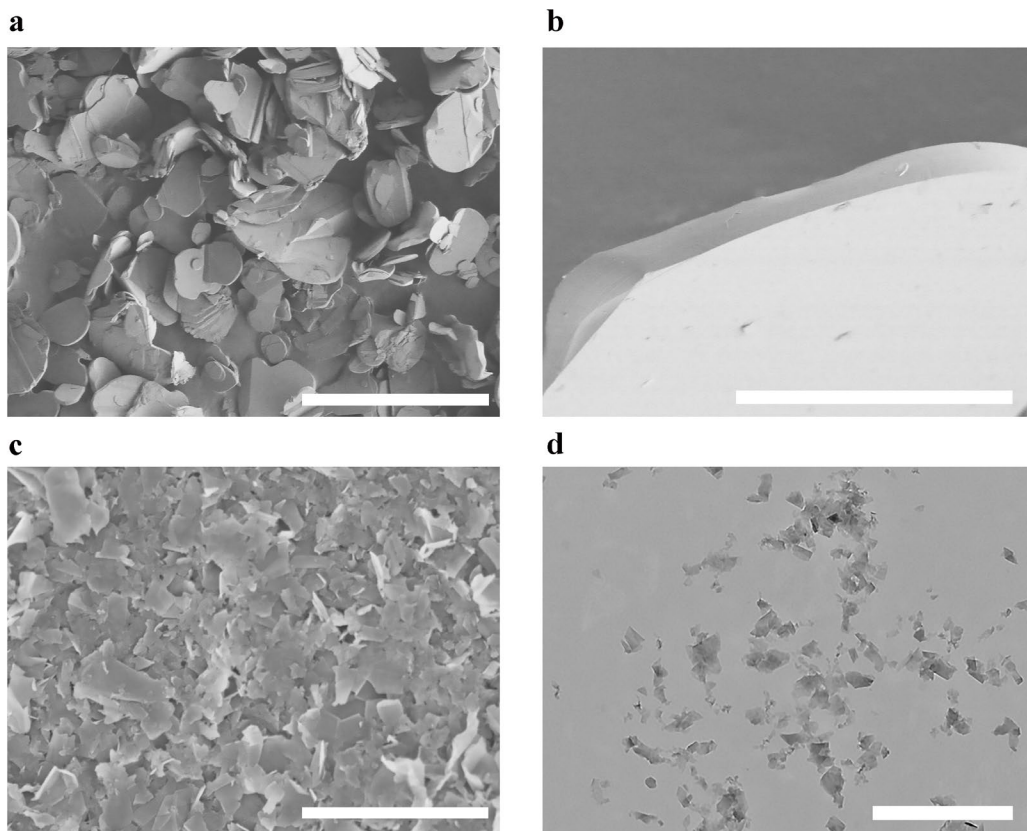
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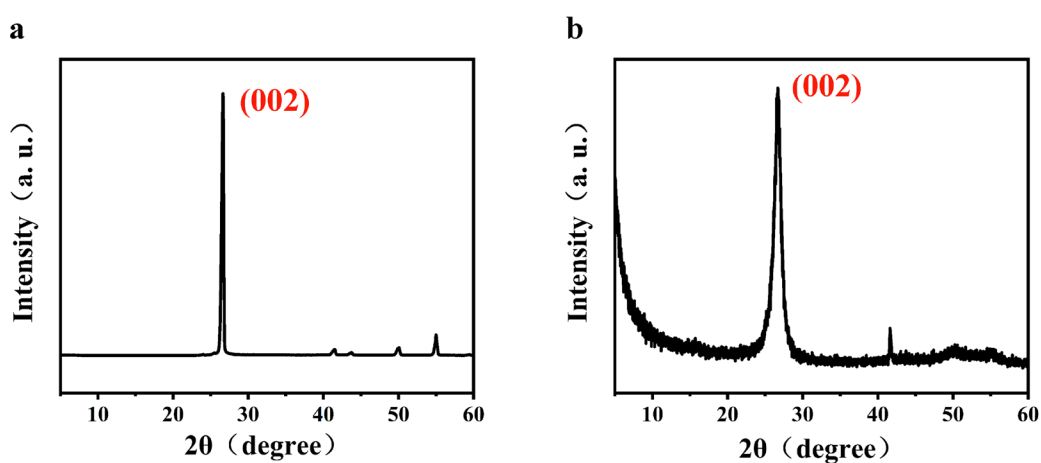
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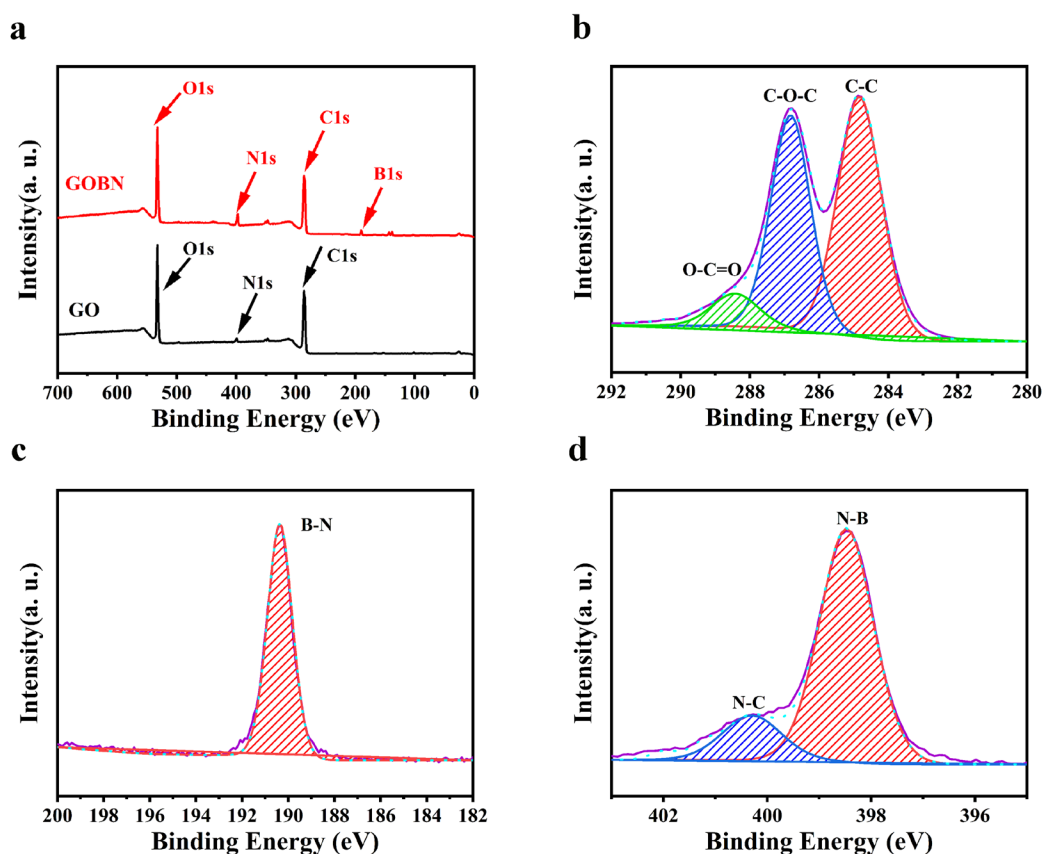
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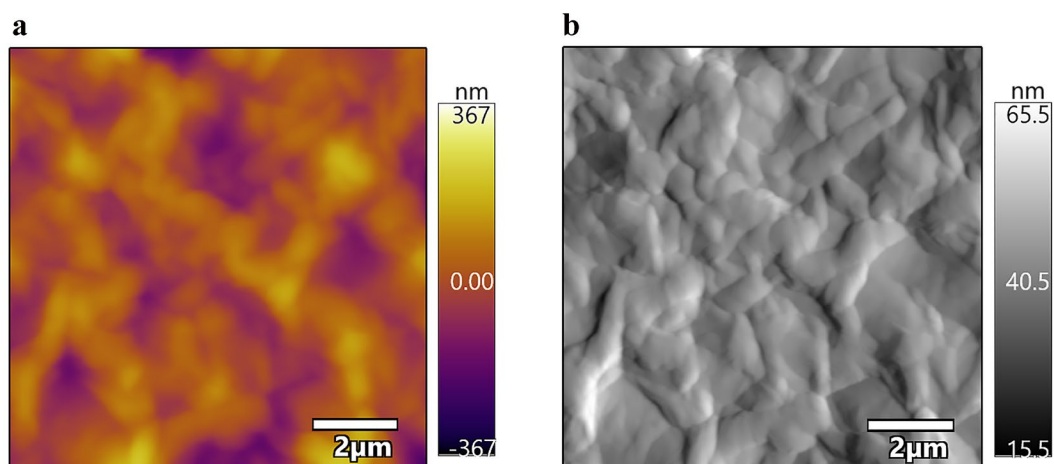
Supplementary Figure 1. (a) SEM image of the bulk BN powder. The scale bar is 100 μm . (b) SEM image of the thick BN. The scale bar is 10 μm . (c) SEM image of the exfoliated BN nanosheets. The scale bar is 2 μm . (d) TEM image of the BN nanosheets. From the SEM and TEM images, we can conclude that the exfoliated BN nanosheets are in the range of 50 nm to 500 nm. The scale bar is 1 μm .



