Electronic Supplementary Information

Confined assembly of ultrathin dual-functionalized Z-MXene nanosheets intercalated GO nanofilms with controlled structure for size-selective permeation

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Experimental

Chemicals and materials

Natural graphite flakes were supplied by Nanjing Xianfeng Nanomaterials Technology Co. Ti_3AlC_2 was purchased from Beijing Kaifa Taote Technology Co. $NaNO_3$, N, N-dimethylacetamide (DMAc), HCl, H_2SO_4, ammonium acetate (AA), LiF, tris hydroxymethyl aminomethane (Tris), dopamine, H_2O_2, polysulfone (PSF), 2-aminoterephthalic acid (ATA) and sulfanilic acid (SA) were purchased from the market. Dyes reactive brilliant red X-3B (M_w=615.34), congo red (CR) (M_w=696.68), methyl blue (MB) (Mw=799.80) and evans blue (EB) (Mw=960.81) were purchased from market. Deionized water was made by RO-EDI system. All the reagents were all analytical standard and used without any further purification.

Table S1. Comparison of the characteristics and performance of GO-based 2D/2D lamellar membranes and GO-based ultrathin nanofiltration membranes.

Sample	**Thickness (nm)**	**Test method**	**Mark molecules**	**Molecular weight**	**Concentration (mg L^{-1})**	**Permeability (L m^{-2} h^{-1} bar^{-1})**	**Rejection (%)**	**Ref.**
GNM/SWCNT	~50	cross-flow	MB RhB FITC	373.9 479.0 389.4	/ / /	110.6	96.4 97.2 98.7	9
GO/FLG	26-33	cross-flow	RhB Acid Blue 9	479.0 792.8	/ /	12	8 96	22
GO/M-PAN	7.5 15 30	cross-flow	MO	327.3	0.02 mM (6.55)	81 86 52	79.6 83.9 91.4	42
PGO	15	cross-flow	MB TO SO	800 316 547	100	18.5	99.8 99.8 99.9	43
GO/PC	20	dead-end	BB MB EB	825.9 799.8 960.8	10	26.9	>90	44
mul-EGO	56	cross-flow	EBT MB AF CR BR	461.4 799.8 585.5 696.7 1017.6	100	37.575	98.6 >98 >98 >98 >98	45
GOCN	66	dead-end	EB MB MnB	960.8 799.8 319.8	10	15.4	95.5 92.6 85.5	46

			MO	327.3			41.2	
a-BN/GO	1210	dead-end	MB	319.8	70	4.15	99.9	47
GO/MoS_2	~4000	dead-end	CR	696.7	10	7.65	99.6	48
			MB	799.8			97.4	
			RhB	479.0			94.6	
MoS_2/GO	300	dead-end	DR80	1373.1	20	48.27	99.9	49
			CR	696.7			99.9	
			RhB	479.0			99.7	
			MB	319.8			99.6	
GO/MXene	~550	dead-end	NR	288.8	10	71.9	99.5	50
			MB	319.8			99.5	
			BB	792.8			99.5	
$Ti_2C_3T_x/GO$	90	dead-end	MR	269.3	10	2.1	61	51
			MnB	799.8		0.3	99.5	
			RosB	479.8		0.67	93.5	
			BB	792.8		0.23	100	
GAT-4	45	cross-flow	X-3B	615.3	10	115.5	84.7	This work
			CR	696.7			99.1	
			MB	799.8			99.2	
			EB	960.8			99.7	
GAT-5	46	cross-flow	CR	696.7		141.5	94.1	

