Supporting information

MgAl-Layered-Double-Hydroxide/sepiolite composite membrane for high-performance water treatment based on layer-by-layer hierarchical architectures

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Experimental section

Fabrication of the MgAl-LDH/Sep membranes

Firstly, The Sep and MgAl-LDH were dissolved in ultrapure water to make a 0.6g/L and 0.5g/L suspension, respectively. Secondly, under continuous stirring, the mixtures were dispersed uniformly by sonicating for 30 min. Finally, the samples were filtered onto the surface of substrate in vacuum one by one to form the hierarchical structure membrane.

Table S1 Compositions of the membranes prepared in this report

Sample / Composition (mg)	LDH-M	Sep-M	M1	M2	M3	M4
LDH	6	0	6	6	6	6
Sep	0	6	6	12	18	24
Al^{3+}	750	750	750	750	750	750

Oil–water emulsion separation and cycling test

The separation process was carried out on a vacuum apparatus. The O/W emulsion were poured onto the membranes and separated under vacuum filtrating. The cycling test corresponded to the anti-fouling ability of the membrane. For each separation cycle, 20 mL of an O/W emulsion was permeated through the membrane under a pressure of 0.1 MPa. The filtrate water was collected and the separation efficiency for each emulsion was calculated by TOC.

Dye removal experiments

In this process, CR and MB as ionic dye were applied severally. Dye removal tests were performed through a filtration system with an effective membrane area of 12.566 cm^2. In detail, 20 mL of synthetic dye solution with diverse concentration was filtered, and then flux and rejection were determined using the equations of (1) and (2). Spectrophotometric measurement was used to obtain the dye concentration at 496 nm and 664 nm (UV–Vis spectrophotometer (UV-762 (Shanghai precision scientific instrument co.))). In addition, to exclude the randomness of the experiment and study the flux stability and pollution resistant capacity of all membranes, each experiment was conducted three times at 1.0 MPa. Furthermore, two dye solutions including 100, 200, 300 mg/mL CR and 10, 20, 30 mg/mL MB were tested on the membrane as anionic and cationic dyes, respectively.