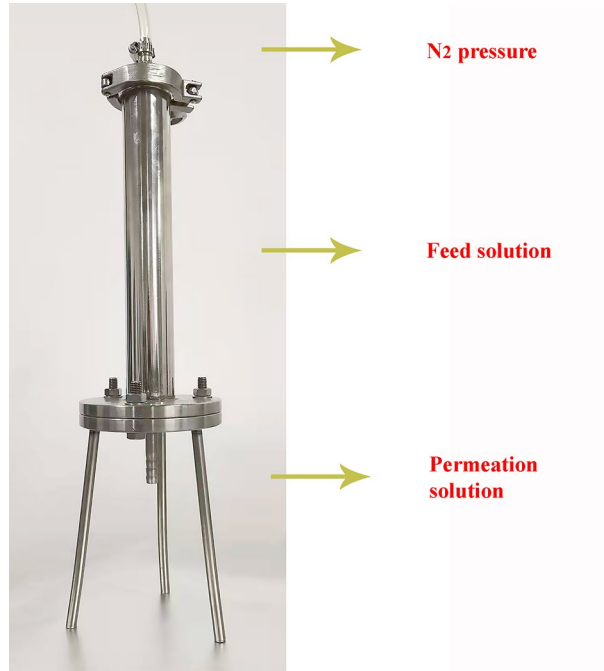
Supplementary Figure 2. (a) XRD pattern of the bulk BN powder. (b) XRD pattern of the exfoliated BN nanosheets. A clear broaden characteristic peak at 26.8° belongs to the BN of (002).



Supplementary Figure 3. (a) XPS survey GO and GOBN membranes. High-resolved XPS spectra of carbon, boron, and nitrogen of GOBN membrane (b, c, d). The boron element signal could be detected in the GOBN membrane and the interaction of the elements could be analyzed with the high-resolved spectra. This confirms the amino group on the BN nanosheets interacted well with the hydroxy and carboxyl of the GO nanosheets in the GOBN membrane.



Supplementary Figure 4. AFM image of the GOBN membrane. A rough surface image could be captured on the GOBN membrane.



