Supporting information for

Fabrication Novel Nanofiltration Membrane using Mg-Fe Layered Double Hydroxide for dye/salt separation

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1. NF membrane preparation condition optimization

The influence of membrane preparation conditions on the membrane performance was studied. In order to obtain NF membrane with optimal separation performance, the concentration of PIP, TMC, Mg-Fe LDH, heat treatment time, and heat treatment temperature were optimized based on the permeation flux and rejection rate for 1000 $mg \cdot L^{-1}$ Na_2SO_4 solution.

1.1 Element mapping images of Mg-Fe LDH

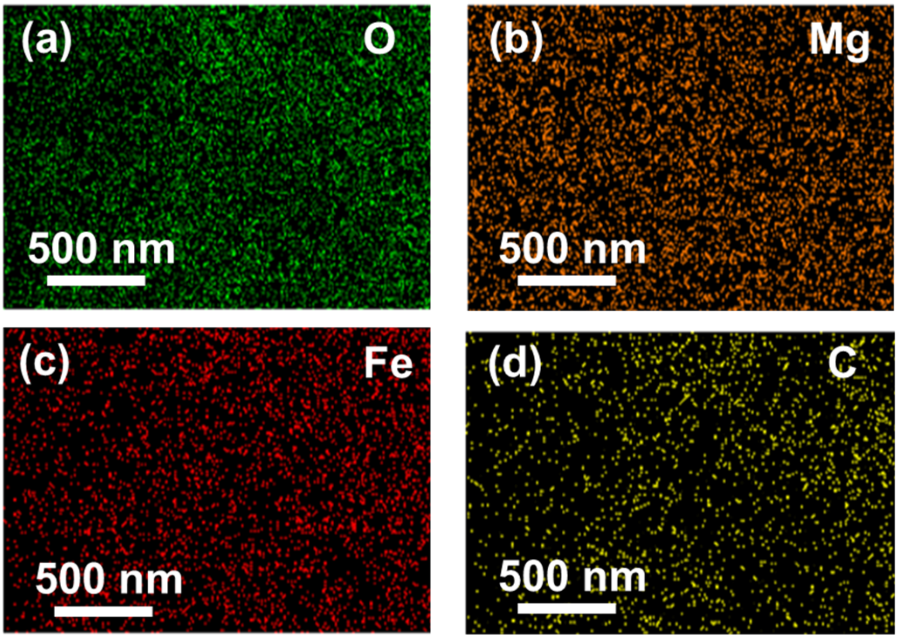

Fig. S1 (a) O, (b) Mg, (c) Fe, and (d) C element mapping images of Mg-Fe LDH

1.2 FT-IR spectra of Mg-Fe LDH

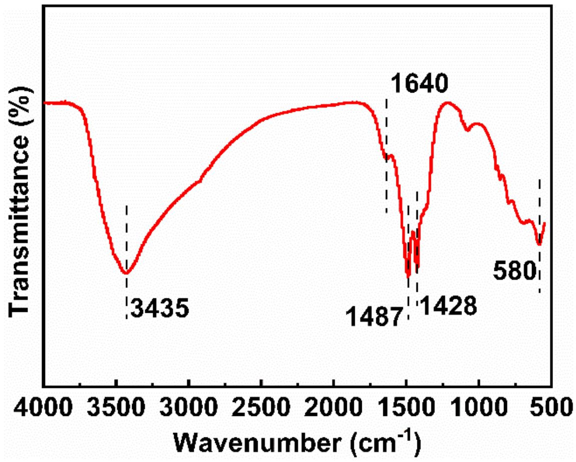

Fig. S2 FT-IR spectra of Mg-Fe LDH

1.3 Wide-scan XPS analysis spectra of different NF membranes

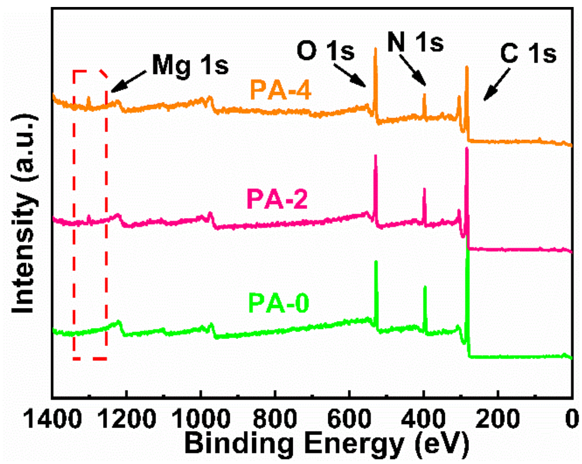

Fig. S3 Wide-scan XPS analysis spectra of different NF membranes

1.4 The effect of PIP concentration

PIP concentration will directly affect the thickness of the selective layer. The effect of aqueous PIP concentration on NF separation performance was investigated firstly and the results were shown in Fig. S4. TMC concentration was fixed at 0.15 wt%, Mg-Fe LDH concentration was 0.1 wt%, heat treatment temperature was 60 ℃ and heat treatment time was 10 min when PIP concentration was changed. As shown in Fig. S4, the permeation flux of Mg-Fe LDH modified NF membrane for Na_2SO_4