Figure S1. Filtering apparatus

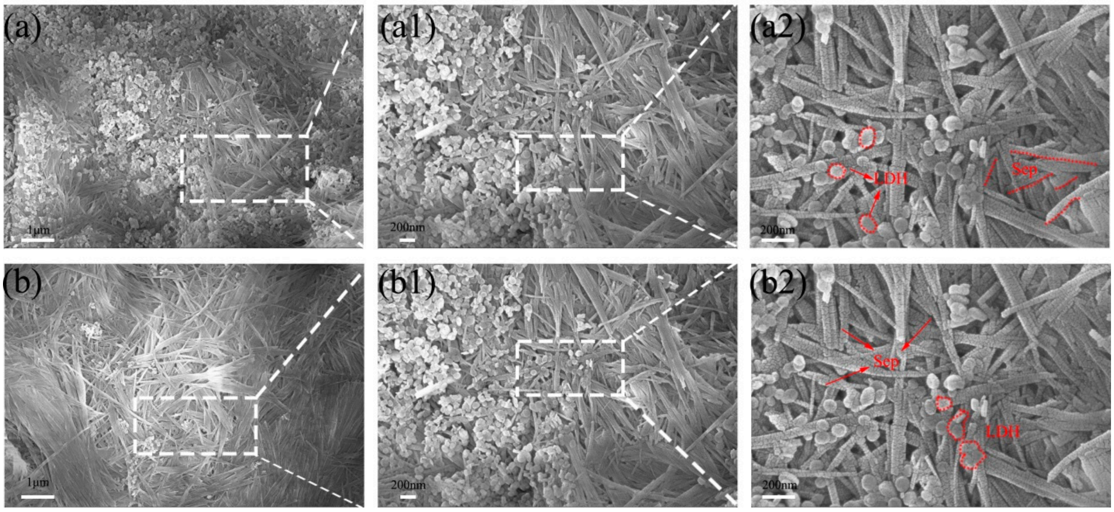


Figure S2. SEM image for M3 (a)-(a2) and M4 (b)-(b2) of membrane

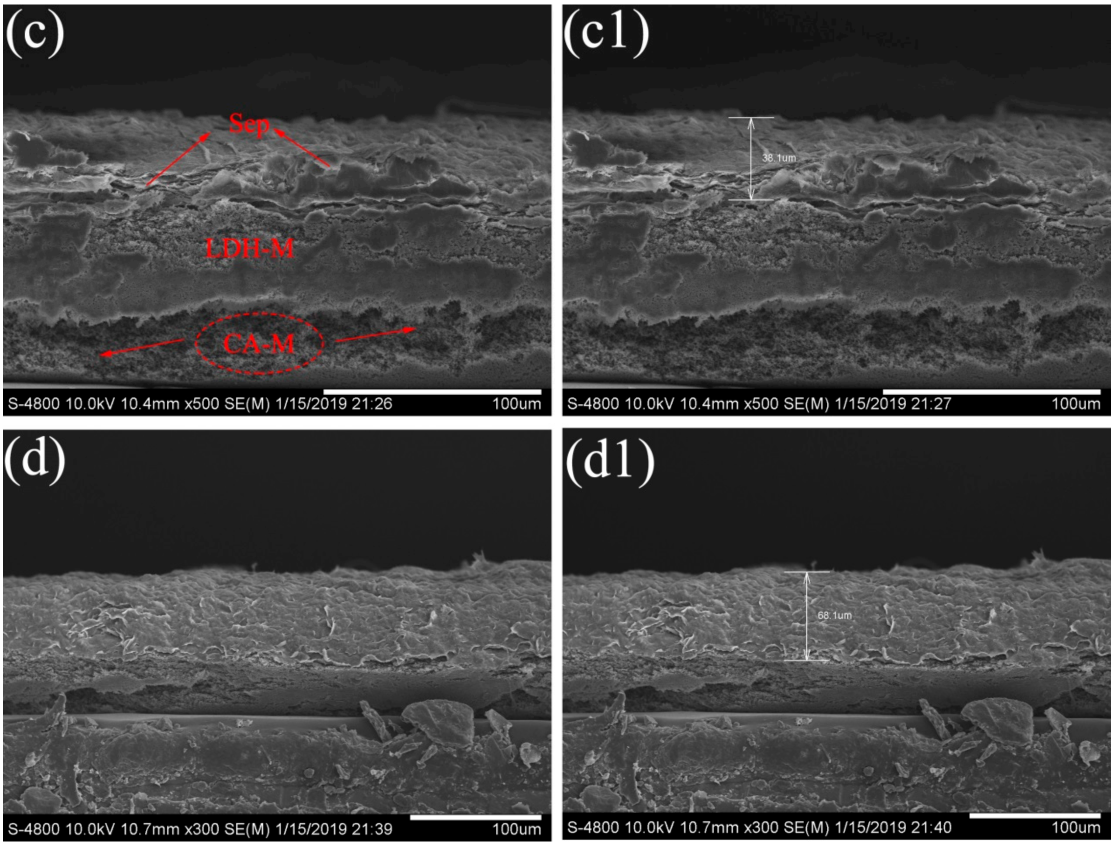


Figure S3. SEM image for M3 (c), (c1) and M4 (d), (d1) of membrane cross-section

Table S2 Surface roughness data

Filter papers	Roughness		Standard deviation(nm)
	R_a(nm)	R_q(nm)	
LDH-M	72.1	106	27.6
M2	166	217	47.2

Table S3 Pore size data

Name	LDH-M	M2
Measured bubble point pressure (bar)	1.1026	3.8881
Measured bubble point flow (L/min)	0.0066	0.0041
Minimum pore size pressure (bar)	3.0256	15.135
Average pore size pressure (bar)	2.155	12.7582
Gas permeability ($m^3/(m^2.pa.s)$)	3.94E-02	7.09E-03
Gas flux ($m^3/(m^2.h)$) (ΔP=0.1000bar)	1.45E+02	1.20E+01
Measured bubble point pore size (μm)	0.4367	0.1239
Optimal pore size (μm)	0.2278	0.0386
Minimum pore size (μm)	0.1592	0.0318
Average pore size (μm)	0.2235	0.0377