Supplementary Figure 5. The dead-end filtration device for the membrane separation measurements.

Note. Concentration polarization is described by the relationship between the solvent flux J_v , the bulk concentration C_B , the concentration at the wall of the membrane C_M , the mass transfer coefficient K , and the concentration in the permeate C_P as:

$$J_v = K \ln((C_M - C_P)/(C_B - C_P)) \quad (1)$$

and the flux is described by:

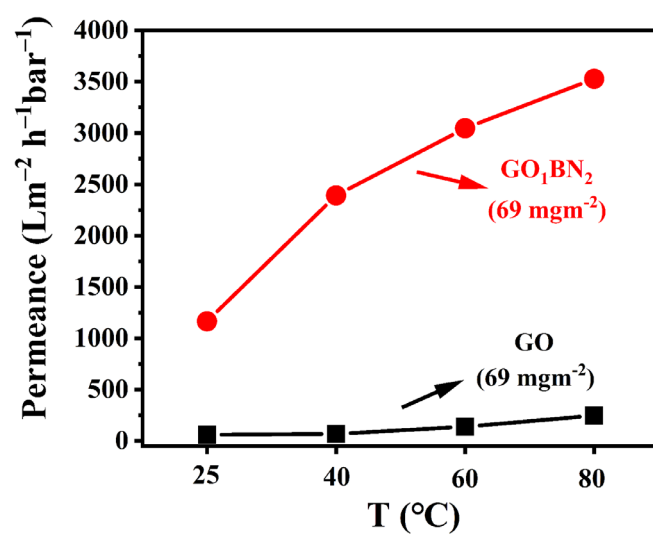
$$J_v = B (AP - OP) \quad (2)$$

where B = pure solvent permeance, AP = Applied Pressure and OP = Osmotic Pressure, and

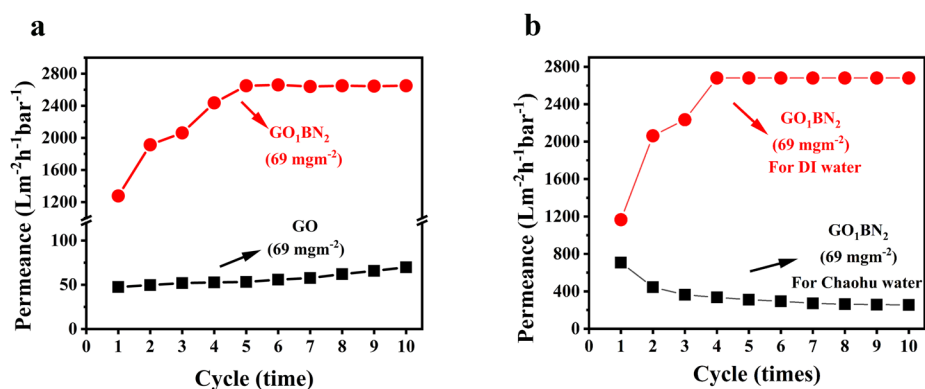
$$OP(\text{from Van't Hoff equation}) = c_M R T \quad (3)$$

Therefore, there is an implicit equation in J_v governing flux when c_P is small relative to c_M and c_F

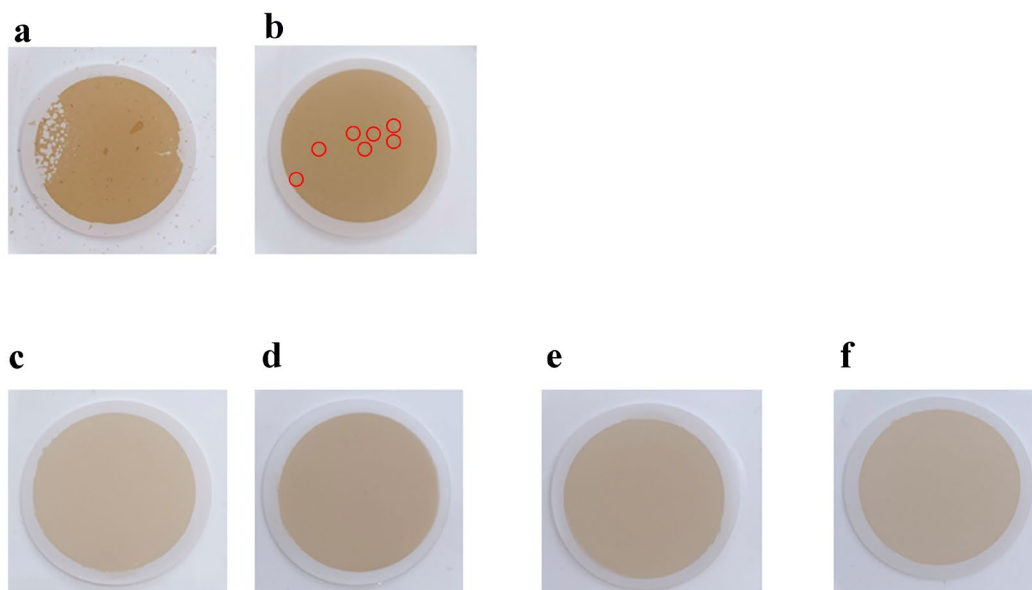
$$J_v - B(AP - C_M R \exp(J_v/K)) = 0 \quad (4)$$



