Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Electronic Supplementary Information

A vertically channeled lamellar membrane for molecular sieving of water from organic solvent

Xiaoting Li, Naixin Wang, Zheng Huang, Lilong Zhang, Ya-Bo Xie, Quan-Fu An, Shulan Ji*

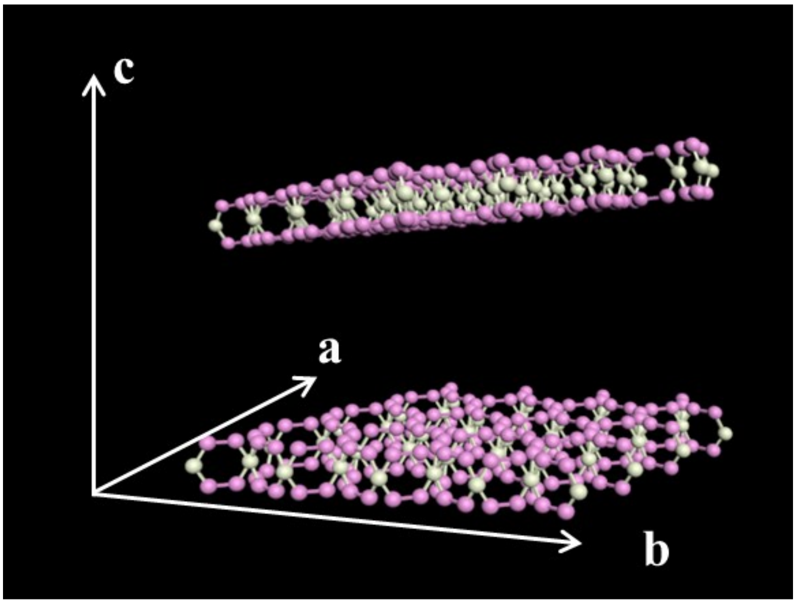

Figure S1. Structure of LDHs with reference.

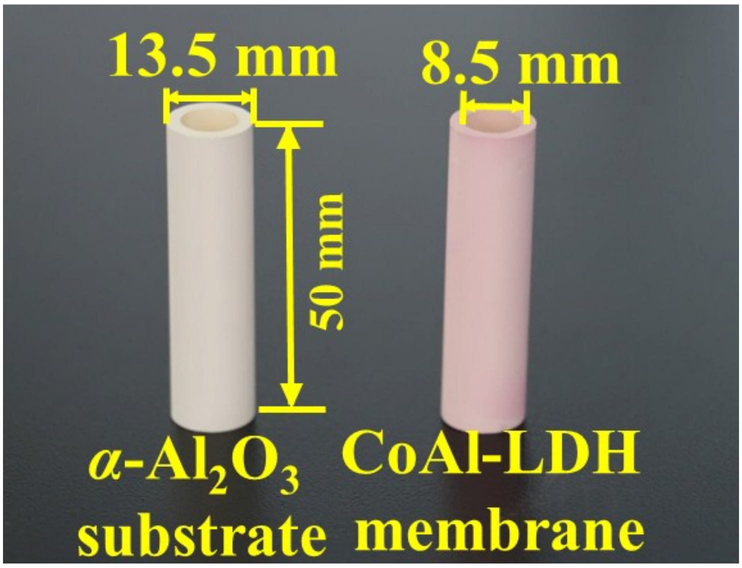

Figure S2. The photo of the α-Al_2O_3 substrate and the CoAl-LDH membrane.

$$Co^{2+} + xF^{-} \rightarrow CoF_{x}^{(x-2)-}$$

$$Al^{3+} + yF^{-} \rightarrow AlF_{y}^{(y-3)-}$$

$$H_2NCONH_2 + H_2O \rightarrow 2NH_3 + CO_2$$

$$CO_2 + H_2O \rightarrow CO_3^{2-} + 2H^{+}$$

$$NH_3 \cdot H_2O \rightarrow NH_4^{+} + OH^{-}$$

$$CoF_{x}^{(x-2)-} + 0.5(2-y)CO_3^{2-} + yOH^{-} + nH_2O \rightarrow Co(OH)_y(CO_3)_{0.5(2-y)} \cdot nH_2O + xF^{-}$$

$$AlF_{x}^{(x-3)-} + 0.5(3-y)CO_3^{2-} + yOH^{-} + nH_2O \rightarrow Al(OH)_y(CO_3)_{0.5(3-y)} \cdot nH_2O + xF^{-}$$

Figure S3. The reaction formulas with the assistance of F^- involved in the hydrothermal process.