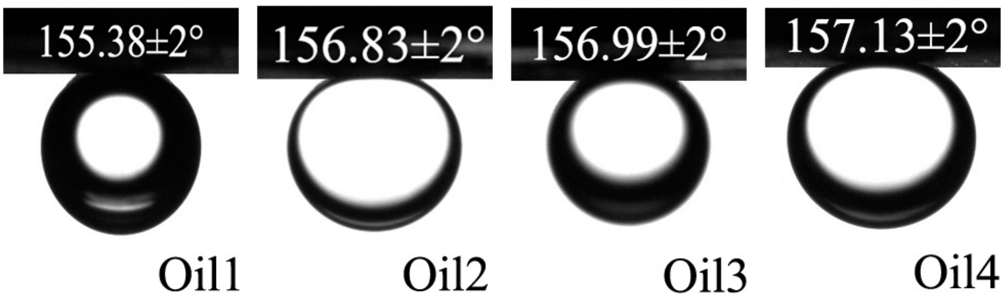


Figure S4. Underwater oil contact angle

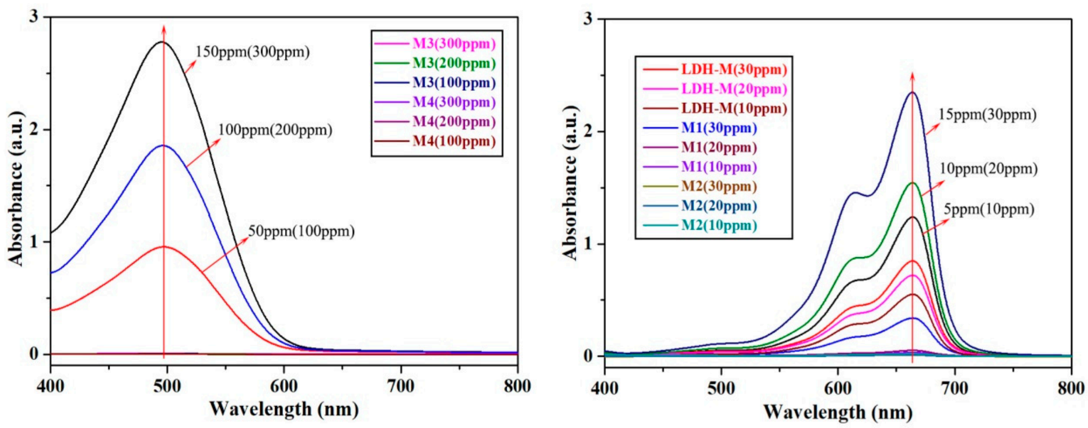
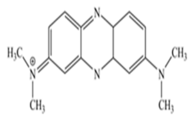
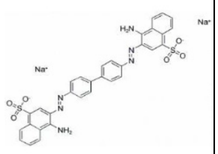
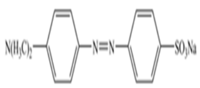
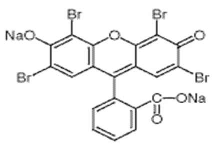


Figure S5. UV–visible absorption spectra

Table S4. The rejection of as-prepared membrane (M2) for different type of organic dye.

Dye name	Molecular formula	Relative molecular mass (g/mol)	Volume (ml)	Concentration (ppm)	Rejection
Methylene Blue (MB)		374	20	30	99.79%
Congo Red (CR)		696.68	20	300	99.89%
Methyl Orange (MO)		327	20	30	99.82%
Acid Red 87 (AR)		691.86	20	300	99.91%

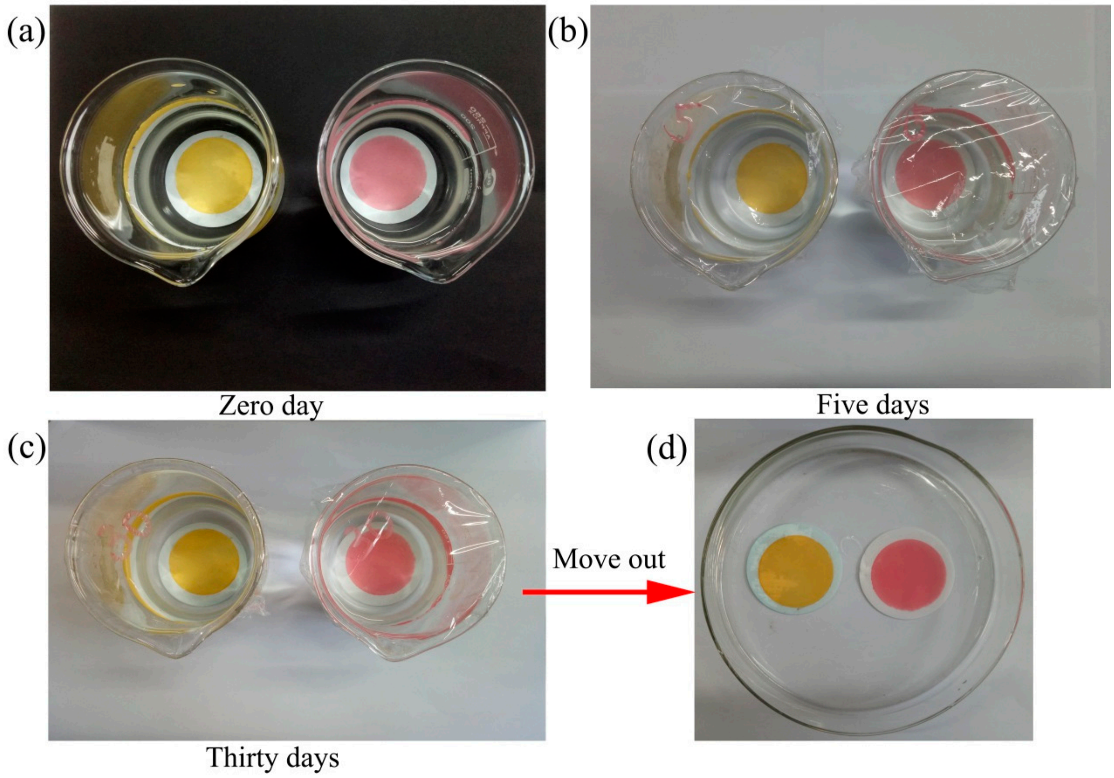


Figure S6.The photographs of the membrane in the water for 30 days (Since the color of the MgAl-LDH/Sep membranes is light yellow and difficult to observe, the membrane is dyed with a small amount of dye for easy observation.)