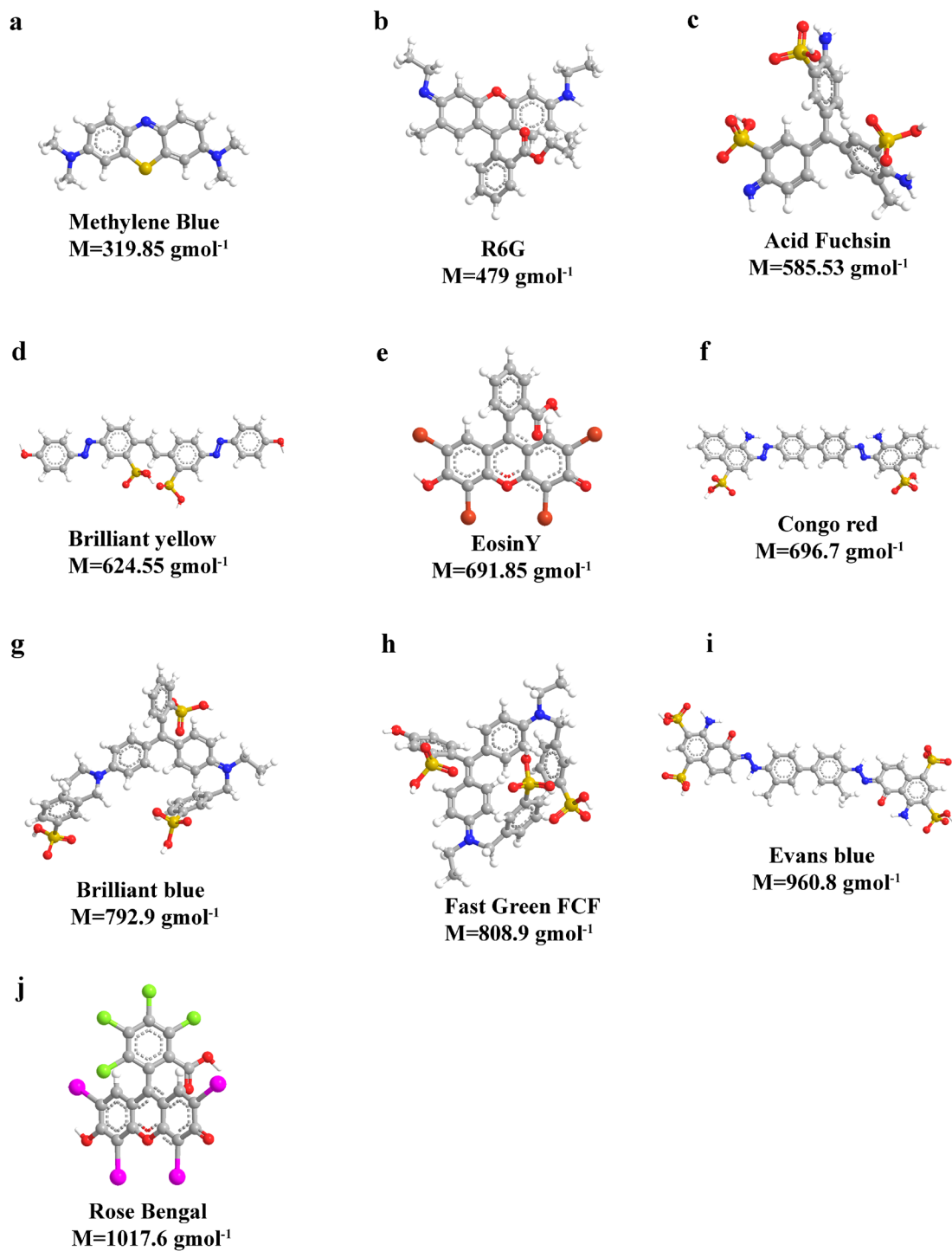
Supplementary Figure 6. The permeance of the membrane versus the temperature of the aqueous.



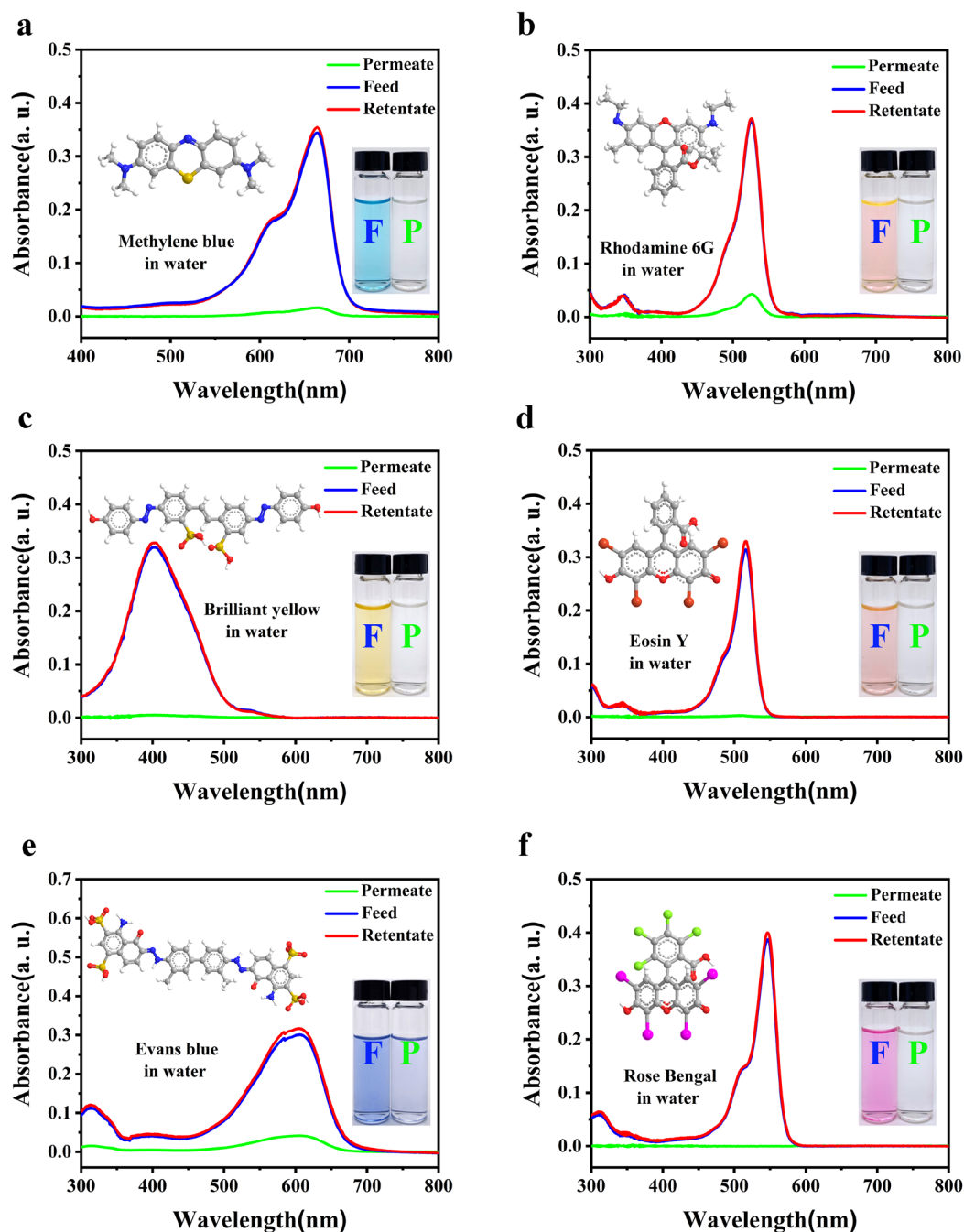
Supplementary Figure 7. The permeance of membrane versus the 10 times cycle filtration of water. (a) Comparisons of the permeance of GO₁BN₂ membrane and GO membrane for DI water. (b) Comparisons of the permeance of GO₁BN₂ membrane versus the 10 times cycle filtration of DI water and real water from Chaohu Lake.