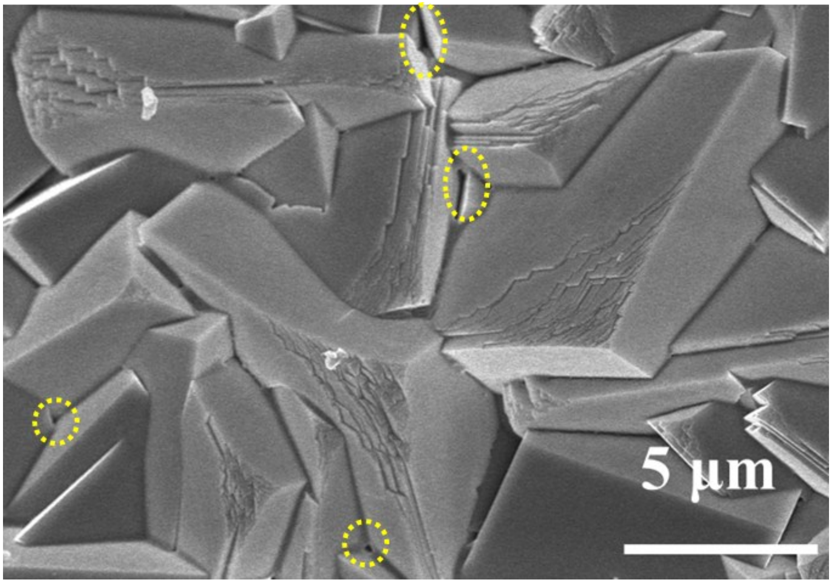

Figure S4. Surface morphology of CoAl-LDH composite membranes without NH_4F. (Preparation conditions: $Co(NO_3)_2 \cdot 6H_2O$, 21 mmol/L; $Al(NO_3)_3 \cdot 9H_2O$, 7 mmol/L; urea, 70 mmol/L; and NH_4F, 100 mmol/L; reaction temperature, 110 °C; reaction time, 24 h)

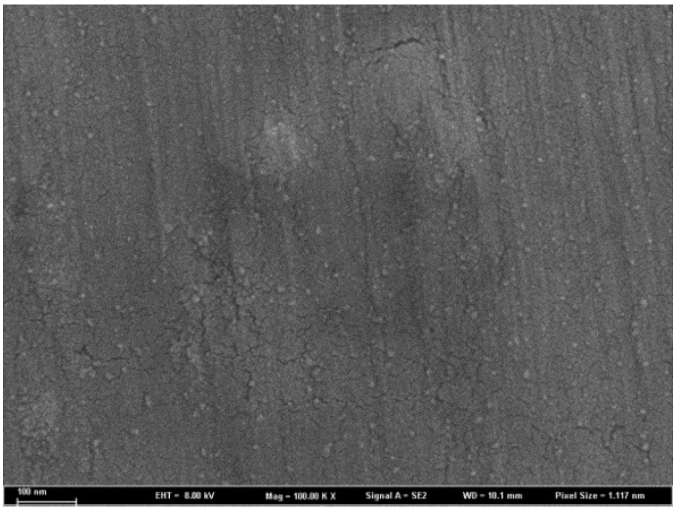

Figure S5. Surface morphology of CoAl-LDH composite membranes after wipe off. (Preparation conditions: $Co(NO_3)_2 \cdot 6H_2O$, 21 mmol/L; $Al(NO_3)_3 \cdot 9H_2O$, 7 mmol/L; urea, 70 mmol/L; and NH_4F, 100 mmol/L; reaction temperature, 110 °C; reaction time, 24 h)