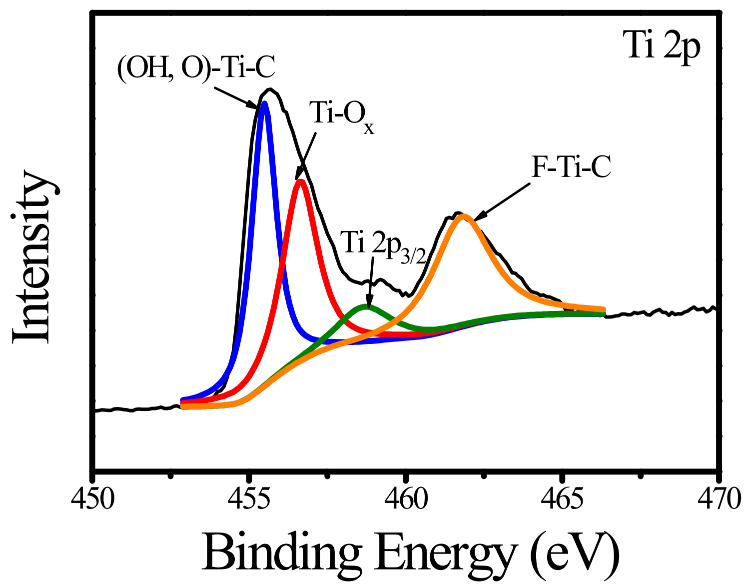

Figure S1. Ti 2p spectra of $Ti_3C_2T_x$ MXene treated by stirring original $Ti_3C_2T_x$ MXene at room temperature for 24 h in an atmosphere of argon.

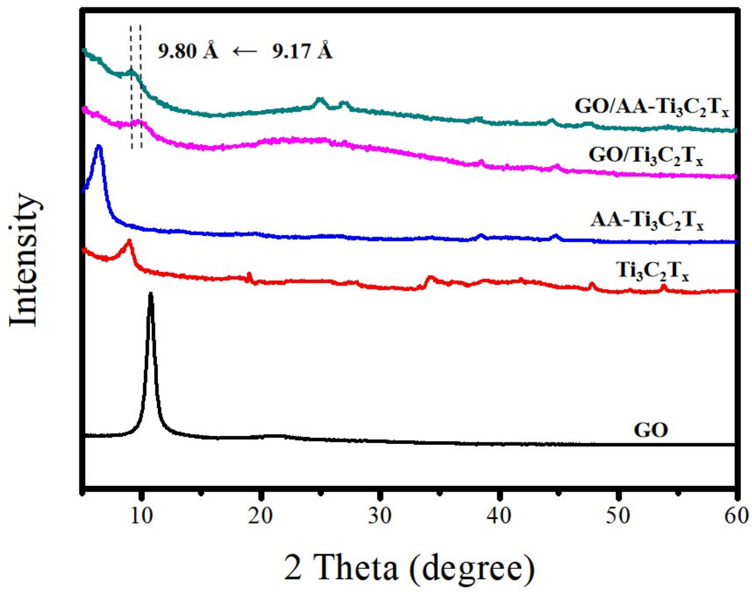

Figure S2. XRD patterns of GO, $Ti_3C_2T_x$, AA-$Ti_3C_2T_x$, GO/$Ti_3C_2T_x$ and GO/AA-$Ti_3C_2T_x$ nanosheets.