Supplementary Figure 8. Photograph of the membranes with a mass density of 69 mgm^{-2} soaked in various solvents for 24 hours. GO membrane in water (a) and ethanol (b); GOBN membrane in water (c), ethanol (d), 0.1 mol/L HCl (e) and 0.1 mol/L NaOH (f), respectively. The incorporation of BN nanosheets into the GO membrane obviously stabilizes the membrane.



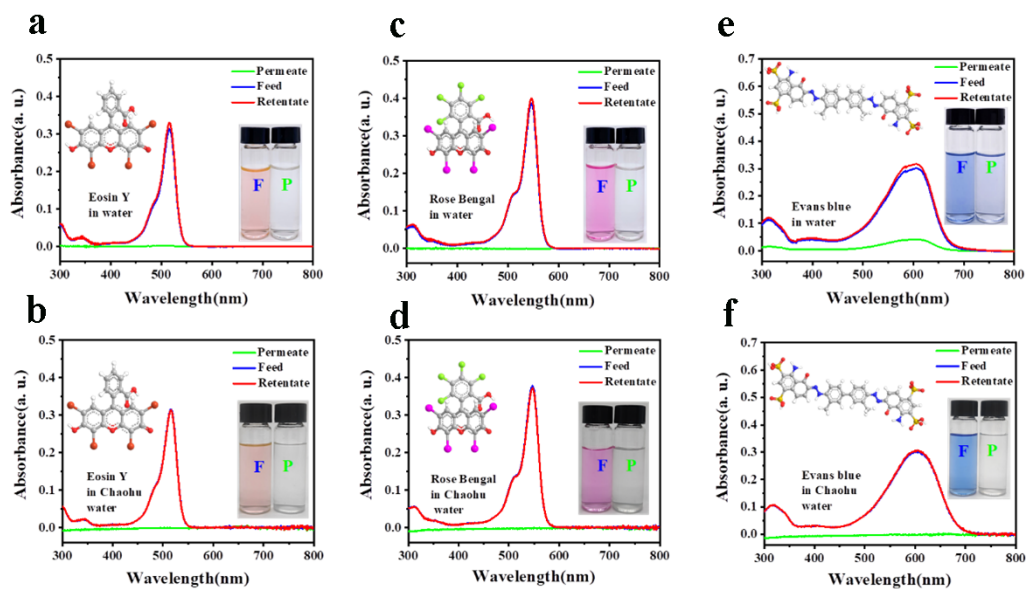
Supplementary Figure 9. The chemical structures of the dyes with the molecular weight were used for the molecular separation experiments.



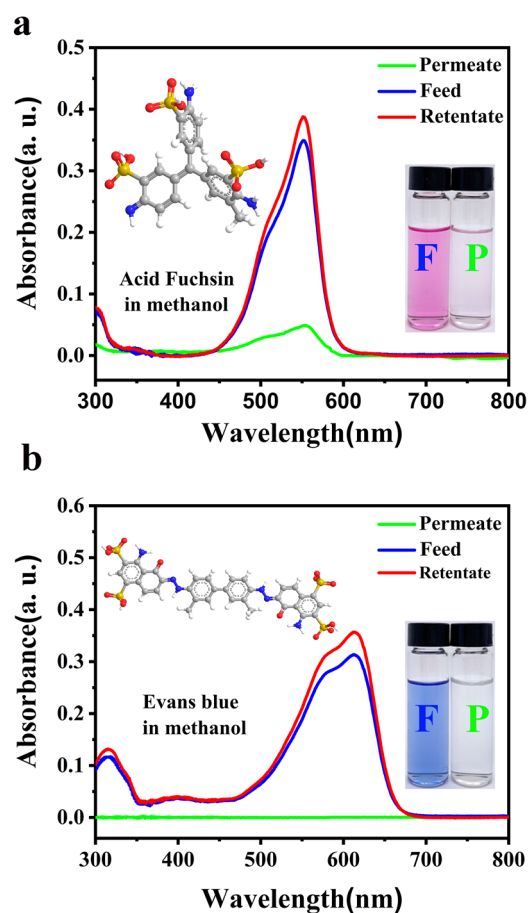
Supplementary Figure 10. UV-Vis absorption spectra of dyes before and after filtration through the fresh GOBN membrane in aqueous. (a) Methylene blue; (b) R6G; (c) Brilliant yellow; (d) Eosin Y; (e) Evans blue; (f) Rose Bengal. The left inset is the molecular structure of tested dyes and the right inset photo shows the feed (F, left) and permeate (P, right) of the dyes solution in water.