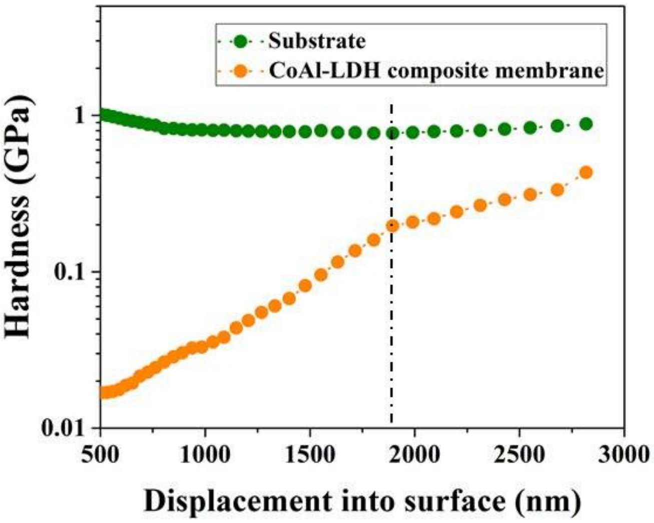

Figure S6. Hardness curves of the ceramic substrate and CoAl-LDH composite membrane with displacement into surface. (Preparation conditions: $Co(NO_3)_2 \cdot 6H_2O$, 21 mmol/L; $Al(NO_3)_3 \cdot 9H_2O$, 7 mmol/L; urea, 70 mmol/L; and NH_4F, 100 mmol/L; hydrothermal temperature, 110 °C; hydrothermal time, 24 h)

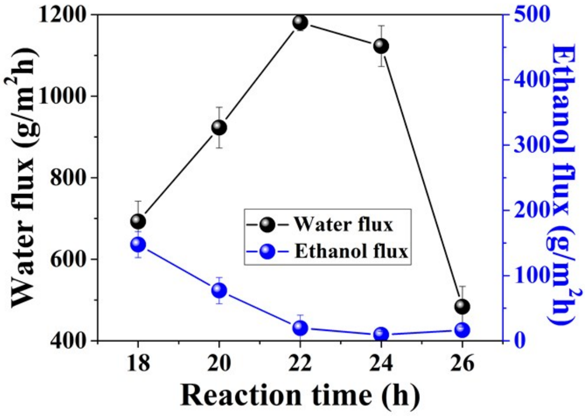

Figure S7. Effects of hydrothermal time on ethanol dehydration performance of the CoAl-LDH composite membranes.

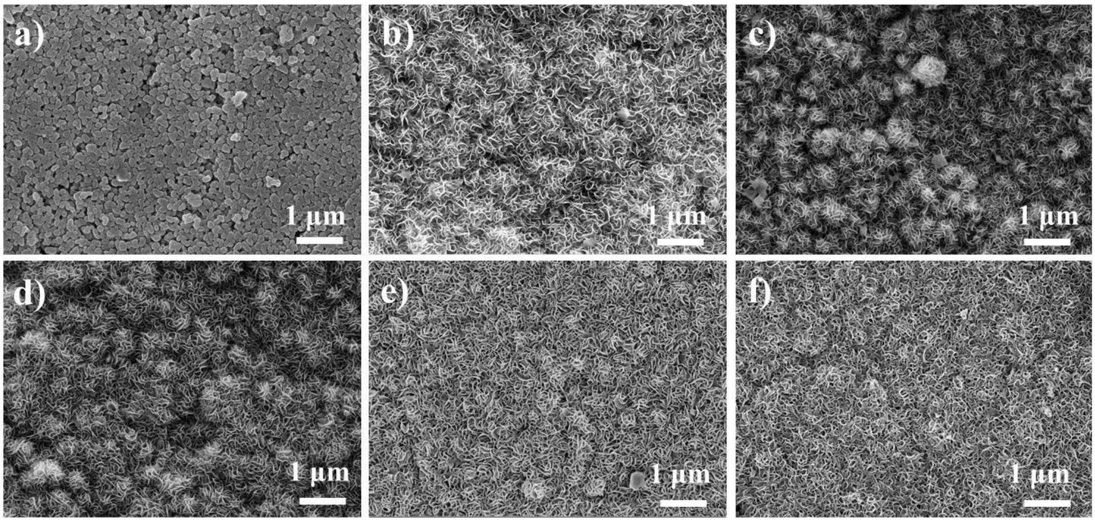

Figure S8. Surface SEM images of a) alumina substrate and CoAl-LDH membranes with different hydrothermal time: b) 18 h; c) 20 h; d) 22 h; e) 24 h; f) 26 h.