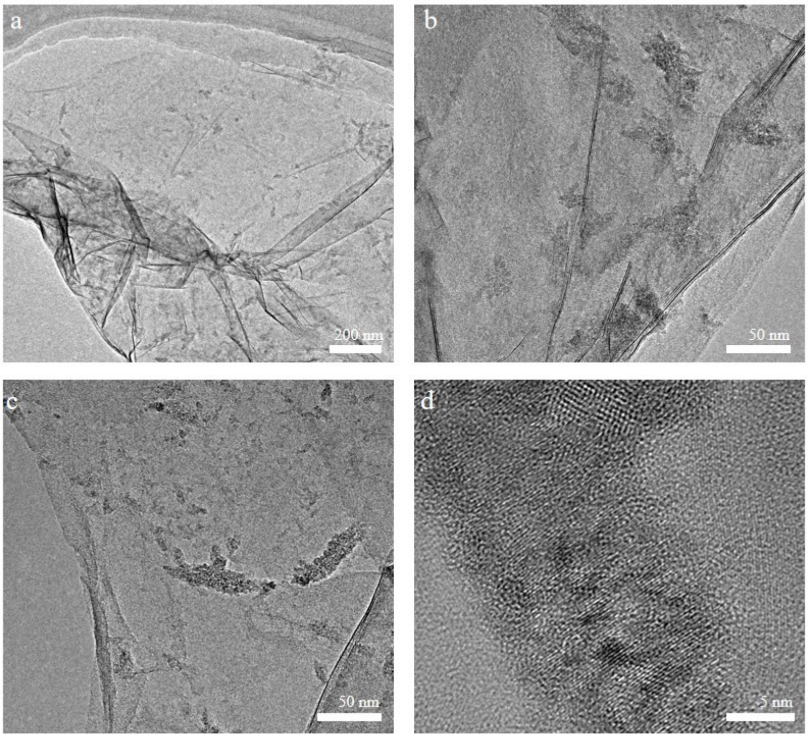

Figure S3. (a-d) TEM images of GO/$Ti_3C_2T_x$ nanosheets with different magnifications.

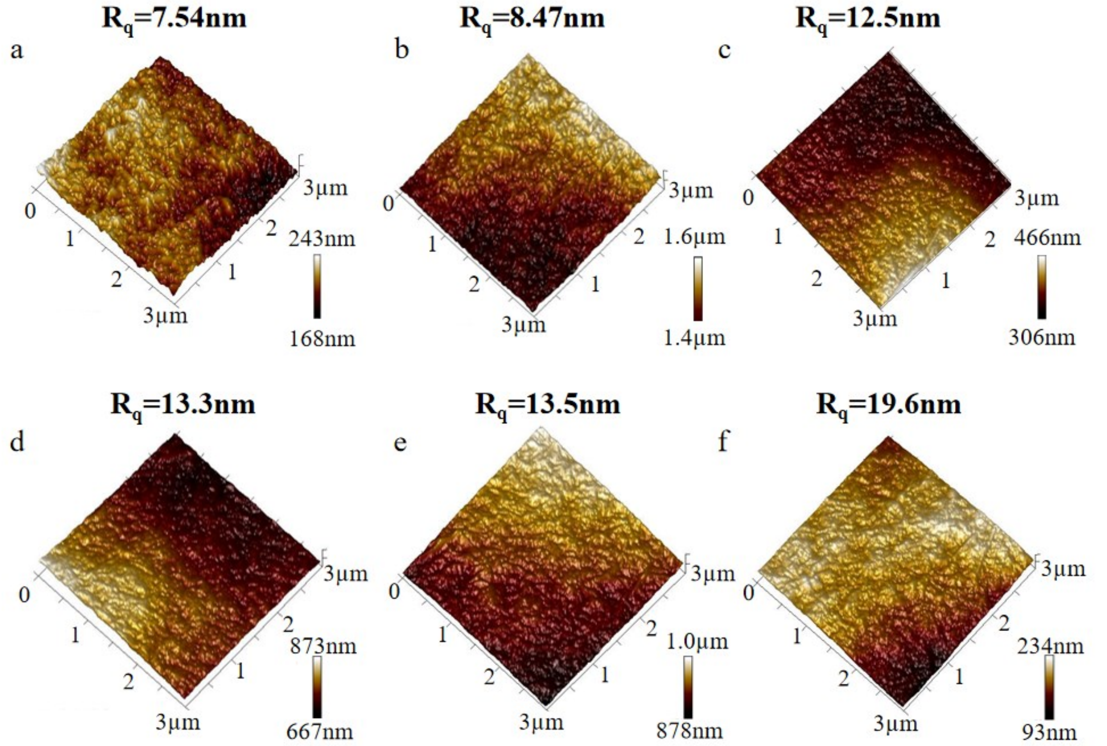

Figure S4. AFM images of GAT-0 (a), GAT-1 (b), GAT-2 (c), GAT-3 (d), GAT-4 (e) and GAT-5 (f).

