Electronic Supplementary Information

General Strategy for Synthesis of Layered Double Hydroxide Nanoscrolls on Arbitrary Substrates: Its Formation and Multifunction

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Figure S1. The high-resolution N 1s spectra of the PDA/PEI coated nanofiber membrane.

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Figure S2. SEM (a) and TEM (b) images of the LDH nanoscrolls.



Figure S3. TGA curve of the pure LDH nanoscrolls.

For the pure LDHs, the weight loss at 800 °C was about 33.75 % (Figure S3). Then the LDH content in the prepared samples was determined by the following formula:

$$0.0586(1-x) + 0.3375x = 0.2326 \tag{1}$$

where the *x* is the LDH content. In equation (1), the first term is the weight loss of organic layer, the second term represents the weight loss of LDH nanoscrolls, and the last term is the weight loss of hybrid membranes at 800 °C. From equation (1) the *x* was calculated to be about 62.38 %.



Figure S4. EDX spectrum of the LDH nanoscroll coated membranes.



Figure S5. (a) The high-resolution Co 2p and (b) Al 2p XPS spectra of the LDH coated nanofiber membrane and the LDH powder. The Co 2p spectra between LDH coated membrane and LDH powder was similar, while a new peak located at 77 eV was observed in Al 2p spectra for LDH coated membranes.



Figure S6. SEM micrographs of the LDHs grown on the nanofibers at different times (at 110 °C and the total metal concentration of 96 mmol/L): (a) 3 h, (b) 6h, (c) 12 h and (d) 24 h.



Figure S7. SEM micrographs of the LDHs grown on the nanofibers at different temperatures with the total metal concentration of 96 mmol/L: (a) 100 °C, (b) 120 °C and (c) their corresponding XRD patterns. (d) The adsorption properties of the LDH coated samples synthesized at different temperatures for 100 mg/L MO aqueous solution.



Figure S8. SEM micrographs of the LDHs grown on the nanofibers at 110 °C with different metal concentrations: (a) 12 mmol/L, (b) 48 mol/L and (c) their corresponding XRD patterns. (d) The adsorption properties of the LDH coated samples synthesized at different metal concentrations for 100 mg/L MO aqueous solution.



Figure S9. (a, b) SEM micrographs of the LDHs grown on the nanofibers for 12 h. (c, d) SEM micrographs of the sample in (a, b) with another 12 h hydrothermal treatment.



Figure S10. (a) Zeta potential of the membrane surfaces at the pH around the distilled water. (b) SEM image of the LDHs grown on the nascent SiO_2 membranes. (c, d) SEM images of the LDHs grown on the PDA coated nanofibers with different magnifications.



Figure S11. SEM images of the (a) aluminum foil, (b) PDA/PEI coated aluminum foil and (c, d) LDHs grown on the PDA/PEI coated aluminum foil with different magnifications.



Figure S12. SEM images of the (a) sponge, (b) PDA/PEI coated sponge and (c, d) LDHs grown on the PDA/PEI coated sponge with different magnifications.



Figure S13. Color of the LDH coated membrane (a) before and (b) after adsorption. (c) FT-IR spectra and (d) XRD patterns of the MO, LDH coated membrane before and after adsorption.

MO was characterized by adsorption bands notably at 1606 cm⁻¹ for the stretching vibrations of aromatic C=C bonds, 1363 cm⁻¹ for C–N bending and 1037 cm⁻¹ for the stretching vibrations of SO₃⁻ groups. After adsorption, the O–H bonds was shifted from 3448 cm⁻¹ to 3401 cm⁻¹, and the v4 band of SO₄²⁻ at 676 cm⁻¹ in the LDH coated membranes disappeared, which was attributed to the interaction between the MO and the hydroxyl groups on the LDH layers.¹ The increased $d_{(003)}$ from 0.889 nm to 1.908 nm shown in XRD patterns (Fig. S13d) further confirmed that the MO ions had already intercalated into the LDH layers.



Figure S14. Molecular structures of (a) crystal violet and (b) methylene green. UV-vis absorption spectra of CV (25 mg/L) adsorbed by LDH coated membranes (c) with and (d) without the existence of MO (25 mg/L). UV-vis absorption spectra of MG (25 mg/L) adsorbed by LDH coated membranes (e) with and (f) without the existence of MO (25 mg/L).

Given that the electrostatic interaction between LDHs and anionic dyes was the critical driving force for the MO remediation,² the adsorption capability of LDHs for cationic dyes was expected to be undesirable (Fig. S14). When the MO was added into the solution containing the cationic dye, both the two organic pollutants could be readily removed by the LDH coated membranes (Table S5).



Figure S15. Removal rate of MO (200 mg/L) by the LDH coated membranes in different recycle runs.

In this work, desorption of the spent membrane was carried out in 1 mol/L $\rm Na_2SO_4$ solution to replace MO with $\rm SO_4^{2-}$



Figure S16. Treatment of MO solution by LDH coated membranes at normal pressure.



Figure S17. Removal rate of Cu²⁺ (100 mg/L) by the LDH coated membranes in different recycle runs.

