Supporting Information for

Green *in-situ* synthsis of ZIF-8 membranes on the inner-surface of PESf hollow fibers and application in hydrogen separation

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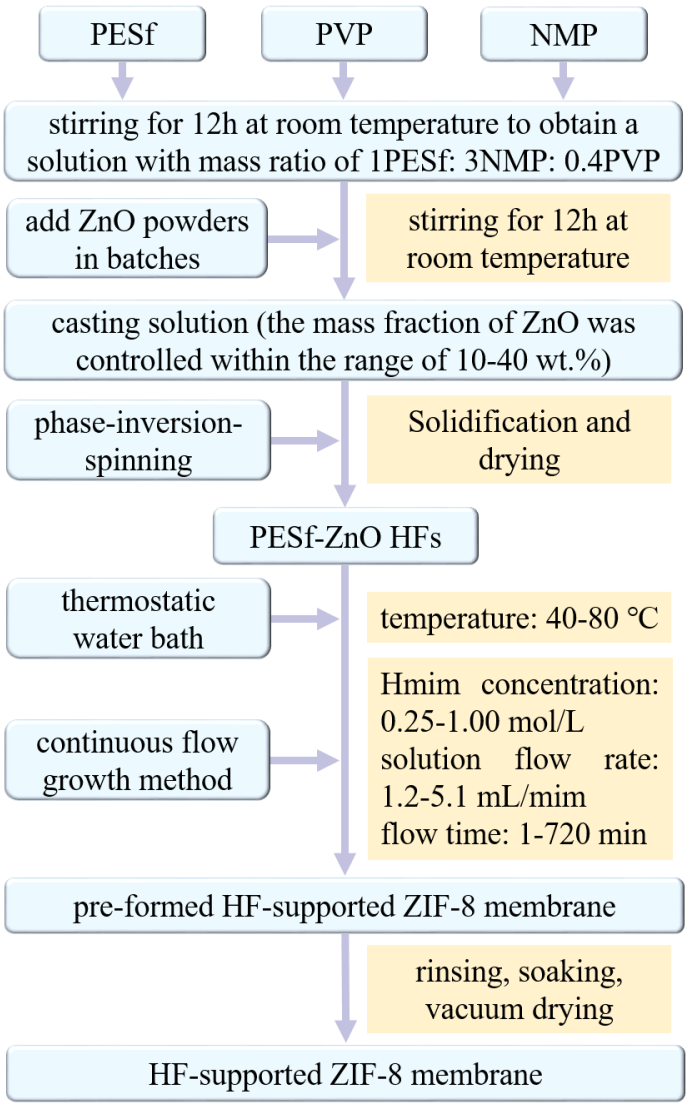
Table S4 Detailed comparison of H_2/CH_4 and H_2/N_2 separation performance of membranes listed in Fig. 9.



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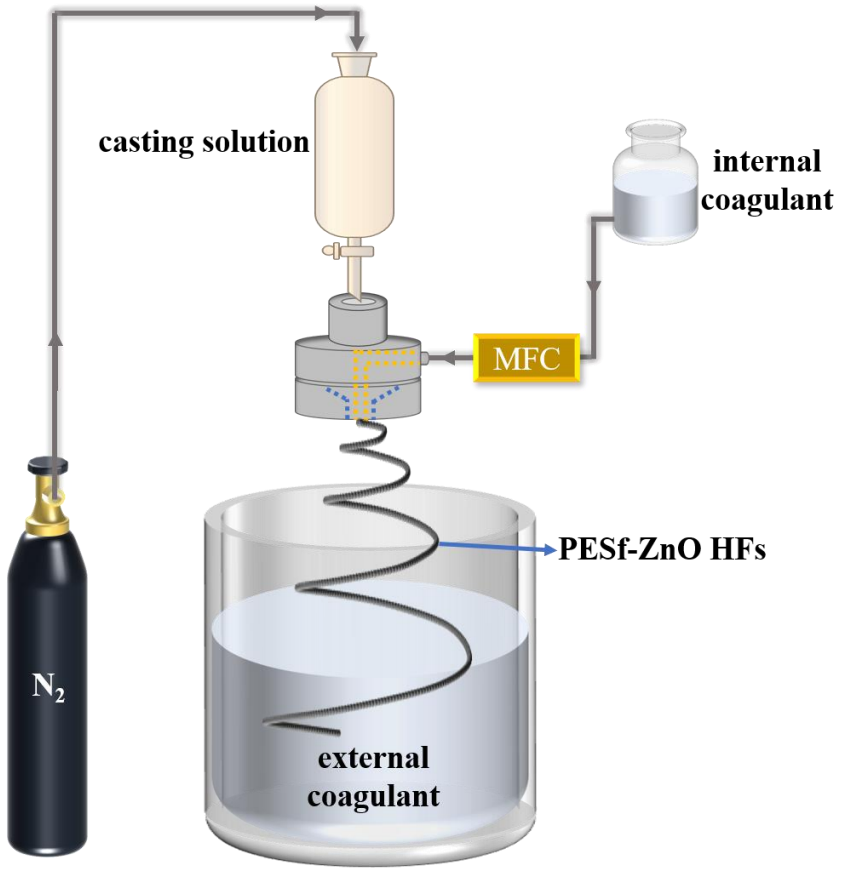


Fig. S2 Schematic diagram of HF membrane prepared by phase-inversion-spinning technology.

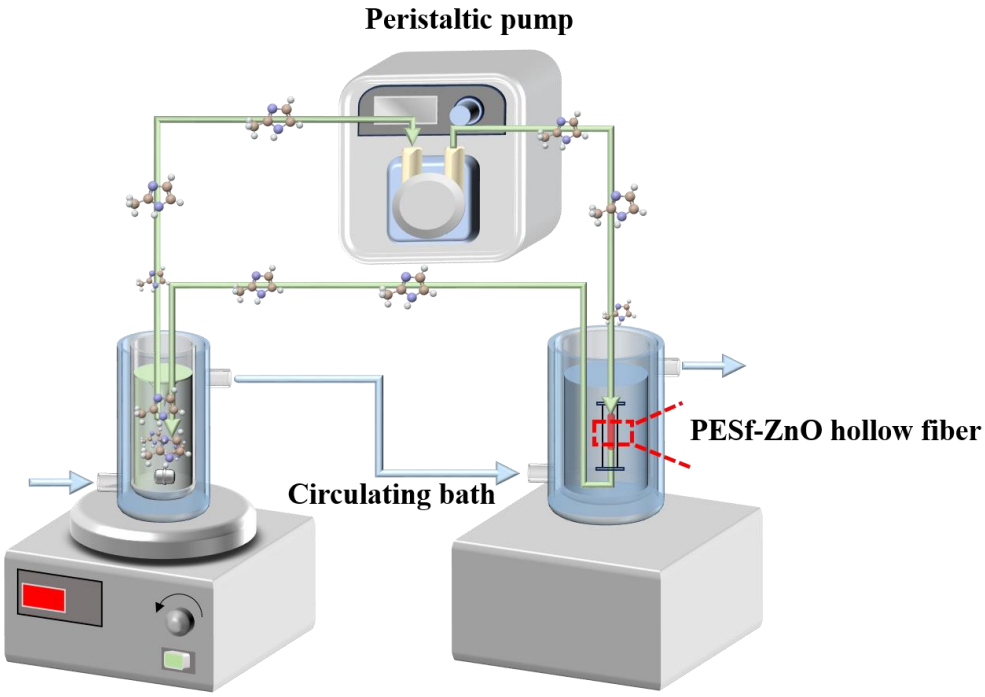


Fig. S3 Schematic drawing of preparation process for ZIF-8 membrane.