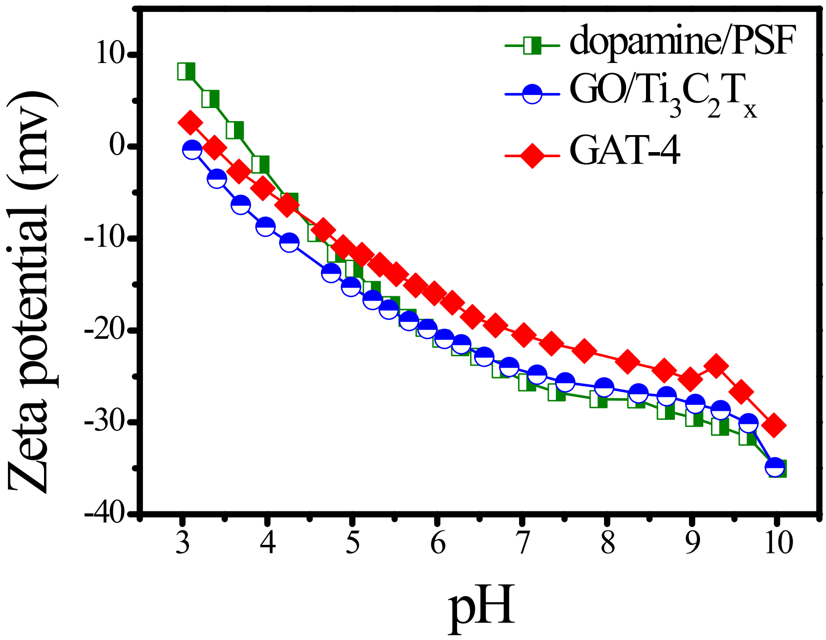

Figure S5. Zeta potential of dopamine coated PSF support and prepared GT and GAT-4 membranes.

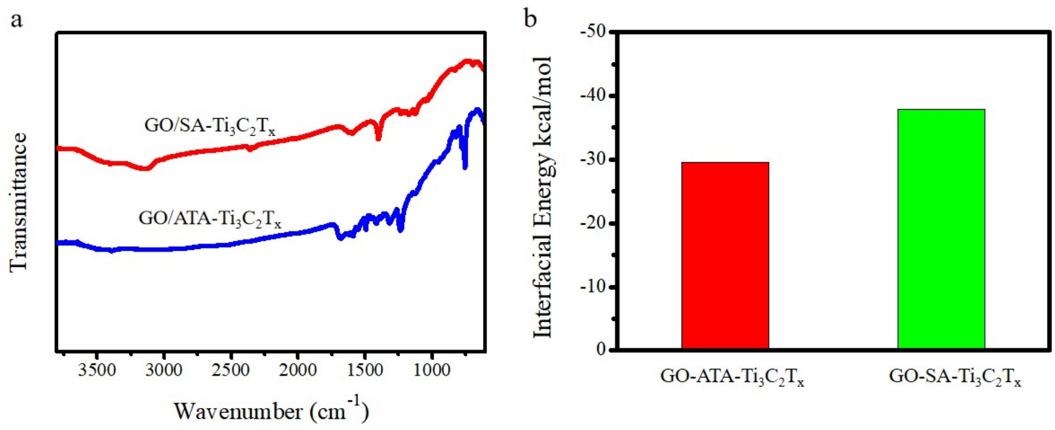

Figure S6. (a) FTIR spectra of the synthesized GO/ATA-$Ti_3C_2T_x$ and GO/SA-$Ti_3C_2T_x$ nanofilms. (b) The interfacial energy for GO-ATA-$Ti_3C_2T_x$ and GO-SA-$Ti_3C_2T_x$ interface calculated from molecular simulations. The peaks at 1215 and 1048 cm^{-1} are ascribed to the stretching vibration peaks of -S=O of -SO_3H in GO/SA-$Ti_3C_2T_x$.[S1] The presence of amino and carboxylic groups on the surface of GO/ATA-$Ti_3C_2T_x$ nanocomposites is confirmed by FTIR analysis (1470 cm^{-1} (-NH bending and 1725 cm^{-1} (C=O stretching).[S2] The results above demonstrated that 2-aminoterephthalic acid and sulfanilic acid were grafted on the surface of $Ti_3C_2T_x$ nanosheets.