Methylene blue ($319.85 \text{ g mol}^{-1}$, concentration is 1.57 mg L^{-1}), Rhodamine 6G (479 g mol^{-1} , concentration is 1.92 mg L^{-1}), Brilliant yellow ($624.55 \text{ g mol}^{-1}$, concentration is 4 mg L^{-1}), Eosin Y ($691.85 \text{ g mol}^{-1}$, 2.7 mg L^{-1}), Evans blue (960 g mol^{-1} , concentration is 3.63 mg L^{-1}), Rose Bengal ($1017.6 \text{ g mol}^{-1}$, concentration is 4.54 mg L^{-1}).

Note: For Rose Bengal, the GOBN membrane was first filtrated with 10 times of DI water, and then tested with a dye solution separation test. This experiment was conducted to prove the stability of the GOBN membrane for molecule separation application.

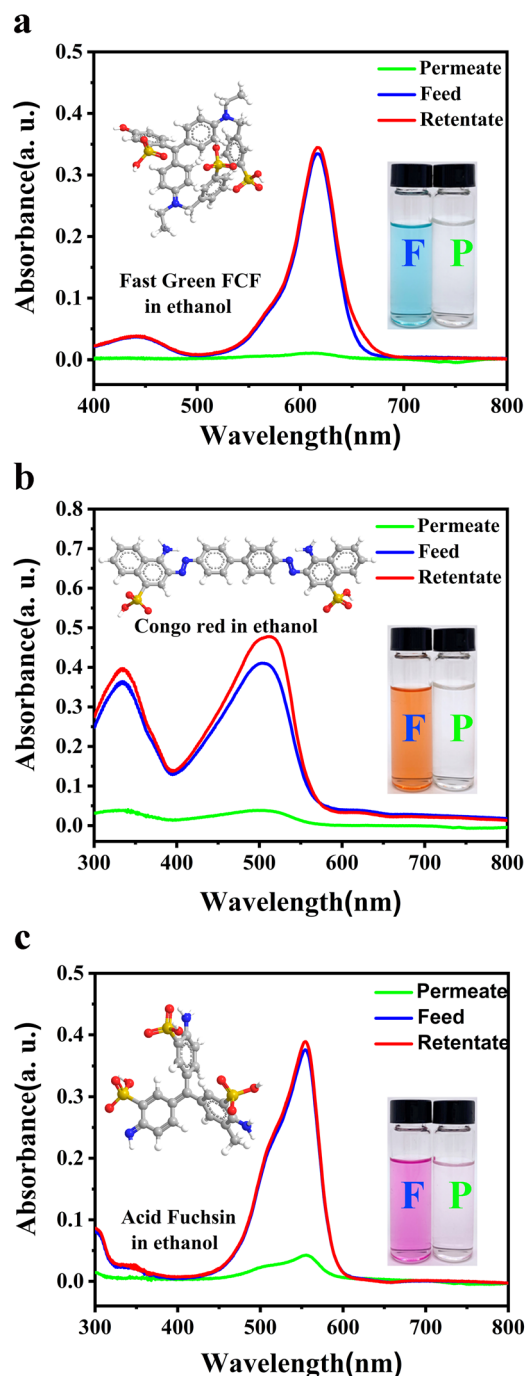


Supplementary Figure 11. Comparisons of the GO1BN2 membrane for organic dyes separation from DI water and real water from Chaohu Lake (31.6117° N, 117.8562° E). (a) Eosin Y in DI water. (b) Eosin Y in real water from Chaohu Lake. (c) Rose Bengal in DI water. (d) Rose Bengal in real water from Chaohu Lake. (e) Evans blue in DI water. (f) Evans blue in real water from Chaohu Lake.



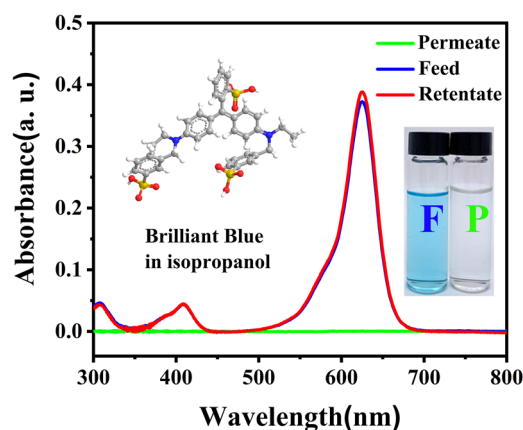
Supplementary Figure 12. UV-Vis absorption spectra of dyes before and after filtration through GOBN membrane in methanol. (a) Acid fuchsin; (b) Evans blue. The left inset is the molecular structure of tested dyes and the right inset photo shows the feed (F, left) and permeate (P, right) of the dyes solution in methanol.

Note: For Evans blue, the GOBN membrane was first filtrated with 10 times methanol, and then tested with a dye solution separation test. This trial was operated to study the stability of the GOBN membrane for molecules separation application in a methanol solution.