gradually decreases with the increase of the PIP concentration, while the rejection rate for Na_2SO_4 increased continuously. With the increase of PIP concentration, the number of PIP molecules fixed into the selective layer increased due to TMC concentration being fixed, the effective thickness of the selective layer increases correspondingly which led to the decrease of permeation flux and the increase of rejection rate. Considering the permeation flux and rejection rate of the NF membrane, the optimal concentration of PIP in this study was chosen as 0.3 wt%.

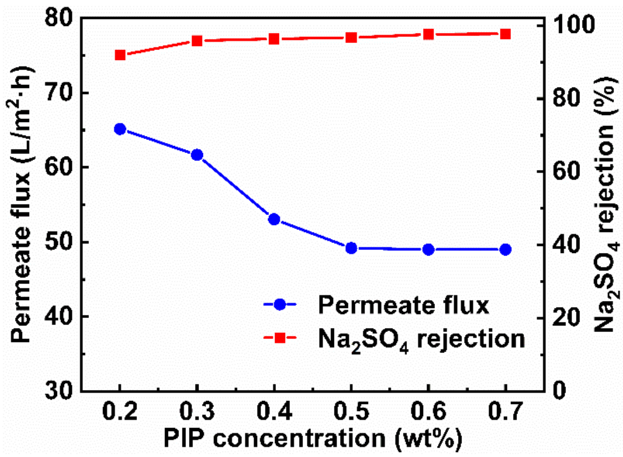

Fig. S4 Effect of PIP concentration on Mg-Fe LDH modified NF membrane performance.

1.5 The effect of TMC concentration

TMC concentration will directly determine the compactness of the selective layer. When Mg-Fe LDH concentration was fixed at 0.1 wt%, PIP concentration was 0.3 wt%, heat treatment temperature was 60 °C and heat treatment time was 10 min, the effect of TMC concentration on NF separation performance was investigated and the results were shown in Fig. S5. As shown in Fig. S5, when the TMC concentration was lower than 0.15 wt%, the permeation flux of Mg-Fe LDH modified NF membrane to the Na_2SO_4 solution increased with TMC concentration increase and the salt rejection rate kept relatively stable. However, when the TMC concentration range in 0.15-0.25 wt%, the permeation flux decreases gradually accompanied by the rejection rate increase slightly. When TMC concentration was higher than 0.25 wt%, the permeation flux increases obviously, and the rejection rate decreases sharply. If the concentration of TMC was lower than 0.15 wt%, the reaction was not complete and the crosslinking rate of the functional layer was not good. Along with the addition of TMC concentration, the crosslinking rate of the functional layer gets better, the surface of the composite layer becomes rougher. As a result, the contacting area of the membrane and water was larger, which promotes the mass transfer of water[1]. As TMC concentration increases (higher than 0.15 wt%), the cross-linking degree of the functional layer increases which leads to increases in rejection rate and the decrease of permeation flux. However, if TMC concentration was too high (higher than 0.25 wt%), the interfacial polymerization reaction became too fast, the formed cross-linking functional layer will relatively loose and the cross-linking structure presents some defects. Some unreacted acyl chloride groups will hydrolyze to carboxyl groups, which causes the permeate flux of the membrane to increase and improves the hydrophilicity of the membrane[2].

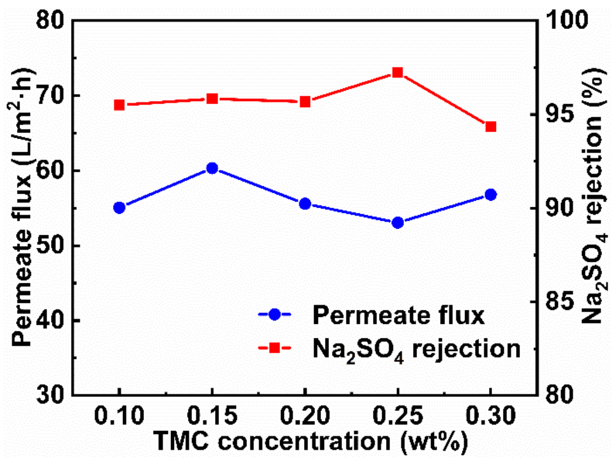

Fig. S5 Effect of TMC concentration on Mg-Fe LDH modified NF membrane performance