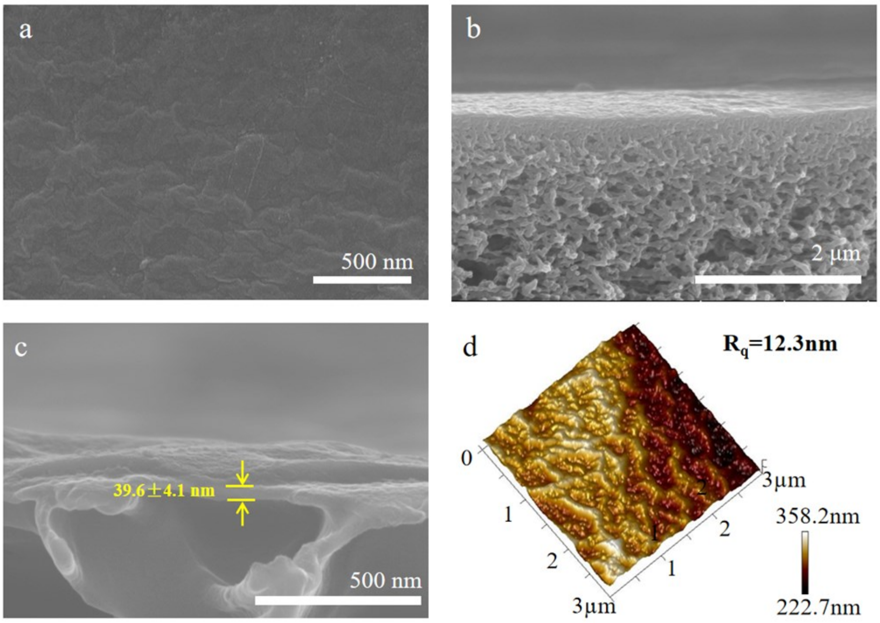

Figure S7. Surface SEM image (a) and cross-section SEM images (b, c) of $GO/Ti_3C_2T_x$ nanofilm. (d) AFM image of $GO/Ti_3C_2T_x$ nanofilm.