(Preparation conditions: $Co(NO_3)_2 \cdot 6H_2O$, 18 mmol/L; $Al(NO_3)_3 \cdot 9H_2O$, 6 mmol/L; urea, 60 mmol/L; and NH_4F, 100 mmol/L; hydrothermal temperature, 110 ^oC)

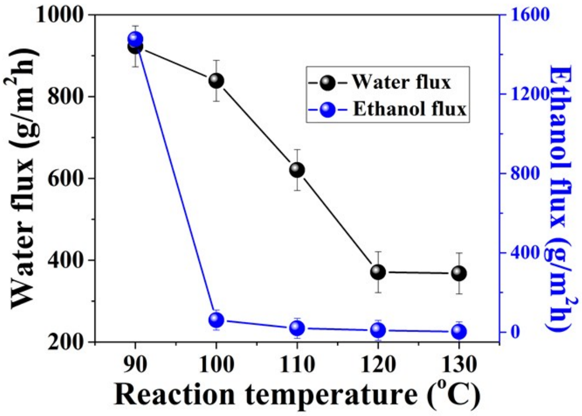

Figure S9. Effects of reaction temperature on ethanol dehydration performance of the CoAl-LDH composite membranes.

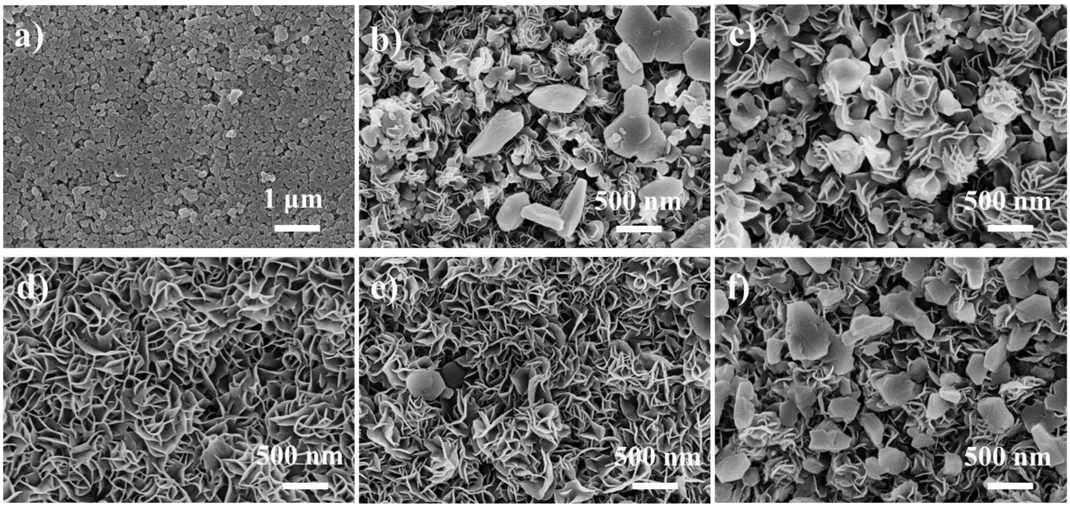

Figure S10. Surface SEM images of a) alumina substrate and CoAl-LDH membranes with different hydrothermal temperature: b) 90 °C; c) 100 °C; d) 110 °C; e) 120 °C; f) 130 °C. (Preparation conditions: $Co(NO_3)_2 \cdot 6H_2O$, 18 mmol/L; $Al(NO_3)_3 \cdot 9H_2O$, 6 mmol/L; urea, 60 mmol/L; and NH_4F, 100 mmol/L; hydrothermal time, 24 h)

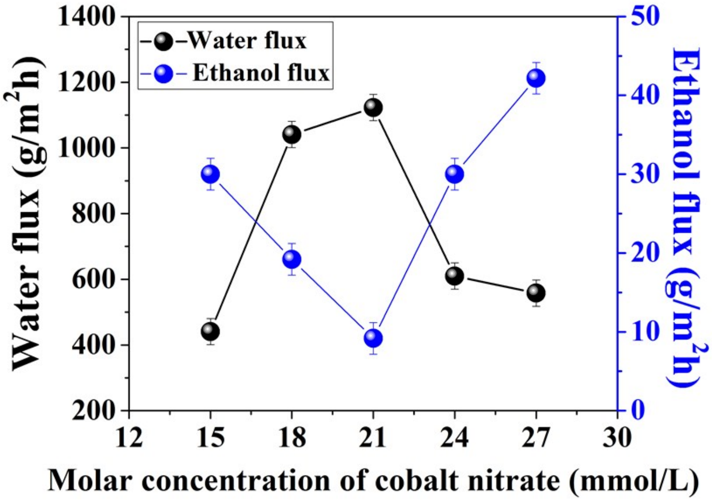

Figure S11. Effects of molar concentration of cobalt nitrate on ethanol dehydration performance of the CoAl-LDH composite membranes.