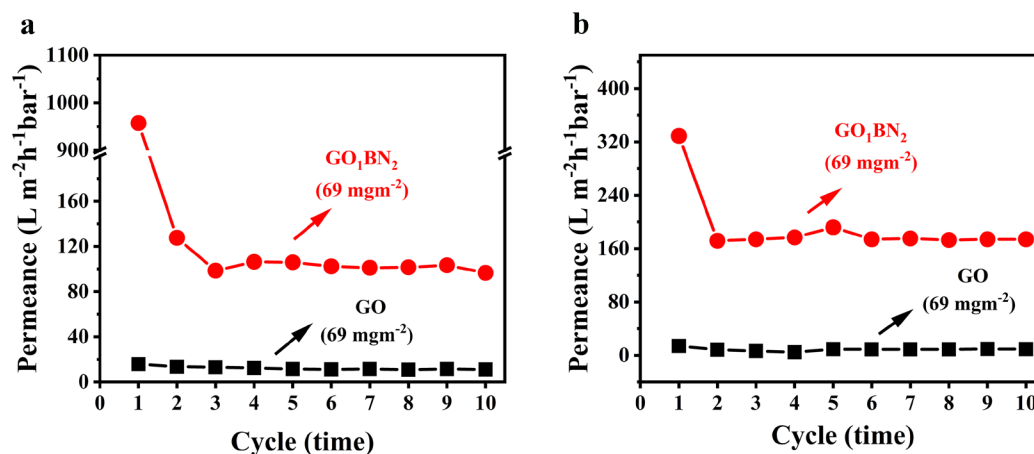


Supplementary Figure 13. UV-Vis absorption spectra of dyes before and after filtration through GOBN membrane in ethanol. (a) Fast green FCF (808.9 g mol^{-1} , concentration is 3.16 mg L^{-1}); (b) Congo red (696.7 g mol^{-1} , concentration is 22.7 mg L^{-1}); (c) Acid fuchsin ($585.53 \text{ g mol}^{-1}$, concentration is 3.34 mg L^{-1}). The left inset is the molecular structure of tested dyes and the right inset photo shows the feed (F, left) and permeate (P, right) of the dyes solution in ethanol.

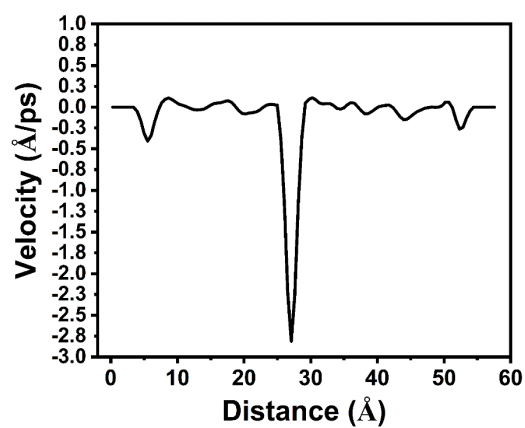
Note: For Fast green FCF, the GOBN membrane was first filtrated with 10 times of ethanol, and then tested with a dye solution separation test. This process was to investigate the stability of the GOBN membrane for molecules separation application in ethanol solution.



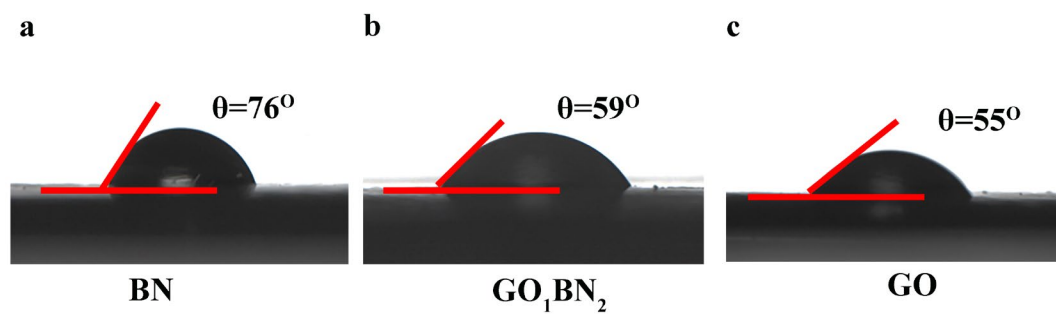
Supplementary Figure 14. UV-Vis absorption spectra of Brilliant blue (concentration of 5 mgL^{-1}) before and after filtration through GOBN membrane in isopropanol; the left inset is the molecular structure of Brilliant blue and the right inset photo shows the feed (F, left) and permeate (P, right) of the Brilliant blue solution in isopropanol alcohol.