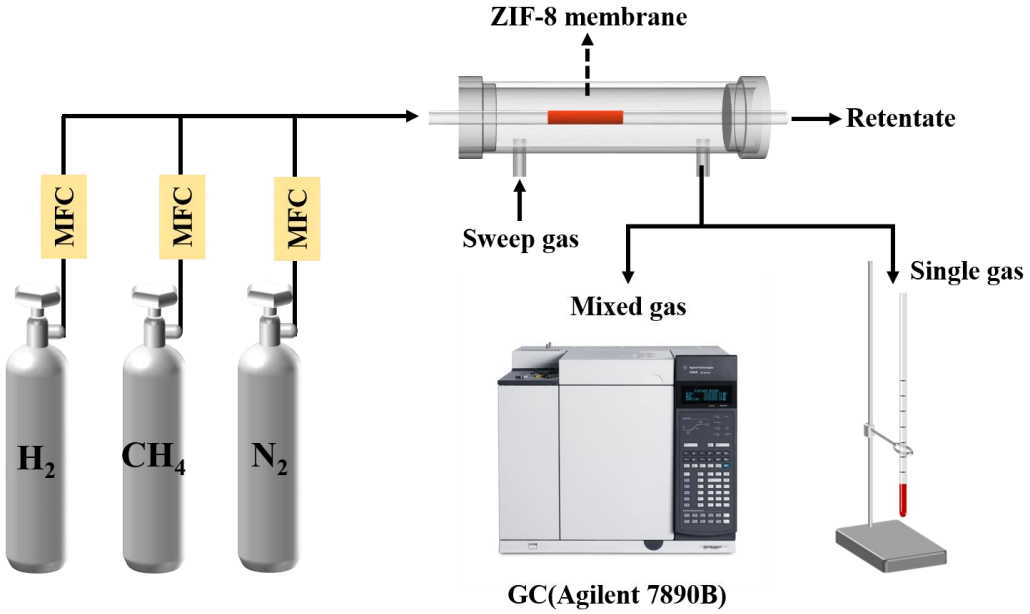


Fig. S4 Schematic diagram of gas permeation device.

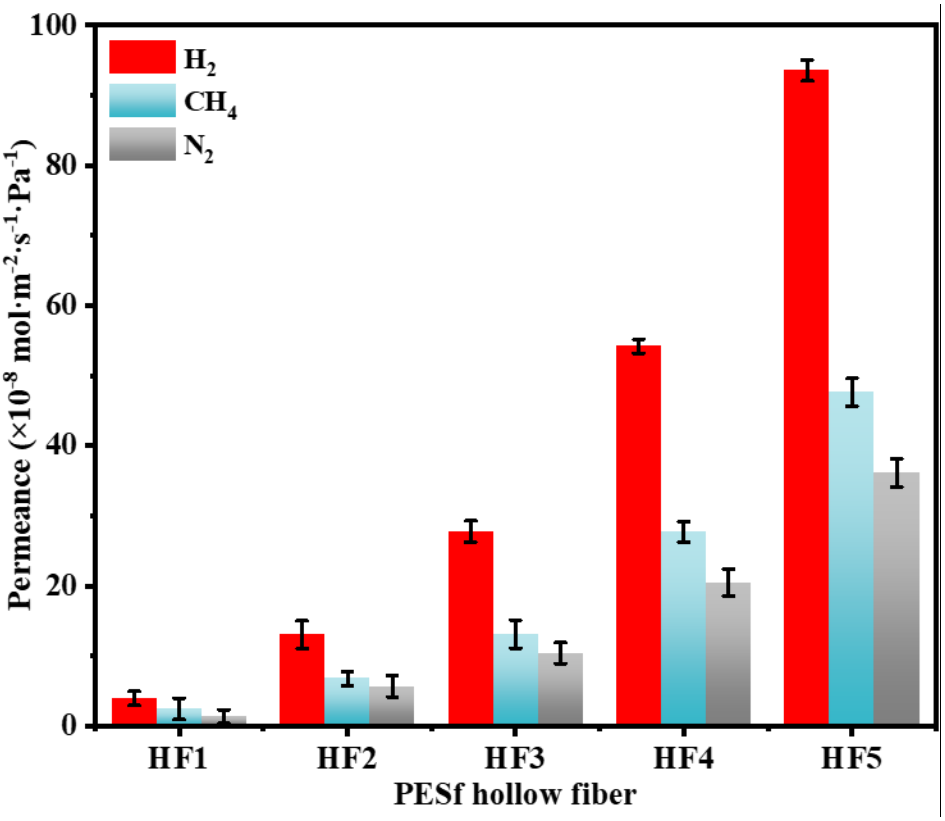
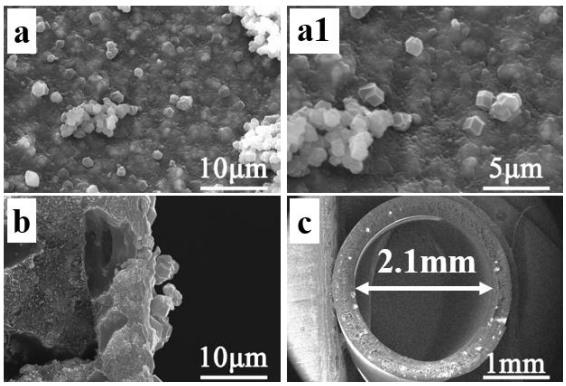


Fig. S5 Gas permeances for PESf hollow fibers.