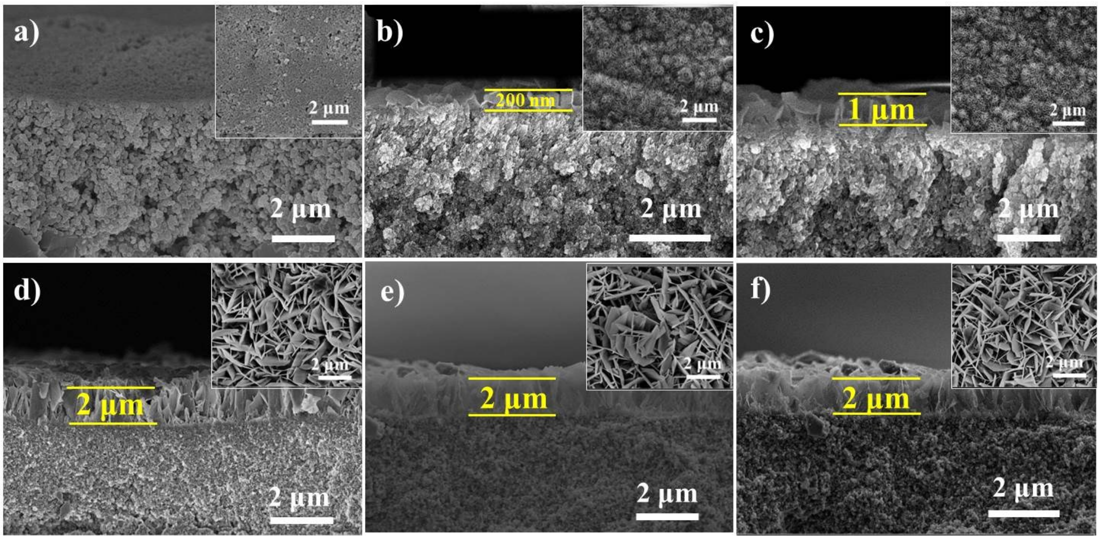

Figure S12. Cross-section and surface SEM images of a) alumina substrate and CoAl-LDH membranes with different molar concentration of cobalt nitrate: b) 15 mmol/L; c) 18 mmol/L; d) 21 mmol/L; e) 24 mmol/L; f) 27 mmol/L. (Preparation conditions: hydrothermal temperature, 110 ^oC; hydrothermal time, 24 h)

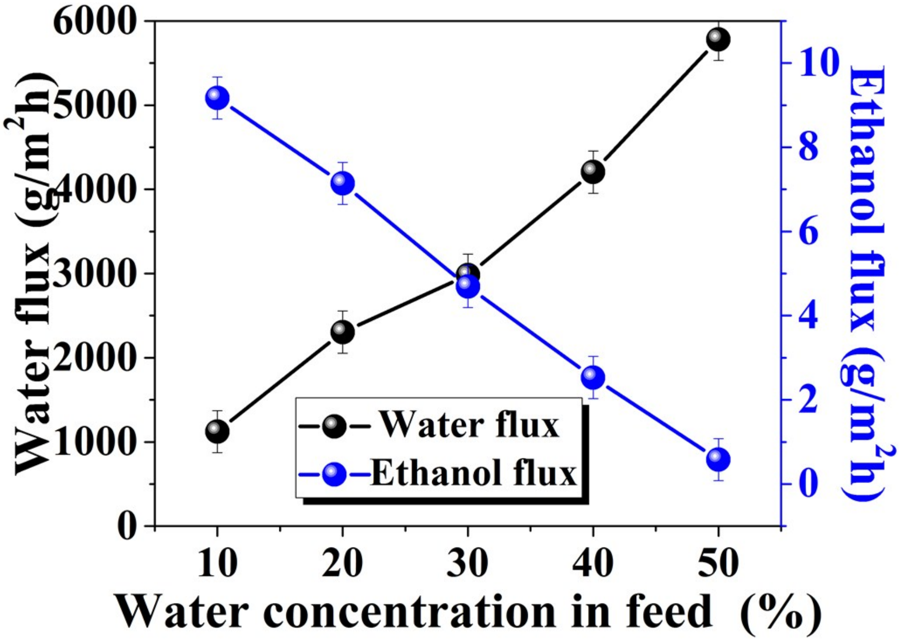

Figure S13. Ethanol dehydration pervaporation performance with water concentration in feed.