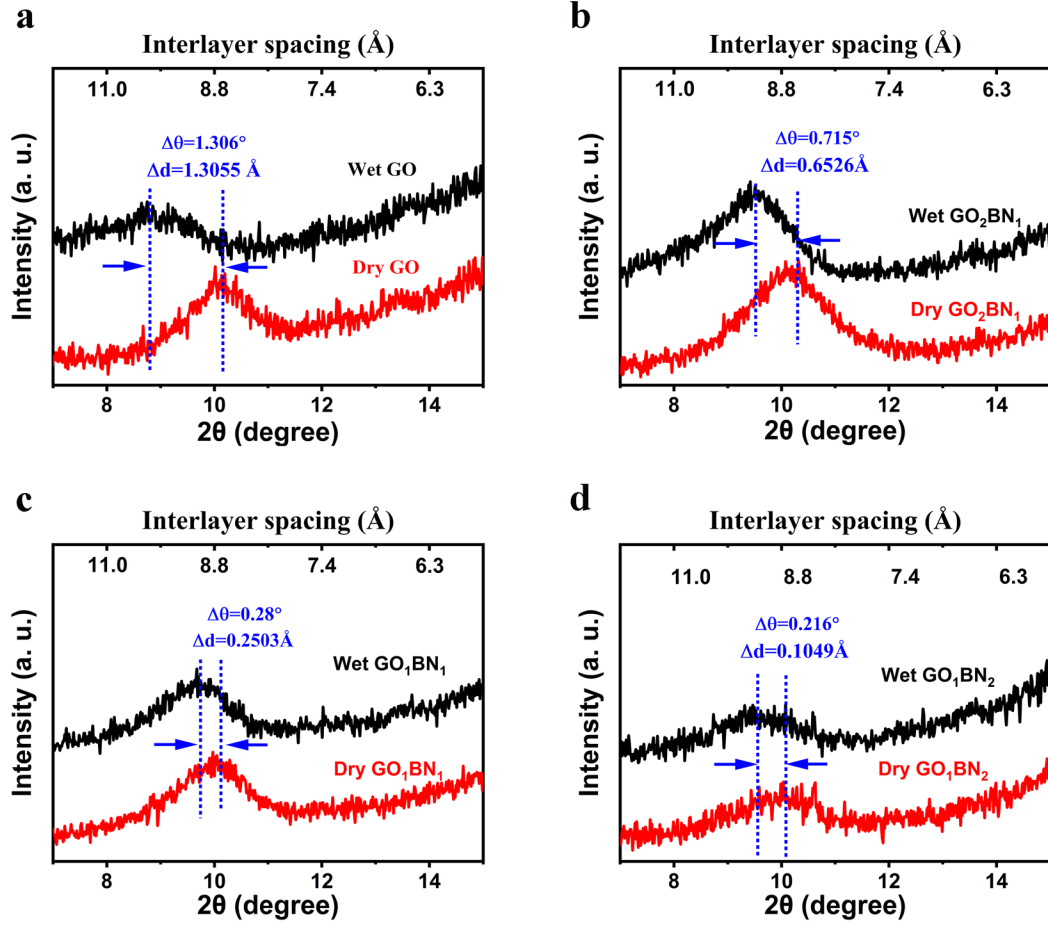
Supplementary Figure 15. The permeance of membrane versus the 10 times cycle filtration of methanol (a) and ethanol (b). The GO₁BN₂ membrane (69 mgm^{-2}) showed a sharp decreasing permeance of methanol and ethanol in the beginning two times of filtration and then stabilized to around 100 and $170 \text{ Lm}^{-2}\text{h}^{-1}\text{bar}^{-1}$, respectively.



Supplementary Figure 16. The velocity profile of water flows across the GOBN channel via the MS simulation.



Supplementary Figure 17. The contact angle of the BN (a), GOBN (b) and GO membrane (c). With the incorporation of BN nanosheets into the GOBN membrane, the contact angle increased from 59° to 76°.



Supplementary Figure 18. XRD patterns of the membrane under dry and wet conditions. (a) GO membrane; (b) GO_2BN_1 membrane; (c) GO_1BN_1 membrane; (d) GO_1BN_2 membrane.

Note: The interlayer spacing of the membrane was calculated by the Scherrer formula below

$$d = \frac{k\lambda}{\beta \cos \theta}$$

Where $\lambda = 0.15406$, $k = 0.89$, β is the half peak width, θ is in degrees, and d is the interlayer spacing in Å.

With the BN content increasing, the GOBN membrane becomes stable under water conditions.

Supplementary Table 1. Comparison of the performance of the reported membranes for the separation of different molecules with different sizes in water.

Membrane	Probe molecule	Flux ($\text{Lm}^{-2}\text{h}^{-1}\text{bar}^{-1}$)	Rejection (%)	Reference
GO-COF	MB	166.8	97.05	1
GO-TA-Ni	MB	51.5	92.9	2
GO-TA-Ni	CR	48.4	98.8	
uGNM	MB	19.6	99.8	3
NCS-GO	EB	573	83	4
NCS-GO	RhB	279	87	
NCS-GO	Cyt.c	568	98	
ZIF-8@f-GOm	MB	49.8	99.8	5
Nematic-GO	MB	67	99.3	6
GQDs-TRG	EB	800	95	7
SWCNT-GO	RhB	710	97.4	8
GO ₁ BN ₂	Eosin Y	1140	98.4	This work
	RB	1280	99.2	
	MB	1310	95.6	
	R6G	1450	89.4	
	BY	1320	98.8	
	EB	1610	86	

Note: RB is short for Rose Bengal, MB is short for Methyl blue, R6G is short for Rhodamine 6G, BY is short for Brilliant yellow, and EB is short for Evans blue.

Supplementary Table 2. Comparison of the performance of the reported membranes for the separation of different molecules with different sizes in an organic solvent.

Membrane	Solvent	Probe molecule	Flux (Lm ⁻² h ⁻¹ bar ⁻¹)	Rejection (%)	Reference
GO	methanol	RhB	16.5	95.9	9
uGO		MB	4	99.9	10
GO-La ³⁺		AF	100	95	11
HPEI/S-rGO-18		EB	73.6	100	12
		MB	72.5	90	12
CW-GO		RB	55	99	13
GO	ethanol	RhB	4.92	97.3	9
mG		RB	156.8	94.5	14
GO-TFN		RhB	41.47	99.4	15
GO ₁ BN ₂	methanol	EB	780	99.1	This work
		AF	880	87.5	
	ethanol	AF	290	89.2	
		FG	310	99.3	
		CR	350	89.6	
	isopropyl alcohol	BB	180	99.5	

Note: EB stands for Evans blue, AF stands for Acid fuchsin, FG stands for Fast Green FCF, CR stands for Congo red, and BB stands for Brilliant Blue.

Supplementary Reference.

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