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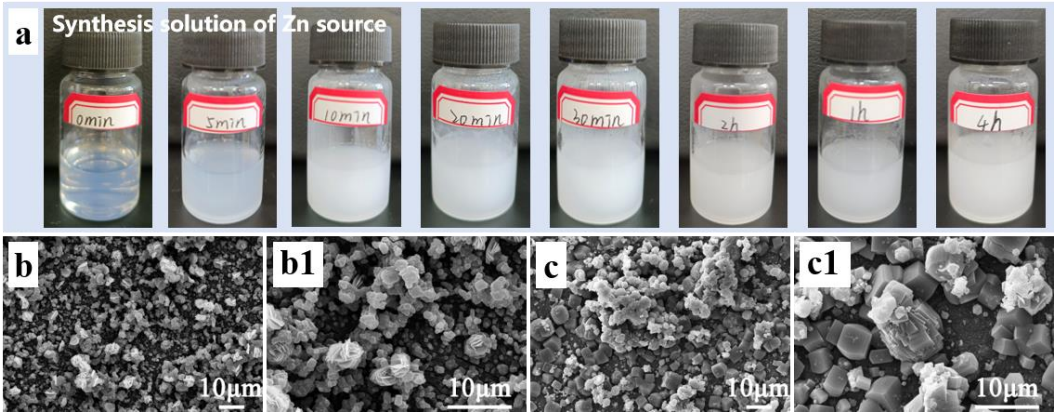

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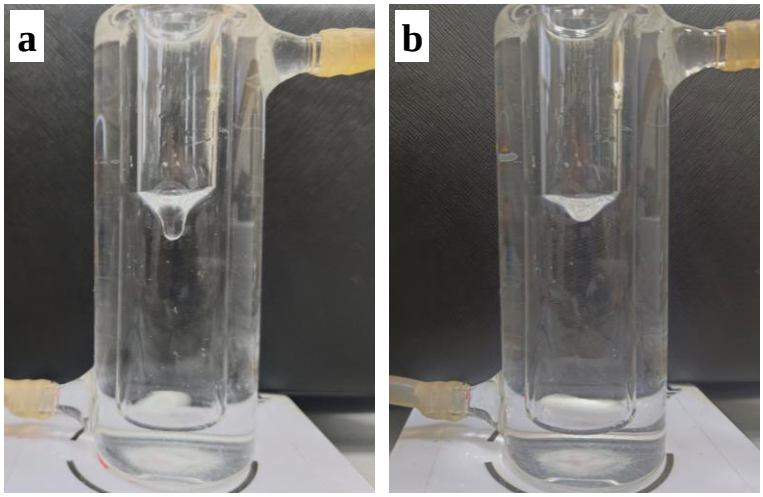
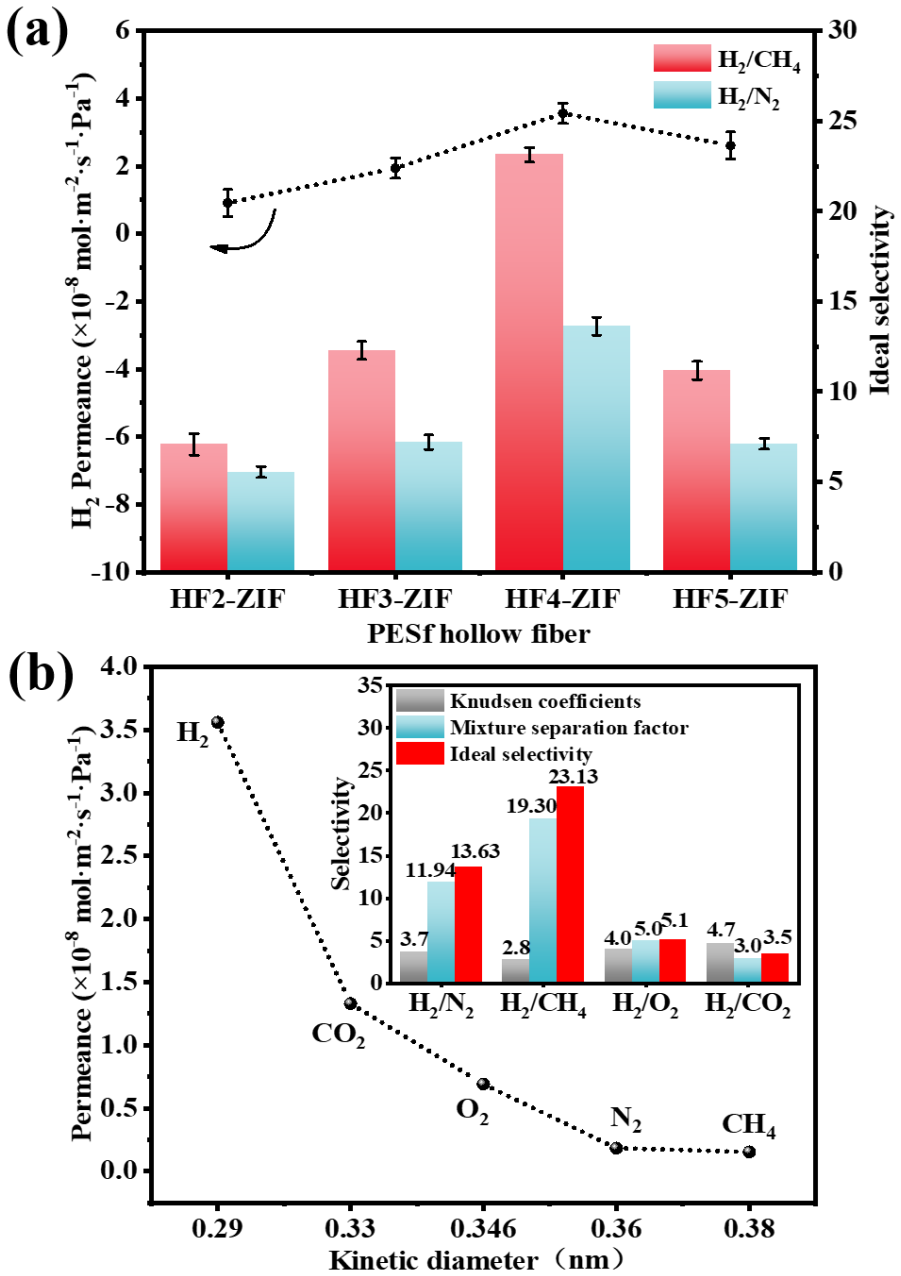


Fig. S8 Photographs of the synthetic solution at the beginning (a) and end (b) of the flow process.