1.6 The effect of Mg-Fe LDH concentration

The presenting of nanoparticles LDH will affect the morphology and structure of NF membranes. The effect of Mg-Fe LDH concentration on NF separation performance was investigated when PIP concentration was 0.3 wt%, TMC concentration was 0.15 wt%, heat treatment temperature was 60 °C, heat treatment time was 10 min. As it can be seen from Fig.S6, the permeation flux of modified NF membrane increases with Mg-Fe LDH concentration increase very obviously, and the rejection rate for Na_2SO_4 decreases slightly. The flux improvement was contributed to the addition of Mg-Fe LDH to provide additional channels for water molecule transportation. At the same time, the hydrophilicity of the NF membrane was improved obviously due to Mg-Fe LDH molecules having abundant hydroxyl groups, which can promote and increase the absorption of water molecules on the NF membrane surface[3]. However, accompanied by an LDH loading increase, the rejection rate of the membrane decreased slightly, which may be due to LDH providing an additional transport channel and forming some defects in the selective layer. Considering the permeation flux and rejection rate of the NF membrane, the optimized Mg-Fe LDH concentration was chosen as 0.1 wt%.

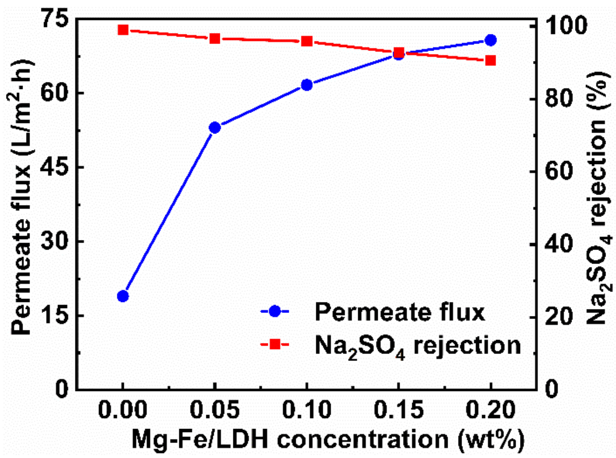

Fig. S6 Effect of Mg-Fe LDH concentration on NF membrane performance

1.7 Effect of heat treatment temperature

When Mg-Fe LDH concentration was fixed at 0.1 wt%, PIP concentration was 0.3 wt%, TMC concentration was 0.15 wt%, and the heat treatment time was 10 min, the effect of heat treatment temperature on NF membrane separation performance was investigated and the results are shown in Fig. S7. With the increase of the heat treatment temperature, the permeation flux of the NF membrane decreases gradually, and the rejection rate increase firstly and then decreases slowly. To some degree, the increase of temperature can improve interfacial polymerization reaction, which leads to the cross-linking degree of selective functional layer increase and makes the selective layer denser. However, if the temperature was too high, some chemical bonds and hydrogen bonds of the cross-linking selective layer will break, and some defects will be formed, which lead to the reduction of the rejection rate. Considering the permeate flux and rejection rate of the NF membrane, the heat treatment temperature was fixed at 60°C in the following study.