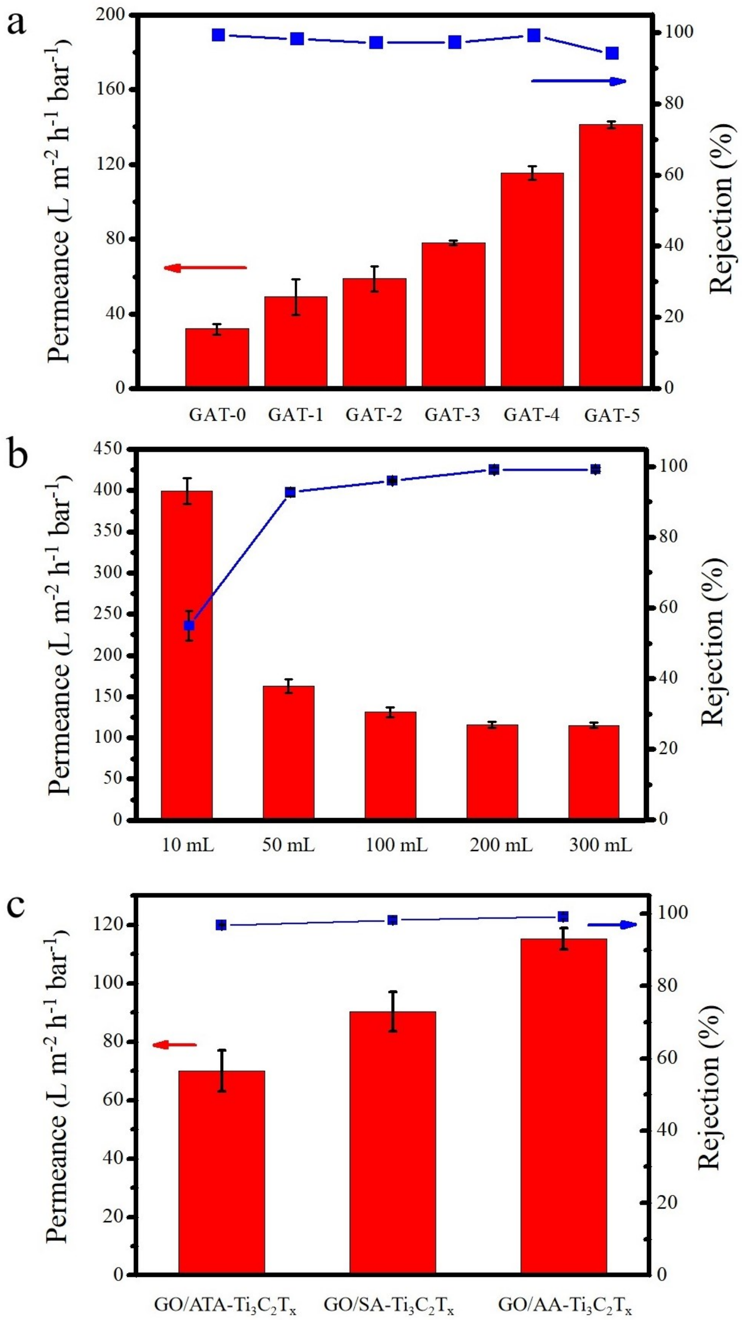

Figure S8. (a) Pure water flux and dye rejection of prepared nanofilms. (b) Permeance of the synthesized GO/AA-$Ti_3C_2T_x$ nanofilms by dispersing same amount of GO/AA-$Ti_3C_2T_x$ in 10, 50, 100, 200 and 300 mL water. (c) Permeance of the synthesized GO/ATA-$Ti_3C_2T_x$, GO/SA-$Ti_3C_2T_x$ and GO/AA-$Ti_3C_2T_x$ nanofilms.

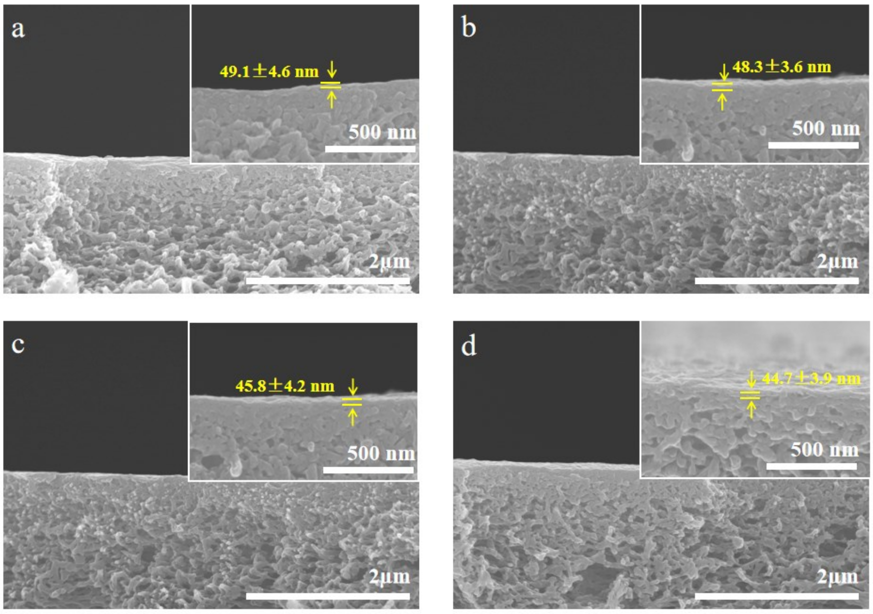

Figure S9. Cross-section SEM images of GO/AA-$Ti_3C_2T_x$ membranes by dispersing same amount of GO/AA-$Ti_3C_2T_x$ in 10 (a), 50 (b), 100 (c) and 300 mL water (d).