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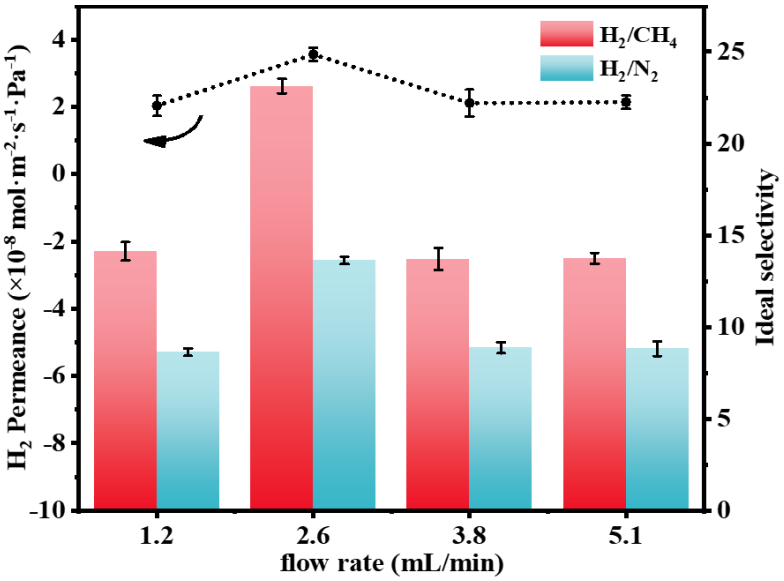


Fig. S10 Gas permeation data of ZIF-8 membranes prepared at different flow rates.

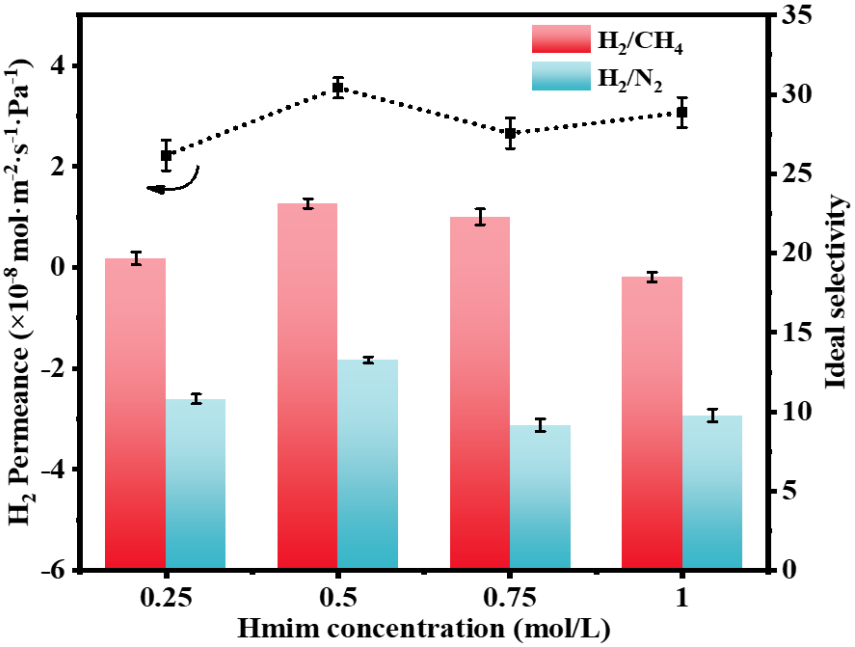


Fig. S11 Gas permeation data of ZIF-8 membranes prepared at different Hmim concentrations.

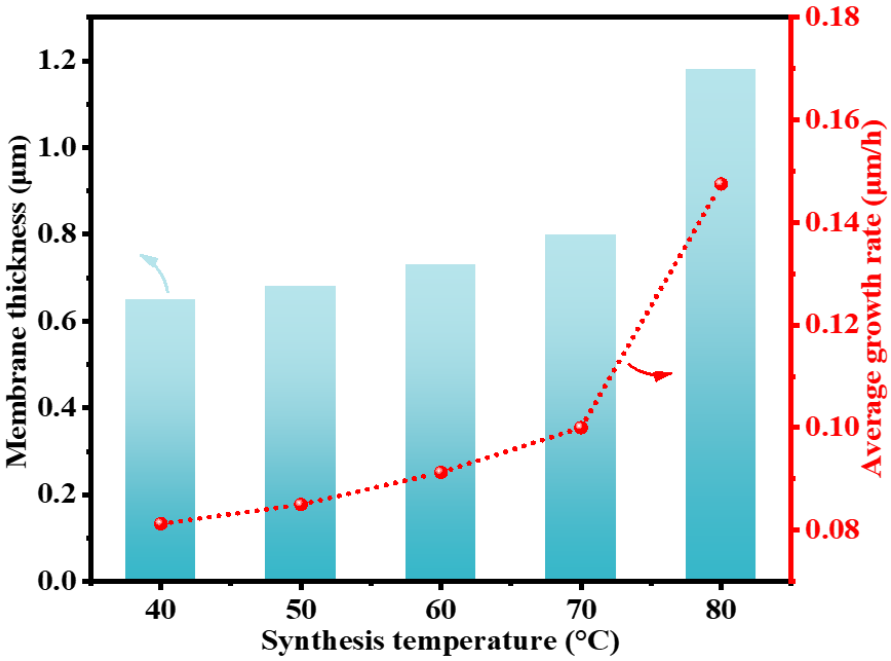
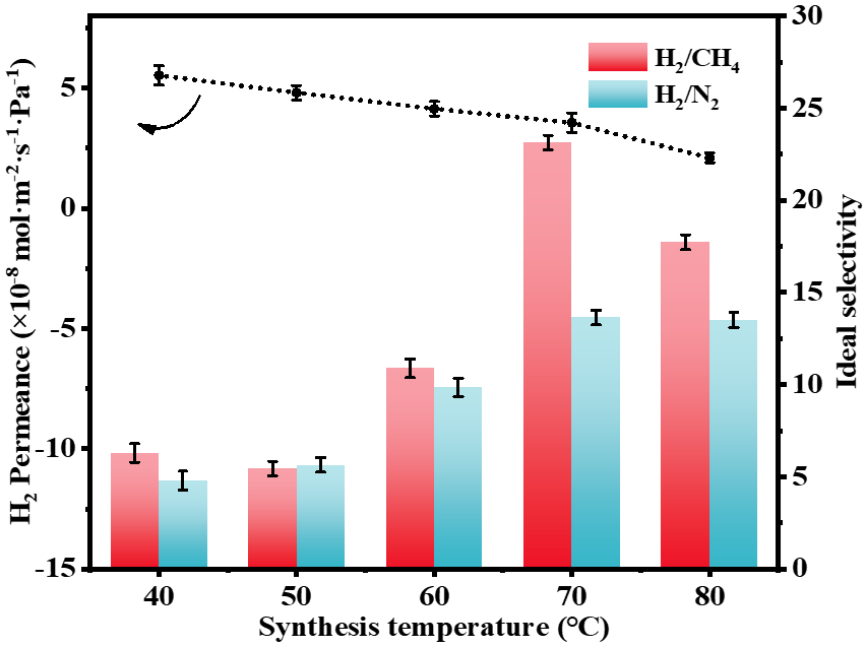


Fig. S12 The thickness and average growth rate of ZIF-8 membranes prepared at different synthesis temperature.