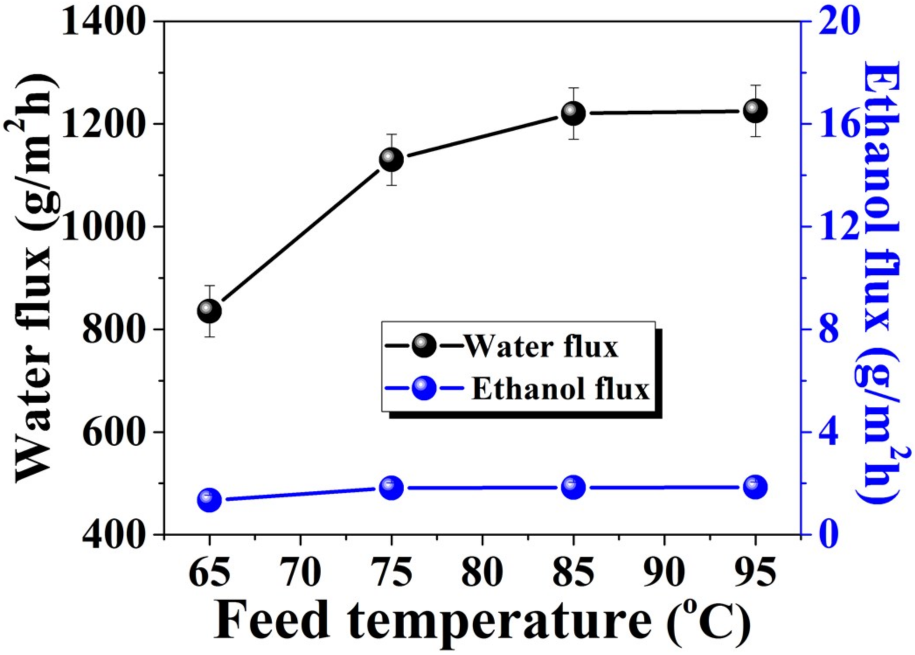

Figure S14. Ethanol dehydration pervaporation performance with feed temperature.

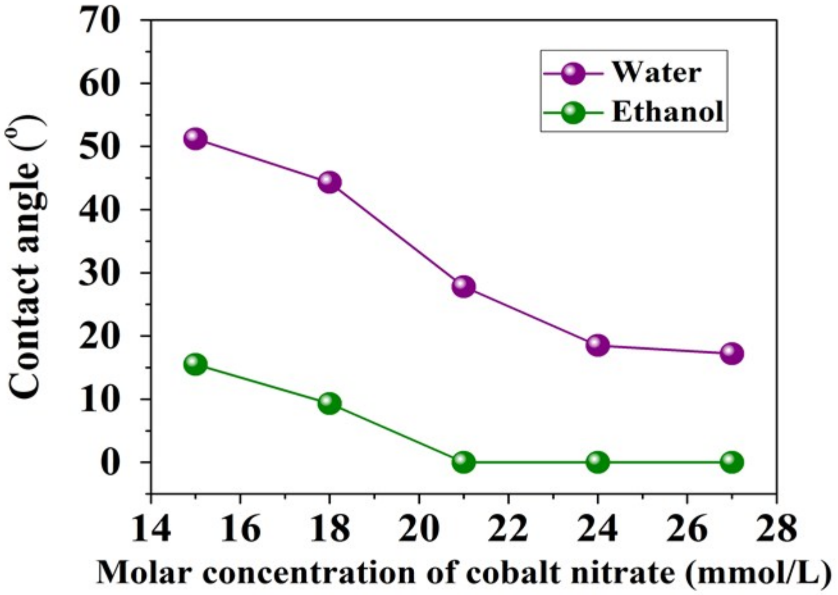

Figure S15. Static contact-angle of alumina substrate and CoAl-LDH composite membranes with different molar concentration.