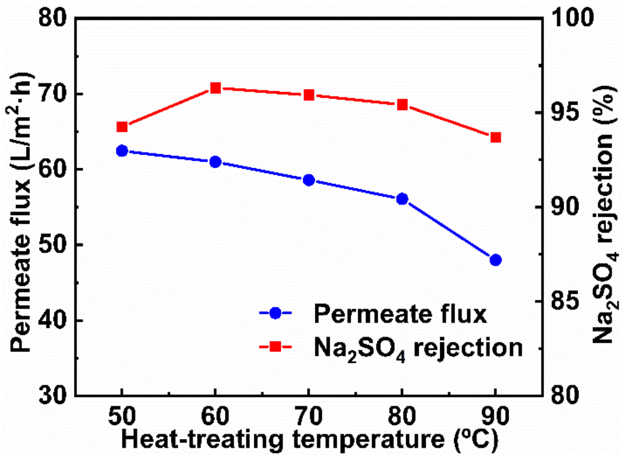

Fig. S7 Effect of heat treatment temperature on NF membrane performance

1.8 Effect of heat treatment time

Effect of heat treatment time on NF membrane separation performance was investigated when Mg-Fe LDH concentration was fixed at 0.1 wt%, PIP concentration was 0.3 wt%, TMC concentration was 0.15 wt%, and the heat treatment temperature was 60°C. As it can be seen from Fig.S8, with the increase of heat treatment time the rejection rate of Na_2SO_4 increased firstly and then decreased. However, accompanied by the change of rejection rate, the permeation flux presented an opposite change trend. The heat treatment process will accelerate the interface polymerization reaction speed. If the heat treatment time was less than 15 min, with heat treatment time prolonging, PIP and TMC will be crosslinked further, which lead the formed selective functional layer denser. As a result, the membrane permeation flux decreased and the salt rejection rate increased. However, as the heat treatment time continues to increase, some cracks and defects will appear in the selective functional layer due to the expansion coefficients of the selective layer and the support layer being different, which contributed to the increase of permeation flux and the decrease the rejection rate. Considering the permeate flux and rejection rate of the NF membrane, the heat treatment time was fixed at 10 min.

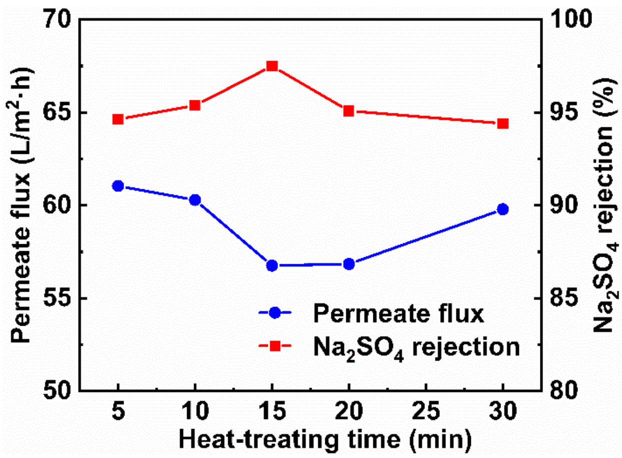

Fig. S8 Effect of heat treatment time on Mg-Fe LDH modified NF membrane.

Table S1 Preparation conditions of different NF membrane

Membrane	LDH (wt%)	PIP (wt%)	TMC (wt%)	Heat treatment temperature (°C)	Heat treatment time (min)
PA-0	0	0.3	0.15	60	10
PA-1	0.05	0.3	0.15	60	10
PA-2	0.1	0.3	0.15	60	10
PA-3	0.15	0.3	0.15	60	10
PA-4	0.2	0.3	0.15	60	10

Table S2 The molecular weight and molecular radius of PEG

Solute	Molecular Weight	r_s
PEG-200	200.00	0.64
PEG-400	400.00	0.94
PEG-600	600.00	1.18
PEG-800	800.00	1.38
PEG-1000	1000.00	1.56

Table S3 Surface chemical compositions of different NF membranes

Membrane	Surface chemical compositions (At.%)				
	C	O	N	Mg	Fe
PA-0	73.70	13.27	13.04	-	-
PA-2	70.37	16.17	11.57	1.42	0.47
PA-4	60.09	22.48	11.63	4.34	1.46

Table S4 Roughness of different NF membranes

Membranes	Ra (nm)	RMS (nm)
PA-0	4.99	6.58
PA-2	2.94	3.71
PA-4	2.43	2.98

Table S5 Characteristic parameters of different membranes

Membrane	PWP（$L\cdot m^{-2}\cdot h^{-1}$）	MWCO（Da）	μ_p（nm）	ρ_p
PSF	800	50,000	3	-
PA-0	30.1 ± 0.3	281	0.415	1.53
PA-2	70.4 ± 1.1	385	0.493	1.69
PA-4	80.8 ± 0.4	597	0.559	1.75

Table S6 Property comparison of different nanomaterial-modified NF membrane

	Nanomaterials	Reactive monomer	Flux ($L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$)	Rejection rate (%)	Contaminant	Reference
1	ODA-h-NCs	PIP/TMC	8.97	95.8	Na_2SO_4	[4]
2	AgNPs	MPD/TMC	10.4	97.7	Na_2SO_4	[5]
3	MWCNTs-OH	PIP/TMC	6.9	97.6	Na_2SO_4	[6]
4	O-MoS_2	PIP/TMC	7.91	97.9	Na_2SO_4	[7]
5	rGO/TiO_2	PIP/TMC	6.0	93.6	Na_2SO_4	[8]
6	r-GO	PIP/TMC	6.6	98.5	Na_2SO_4	[9]
7	GO	PIP/TMC	10.4	94.6	Na_2SO_4	[10]
8	GO/MWCNTs	PIP/TMC	13.9	94.0	Na_2SO_4	[10]
9	Blank	PIP/TMC	5.7	98.9	Na_2SO_4	This work
10	Mg-Fe LDH	PIP/TMC	15.2	96.4	Na_2SO_4	This work

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