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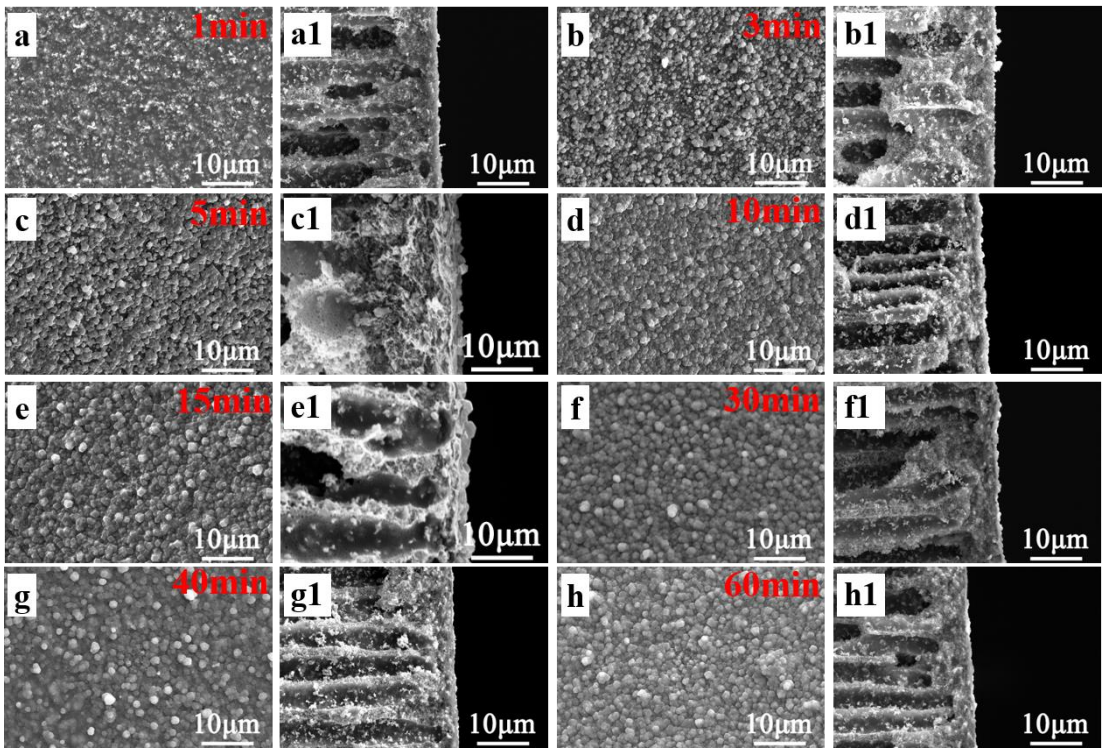
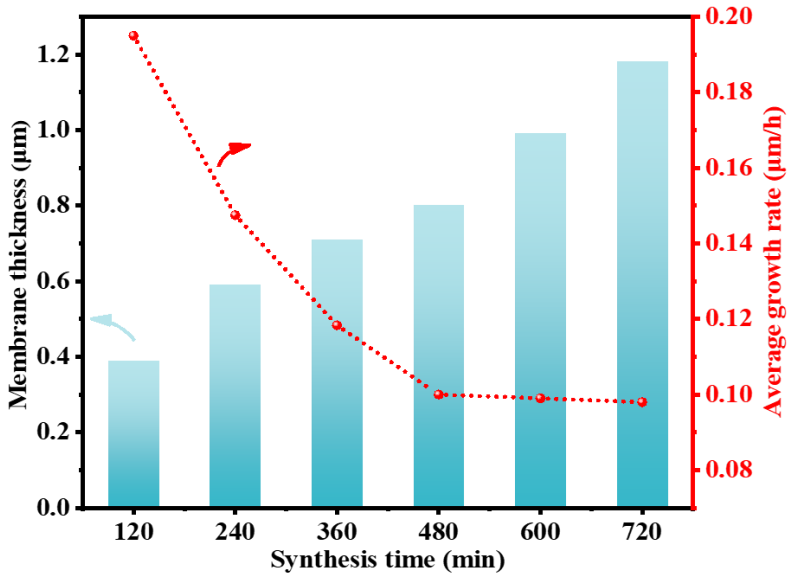


Fig. S14 SEM images of ZIF-8 membranes prepared under different synthesis time at 70 °C (a, a1: 1 min; b, b1: 3 min; c, c1: 5 min; d, d1: 10 min; e, e1: 15 min; f, f1: 30 min; g, g1: 40 min; h, h1: 60 min; a, b, c, d, e, f, g, h: surface morphology; a1, b1, c1, d1, e1, f1, g1, h1: cross-section view).



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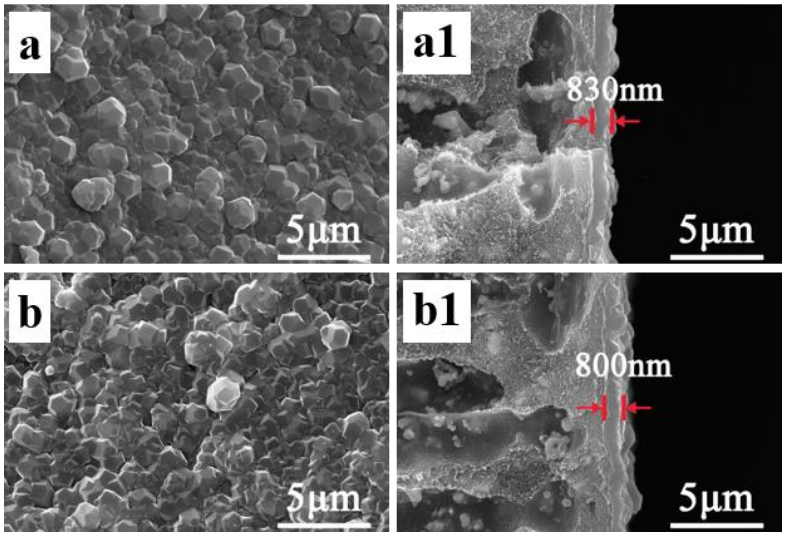