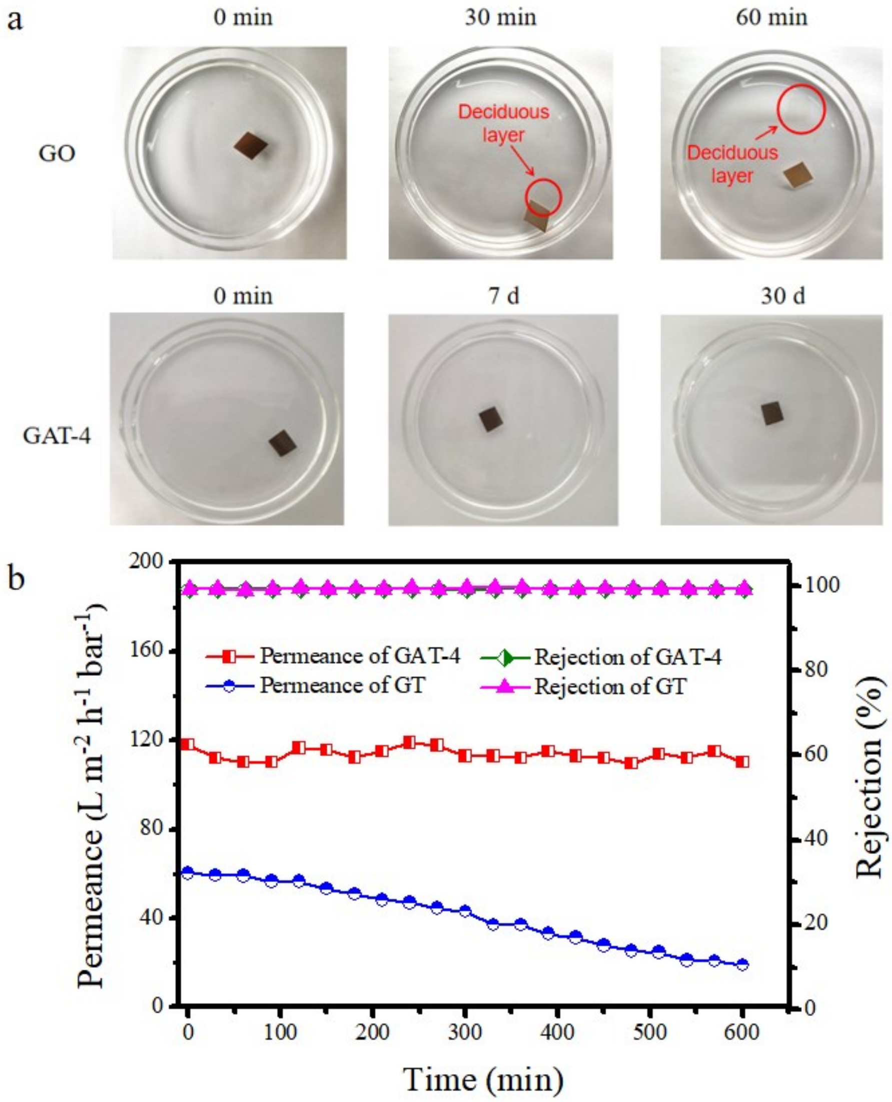

Figure S10. (a) Photographs of the GO and GAT-4 membranes before and after soaking in water for a period of time. (b) The long-term test on continuous dye separation performance for GAT-4 and GT membranes.

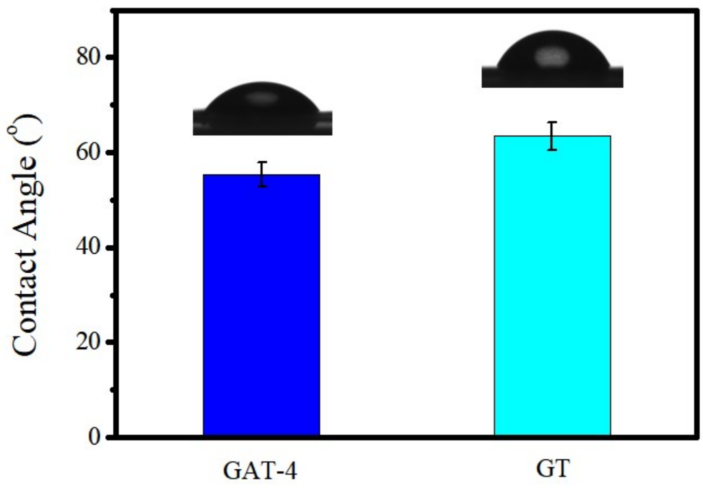

Figure S11. Water contact angles of GAT and GT membranes.

References:

S1. S. Saha, M. Jana, P. Samanta, N. C. Murmu, T. Kuila, *J. Mater. Chem. A*, 2015, **3**, 19461.

S2. Z. Z. Liu, F. R. Cao, M. Wang, M. Wang, L. Li, *Angew. Chem. Int. Ed*., 2020, **59**, 4161.