The desorption of Cu^{2+} from the LDH coated membrane was performed by immersing the exhaust membranes into 0.01 mol/L CoSO₄ aqueous solution according to the method for metal unloading.³



Figure S18. Photograph of an underwater oil droplet (1,2-dichloroethane was selected so that oil droplet could sink down) on the surface of the LDH coated membranes.



Figure S19. Optical images of the MO degradation process with (left) and without (right) hybrid membrane at different times.

Samples Parameters	Blank	PDA/PEI coated	LDH coated
Specific surface area (m^2/g)	1.49	1.89	28.82
Pore diameter (nm)	—	—	33.77
Pore volume (cm^3/g)	—	_	0.2398

Table S1. Textural properties of the nascent silica membranes, membranes modified by PDA and PEI and membranes after hydrothermal treatment.

Samples Parameters	PDA/PEI coated	LDH coated
First-Order		
q _e (mg/g)	16.87	129.46
k ₁ (/min)	0.0458	0.0269
\mathbb{R}^2	0.9052	0.9802
Second-Order		
q _e (mg/g)	18.52	148.82
$k_2 (g/(mg \cdot min))$	0.0034	0.0002
\mathbb{R}^2	0.8597	0.9950
Elovich		
α (mg/g)	4.81	12.13
β (g·min/mg)	0.3382	0.0349
R ²	0.7447	0.9673

Table S2. Kinetic parameters of MO (100 mg/L at time zero) adsorption on prepared membranes at 30 °C.

In Table S2, q_e is the equilibrium sorption capacity, k_1 and k_2 are the pseudo-first-order rate constant and pseudo-second-order rate constant, respectively, and α and β are the initial adsorption rate and the desorption constant, respectively.

The adsorbed amount of MO at different times could be well fitted by using the pseudosecond-order model, indicating that the rate limiting step in the adsorption process was the chemisorption of MO on the LDH coated membranes.⁴

Samples Parameters	LDH coated	
$\begin{array}{l} \textbf{Langmuir model} \\ Q_{MAX}(mg/g) \\ K_{L}(L/mg) \\ R^{2} \end{array}$	702.83 0.035 0.9856	
$\label{eq:kappa} \begin{array}{l} {\bf Freundlichmodel} \\ {\bf K}_{\rm F}(mg/g\cdot(L/mg)^{1/n}) \\ 1/n \\ {\bf R}^2 \end{array}$	97.86 0.358 0.9032	

Table S3. Adsorption isotherm parameters of MO on prepared membranes at 30 °C.

In Table S3, Q_{MAX} is the maximum adsorption capacity, K_L is the constant related to the free energy of adsorption, K_F is the Freundlich isotherm constant and the 1/n is the adsorption intensity.

It was found that the adsorption of LDH coated membranes fitted the Langmuir model much better.

Q (mg/g) Refs Adsorbents 12 CA/MWCNTs fibers Carbohydrate Polymers J Mol Liq Synthetic Metals Grafted ficus carica fibers 51 59 PANI (skin)/PA6 (core) 91 J Hazard Mater PVA/PAA@PDA Co0.3Ni0.7Fe2O4@SiO2 116 J. Mater. Chem. A Fiber Polym PAN/PAMAM 120 PANF-g-HPEI 194 J Hazard Mater LDH coated membranes 702 this work

Table S4. Comparison of adsorption capacity of various adsorbents for MO.

Table S5. The removal rate of CV and MG by the LDH coated membranes with and without the existence of MO.

Removal rate	CV		MG	
Dyes (%)	No MO	With MO	No MO	With MO
CV MG MO	15.27 	67.89 		

Table S6. Kinetic parameters of Cu^{2+} (100 mg/L at time zero) adsorption on LDH coated membranes at 30 °C.

Samples Parameters	LDH coated
First-Order	
q _e (mg/g)	155.63
k_1 (/min)	0.0034
R ²	0.9796
Second-Order	
q _e (mg/g)	216.12
$k_2 (g/(mg \cdot min))$	0.0001
R ²	0.9862
Elovich	
α (mg/g)	2.6473
β (g·min/mg)	0.0319
R ²	0.9070

Table S7. Adsorption isotherm parameters of Cu^{2+} on LDH coated membranes at 30 °C.

Samples Parameters	LDH coated
Langmuir model	
$O_{MAX}(mg/g)$	285.28
K_{I} (L/mg)	0.081
R^2	0.9893
Freundlich model	
$K_{\rm r}$ (mg/g·(L/mg) ^{1/n})	59.79
1/n	0.307
\mathbf{R}^2	0.8793

Adsorbents	Q (mg/g)	Refs
Cl-LDHs	38	Chem. Eng. J
LS-LDHs	51	J. Colloid Interface Sci.
PEI nanofibers	71	J. Membr. Sci
PANI-PS	171	ACS Appl. Mater. Interface
MoS ₄ -LDHs	181	J. Am. Chem. Soc
LDH coated membranes	285	this work
Chitosan nanofibers	485	J. Membr. Sci
PVA/SiO ₂ nanofibers	504	Chem. Commun

Table S8. Comparison of adsorption capacity of various adsorbents for Cu^{2+} .

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