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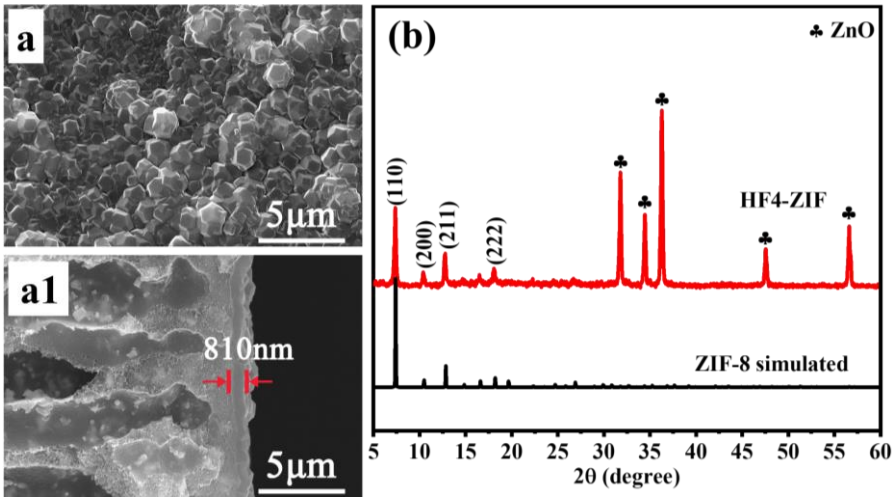

Fig. S17 SEM images (a: surface morphology; a1: cross-section view) and XRD patterns (b) of ZIF-8 membrane after thermal and long-term stability tests.

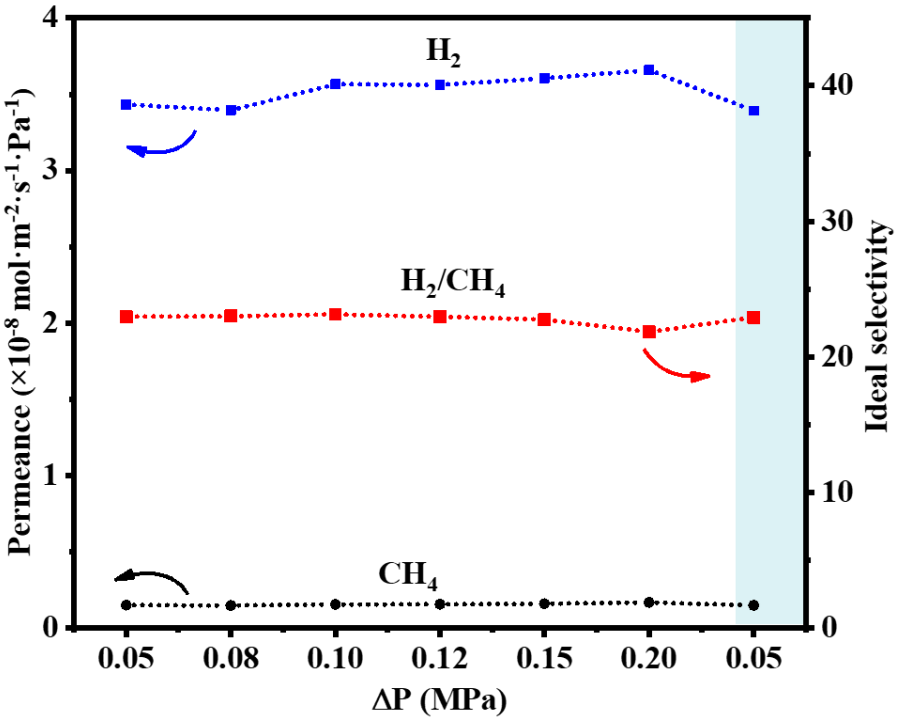

Fig. S18 Gas permeances as a function of pressure difference.

Table S1 ZIF-8 membranes prepared by different synthesis methods.

Preparation method	**Support**	**The composition of synthetic solution (molar ratio)**	**Metal source in the solution**	**Alkaline additives in the solution**	**Solvents**	**Coupling agents or other reagents**	**Synthesis temperature**	**Synthesis time**	**Reaction vessel**	**Ref.**
In-situ solvothermal synthesis	α-Al_2O_3 disc	1$ZnCl_2$: 8.3Hmim: 5.6HCOONa: 250MeOH	$ZnCl_2$ (0.55 g)	HCOONa (1.43 g)	MeOH (40 mL)	-	120 °C	4 h	Teflon-lined stainless steel autoclave	[1]
	PDA-modified α-Al_2O_3 disc	1$ZnCl_2$: 2Hmim: 1HCOONa: 313MeOH	$ZnCl_2$ (0.538 g)	HCOONa (0.268 g)	MeOH (50 mL)	Dopamin (2 mg/mL); Tris-HCl (10 mM)	85 °C	24 h	Teflon-lined stainless steel autoclave	[2]
	PDA-functionalized stainless-steel-net (SSN)	1$ZnCl_2$: 2Hmim: 1HCOONa: 313MeOH	$ZnCl_2$ (0.538 g)	HCOONa (0.268 g)	MeOH (50 mL)	Dopamine (2 mg/mL); Tris-HCl (10 mM)	85 °C	24 h	Teflon-lined stainless steel autoclave	[3]
	PDA-functionalized stainless-steel-net (SSN)	1$ZnCl_2$: 2Hmim: 1HCOONa: 313MeOH	$ZnCl_2$ (0.538 g)	HCOONa (0.268 g)	MeOH (50 mL)	Dopamine (2 mg/mL); Tris-HCl (10 mM)	85 °C	48 h	Teflon-lined stainless steel autoclave	[4]
Secondary seeding growth method	Hollow ceramic fiber	1$ZnCl_2$: 2.1Hmim: 1HCOONa: 306MeOH	$ZnCl_2$ (0.55 g)	HCOONa (0.272 g)	MeOH (50 mL)	C_3H_6O	120 °C	8 h	Teflon-lined stainless steel autoclave	[5]