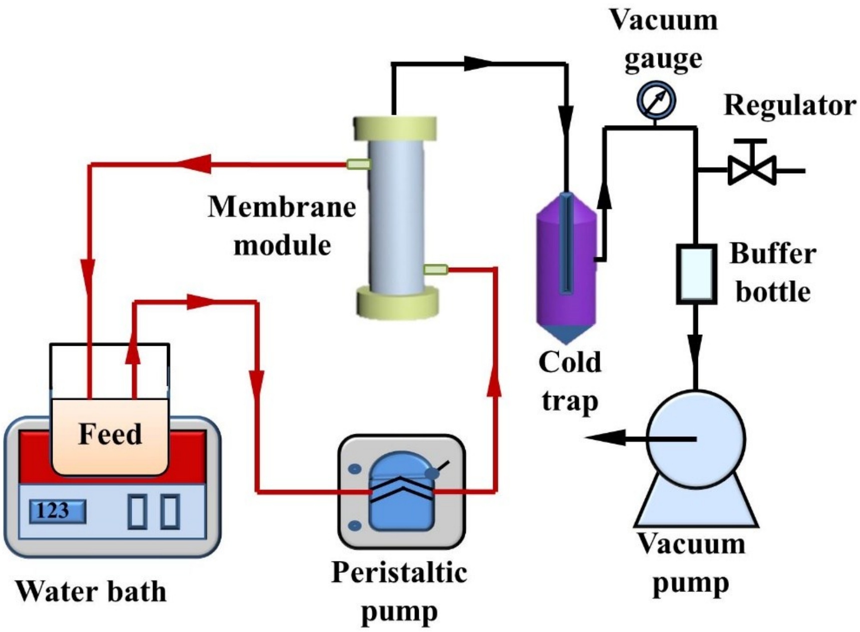

Figure S16. Schematic diagram of the pervaporation apparatus.

Table S1 Comparison with other membranes for water/organics separation

Membranes	Organic	Feed	Temperature	Flux	Water in	Ref

	solvents	content (wt.%)	(^{o}C)	(g/m^{2}h)	permeate (wt.%)	
PVA	Ethanol	90	40	410	98.2	[1]
HPA/SA	Ethanol	90	60	320	99.11	[2]
PSF	Ethanol	90	25	800	98.73	[3]
DETA/ TM*C*	Ethanol	90	25	1220	98.4	[4]
MPD/TMC	Ethanol	85	50	1288	87.6	[5]
PEI/ PVS$_{60}$	Ethanol	79.4	58.5	600	99.5	[6]
Nexar^{TM}/ PEI	Ethanol	85	50	1160	95.8	[7]
(PAA/PEI)$_4$	Ethanol	95	40	140	98.45	[8]
m-Tolidine-H-TMC/ PAN	Ethanol	90	25	2191	99.5	[9]
TDI cross-MPD/TMC	Ethanol	85	50	2000	95.8	[10]
PVA/NR/ zeolite 4A	Ethanol	95	30	3600	94	[11]
Poly(acrylonitrile butyl acrylate)	Acetic acid	99.5	30	3970	99.84	[12]
PVA/ zeolite 4A	Ethylene glycol	80	70	2800	99.8	[13]
CS/ZIF-7	Ethanol	90	25	1206	98.35	[14]
CS/ OAS POSS	Ethanol	90	30	37	95.7	[15]
Ultem^{R}/polyimide/ POSS	Ethanol	90	60	1800	94.7	[16]
Cu$_3$(BTC)$_2$	Isopropanol	90	50	400	96.46	[17]
P84/ Zeolite 13X	Isopropanol	90	60	110	99.67	[18]
PBI/ZIF-8	Isopropanol	85	60	103	99.6	[19]

PERVAP^{R}2510 Dense	Isopropanol	85	60	64.4	99.8	[20]
Zeolite T	Ethanol	90	65	1770	99.2	[21]
Zeolite T	Isopropanol	90	65	2150	100	[21]
Zeolite T	Isopropanol	90	75	1100	99	[22]
Zeolite T	Ethanol	90	75	2200	100	[22]
Zeolite T	Ethanol	90	75	2120	99.3	[23]
Zeolite T	Ethanol	90	75	2520	100	[23]
NaA/PES-PI	Isopropanol	90	75	11100	100	[24]
NaA/PES-PI	Ethanol	90	75	10600	100	[25]
NaY/ ceramic	Isopropanol	90	75	2500	97.5	[25]
NaY/ ceramic	*n*-Butanol	95	75	2000	98.75	[25]
NaX	Ethanol	90	75	1900	95	[25]
LDH	Ethanol	90	75	1132	99.19	This work
LDH	Propanol	90	75	3484	100	This work
LDH	Isopropanol	90	75	2580	100	This work
LDH	*n*-Butanol	90	75	3200	100	This work
LDH	Isobutanol	90	75	3300	100	This work
LDH	Ethyl acetate	90	75	4640	100	This work

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