	α-Al_2O_3 disc	1$Zn(NO_3)_2·6H_2O$: 74.5Hmim: 6000H2O	$Zn(NO_3)_2·6H_2O$ (0.11 g)	-	H_2O (40 mL)	MeOH	90 °C	24 h	Autoclave	[6]
	Al_2O_3 hollow fiber	1$ZnCl_2$: 1.5Hmim: 1.5HCOONa: 250MeOH	$ZnCl_2$ (0.674 g)	HCOONa (0.506 g)	MeOH (50 mL)	-	100 °C	2 h	Teflon-lined stainless steel autoclave	[7]
	ZIF-8-PVDF/PVDF dual-layer polymeric hollow fiber	1$ZnCl_2$: 1.5Hmim: 1.5HCOONa: 250MeOH	$ZnCl_2$	HCOONa	MeOH	DMAc (N, N dimethylacetamide); PEG 400 (polyethylene glycol 400)	100 °C	2.5 h	Teflon-lined stainless steel autoclave	[8]
	Zn foam substrates	1$Zn(NO_3)_2·6H_2O$: 4Hmim: 2HCOONa: 451MeOH	$Zn(NO_3)_2·6H_2O$ (0.9766 g)	HCOONa (0.4477 g)	MeOH (60 mL)	CH_3CH_2OH	25 °C	24 h	Beaker	[9]
Plasma-assisted synthesis	α-Al_2O_3 disc	1$ZnCl_2$: 2Hmim: 1.4HCOONa: 313MeOH	$ZnCl_2$ (0.538 g)	HCOONa (0.3694 g)	MeOH (50 mL)	-	85 °C	24 h	Teflon-lined stainless steel autoclave	[10]
Electrochemical deposition	Polyamide	1$Zn(OAc)_2$: 2Hmim: 500MeOH	$Zn(OAc)_2$ (0.827 g)	-	MeOH (90 mL)	-	25 °C	15-75 min	Electrolytic cell	[11]
	Polypropylene support	1$Zn(OAc)_2·2H_2O$: 60Hmim: 53H_2O	$Zn(OAc)_2·2H_2O$ (0.18 g)	-	H_2O (80 mL)	-	25 °C	1 h	Electrolytic cell	[12]
Contra-diffusion growth	α-Al_2O_3 disc	A: 1$ZnCl_2$: 137MeOH B: 1Hmim: 0.12HCOONa: 15.6MeOH	$ZnCl_2$ (0.98 g)	HCOONa (0.5 g)	MeOH (80 mL)	-	B: 120 °C	A: 1 h B: 4 h	Teflon-lined stainless steel autoclave	[13]

	Porous nylon	1$Zn(NO_3)_2{\cdot}6H_2O$: 2Hmim: (8-64)$NH_3{\cdot}H_2O$: 2200H_2O	$Zn(NO_3)_2{\cdot}6H_2O$ (0.147 g)	-	H_2O (40 mL)	$NH_3{\cdot}H_2O$	22±2 °C	48 h	Home-made setup	[14]
Current-driven synthesis process	AAO	1$Zn(OAc)_2{\cdot}2H_2O$: 2Hmim: 291MeOH: (0.1-0.4)dclm: 129DMF	$Zn(OAc)_2{\cdot}2H_2O$ (0.66 g)	-	MeOH (60 mL); DMF (N, N-dimethylformamide, 30 mL)	CH_3COOH	25 °C	20 min	Electrolytic cell	[15]
Gel–vapour deposition	PVDF hollow fiber	Hmim	-	-	-	CH_3CH_2OH; C_2H_7NO; PEG (poly(ethylene glycol)); DMAc (N,N-dimethylacetamide)	150 °C	6 h	Teflon-lined stainless steel autoclave	[16]
Interfacial synthesis	PES supports	A: $Zn(NO_3)_2{\cdot}6H_2O$ in H_2O B: Hmin in 1-Octanol	$Zn(NO_3)_2{\cdot}6H_2O$	-	H_2O; 1-Octanol	PEG; NMP; C_6H_{14}	25 °C	0.5-4 h	-	[17]
Interfacial microfluidic processing	PSf hollow fiber	$Zn(NO_3)_2{\cdot}6H_2O$ (0.1 mol/L) solution with a Hmim (0.3 mol/L) and HCOONa solution in MeOH	$Zn(NO_3)_2{\cdot}6H_2O$ (0.357 g)	HCOONa	MeOH (12 mL)	-	25 °C	2.5 h	Polypropylene syringes	[18]

Microwave assisted heating	α-Al_2O_3 hollow fiber	1$ZnCl_2$: 2Hmim: 1.3HCOONa: 323MeOH	$ZnCl_2$ (2.39 g)	HCOONa (1.49 g)	MeOH (180 mL)	-	140 °C	5 h	Teflon vessel	[19]
	TiO_2 disc	1 $ZnCl_2$: 1.5 Hmim: 1 HCOONa: 249.5 MeOH	$ZnCl_2$ (1.078 g)	HCOONa (0.54 g)	MeOH (80 mL)	-	100 °C	4 h	Teflon-lined stainless steel autoclave	[20]
Semisolid processing method	Al_2O_3 disc	1 $Zn(OAc)_2{\cdot}2H_2O$: 2: Hmim: 18 DMAc: 46 H_2O	$Zn(OAc)_2{\cdot}2H_2O$ (1.32 g)	-	DMAc (10 mL); H_2O (5 mL)	MeOH; $CHCl_3$	200 °C	15 min	-	[21]
Homologous metal induction	PESf hollow fiber	1$Zn(NO_3)_2{\cdot}6H_2O$: 1.5Hmim: 0.65HCOONa: 450MeOH	$Zn(NO_3)_2{\cdot}6H_2O$	HCOONa	MeOH	NMP	60 °C	6 h	Teflon-lined stainless steel autoclave	[22]
	Al_2O_3-ZnO hollow fiber	1$Zn(NO_3)_2{\cdot}6H_2O$: 1.4Hmim: 1HCOONa: 448MeOH	$Zn(NO_3)_2{\cdot}6H_2O$ (2 g)	HCOONa (0.46 g)	MeOH (122 mL)	CH_3CH_2O; NMP	100 °C	8 h	Teflon-lined stainless steel autoclave	[23]
	α-Al_2O_3 disc	Hmim solution in DI water (MeOH; DMF; DI water–MeOH mixtures) (0.691 mol/L)	$Zn(NO_3)_2{\cdot}6H_2O$	-	DI water (MeOH; DMF; DI water–MeOH mixtures)	HNO_3	30-90 °C	12 h (48 h)	-	[24]
	ZnO hollow fiber	1$Zn(NO_3)_2{\cdot}6H_2O$: 1.5Hmim: 0.65HCOONa: 450MeOH	$Zn(NO_3)_2{\cdot}6H_2O$ (2 g)	HCOONa (0.46 g)	MeOH (122.5 mL)	NMP	80 °C	8 h	Teflon-lined stainless steel autoclave	[25]

In-situ synthesis	PESf hollow fiber	1Hmim: 111H_2O	-	-	H_2O (50 mL)	NMP	70 °C	8 h	Glass container	This work

Table S2 Hazard codes and boiling points of solvents according to the "ECHA C&L Inventory database".

Solvent	Hazard codes	Boiling point (°C)
N, N-Dimethylformamide (DMF)	H302, H312, H315, H332, H319	153
N, N-Dimethylacetamide (DMAc)	H360D, H312, H332	165
N-Methyl Pyrrolidone (NMP)	H315, H319, H335, H360D	203
Methanol (MeOH)	H225, H301, H311, H331, H370	64.5
Ethanol (EtOH)	H225	78.3
1-Octanol ($C_8H_{18}O$)	H302, H315, H319, H332, H335	195
Acetone (C_3H_6O)	H225, H315, H319, H336	56
Zinc nitrate ($Zn(NO_3)_2·6H_2O$)	H272, H302, H315, H319	105-131
Zinc chloride ($ZnCl_2$)	H314, H302, H400, H410	732
Zinc acetate ($Zn(OAc)_2$)	H302, H318, H319, H335, H411	330
Zinc acetate dihydrate ($Zn(OAc)_2·H_2O$)	H302, H315, H319, H335, H4110	242-244

2-Methylimidazole (Hmim)	H302, H314, H360, H371, H373, H341, H351	267-268
4, 5-dichloroimidazole (dclm)	H315, H319, H335	334.1±22
Sodium formate (HCOONa)	H319, H336	360
Dopamine (DPA)	-	337.7
Tris Hydrochloride (Tris HCl)	H315, H319, H335	225
polyethylene glycol (PEG)	H302, H3115, H319, H335	200-250
Ethanolamine (C_2H_7NO)	H302, H312, H314, H332, H335	170
Ammonium hydroxide ($NH_3 \cdot H_2O$)	H314, H318, H335, H400	36
Hexane (C_6H_{14})	H225, H315, H336, H304, H373, H361e, H411	69
Chloroform ($CHCl_3$)	H331, H302, H315, H319, H351, H372, H361d	61.3
Acetic acid (CH_3COOH)	H226, H314, H315, H319	117-118
Nitric acid (HNO_3)	H272, H314, H330	83

Table S3 Residence time corresponding to different flow rates.

The speed of pump (RPM)	Flow rate of solution (mL/min)	Residence time (min)
5	1.2	2.6
10	2.6	1.2
15	3.8	0.8
20	5.1	0.6

Table S4 Detailed comparison of H_2/CH_4 and H_2/N_2 separation performance of membranes listed in Fig. 9.

Membrane	**Support**	**Thickness (μm)**	**H_2 permeance ($mol \cdot m^{-2} \cdot s^{-1} \cdot Pa^{-1}$)**	**H_2/CH_4 selectivity**	**H_2/N_2 selectivity**	**Synthetic method; solvent**	**Stability**	**Ref.**
ZIF-8	PESf hollow fiber	0.8	3.56×10^{-8}	23.1	13.6			This work
	α-Al_2O_3 disc	25	2.4×10^{-7}	14	9.6	Solvothermal synthesis; methanol	-	[1]
	α-Al2O3 disc	20	1.8×10^{-7}	31.5	16.2	Solvothermal synthesis; methanol	Pressure	[2]

	Stainless-steel-net (SSN)	-	2.12×10^{-5}	23.2	15	Solvothermal synthesis; methanol	Pressure	[3]
	Stainless-steel-net (SSN)	-	2.02×10^{-5}	-	4.111	Solvothermal synthesis; methanol	-	[4]
	Hollow ceramic fiber	20	7.29×10^{-7}	10.8	9.2	Solvothermal synthesis; methanol	Heating, pressure	[5]
	Al_2O_3 disc	0.9	1.81×10^{-6}	9.56	-	Solvothermal synthesis; DI water	Long-time, heating	[6]
	Al_2O_3 hollow fiber	7	14.6×10^{-8}	-	30.9	Solvothermal synthesis; methanol	Long-time	[7]
	ZIF-8-PVDF/PVDF dual-layer polymeric hollow fiber	7.8	1.5×10^{-7}	-	69.5	Solvothermal synthesis; methanol	Heating, pressure	[8]
	Zn foam substrates	244	8.1×10^{-6}	7.3	6	Room temperature; methanol	Heating, pressure, long-time	[9]
	Porous nylon	2.5	11.3×10^{-7}	-	4.6	Room temperature; water	-	[14]

	PVDF hollow fiber	0.87	1.19×10^{-5}	27.3	22.4	Gel–vapour deposition	-	[16]
	PSf hollow fiber	3.6	4.7×10^{-9}	17.2	18.3	Microfluidic synthesis; methanol	Pressure	[18]
	α-Al_2O_3 hollow fiber	22	6.1×10^{-7}	12.4	1.8	Microwave assisted; methanol	Heating, pressure	[19]
	TiO_2 disc	42	6.74×10^{-8}	11.2	4.1	Solvothermal synthesis; methanol	Heating	[20]
	PESf hollow fiber	6.2	1.06×10^{-7}	2.95	4.57	Solvothermal synthesis; methanol	Ultrasonic, long-time, reproducibility	[22]
	ZnO hollow fiber	10.5	1.2×10^{-6}	4.29	3.73	Solvothermal synthesis; methanol	Ultrasonic, reproducibility	[25]
	α-Al_2O_3 hollow fiber	4.8	1.19×10^{-6}	9.6	10.2	Solvothermal synthesis; methanol	Ultrasonic, reproducibility	[26]
	α-Al_2O_3 tube	6	1.88×10^{-7}	11.4	10.3	Solvothermal synthesis; methanol	-	[27]
	SSN	10	1.47×10^{-7}	12.4	9	Electrodeposition method; methanol	Long-time	[28]

	AAO disc	2.5	4.71×10^{-6}	9.76	12.53	Room temperature; ethanol-water	-	[29]
	α-Al_2O_3 tube	1	2.3×10^{-7}	14.8	13.7	Room temperature; water	-	[30]
	α-Al_2O_3 tube	8	2.08×10^{-7}	10.4	10.3	Solvothermal synthesis; methanol	-	[31]
	α-Al_2O_3 disc	12	1.0×10^{-7}	16	-	Solvothermal synthesis; methanol	-	[32]
	α-Al_2O_3 hollow fibe	2	4.3×10^{-7}	12.13	11.06	cycling flow; DI water	Long-time	[33]
	PESf disc	35	2.0×10^{-7}	10.5	12.4	Solvothermal synthesis; methanol	Heating	[34]
	Al_2O_3 disc	0.29	1.8×10^{-8}	2.8	3.2	Vapor-solid synthesis	-	[35]
	α-Al_2O_3 disc	0.5	1.9×10^{-8}	9.5	10	stepwise deposition method; methanol	Long-time	[36]
ZIF-8/GO	AAO disc	0.1	5.5×10^{-8}	11.2	11.1	mutual diffusion; methanol	-	[37]

ZIF-67	Al_2O_3-ZnO hollow fiber	7.5	5.24×10^{-7}	5.17	7.74	Solvothermal synthesis; methanol	Heating, ultrasonic, Reproducibility	[23]
	Al_2O_3 disc	7	2.22×10^{-7}	14.92	-	Solvothermal synthesis; methanol	Reproducibility	[10]
	ZnO hollow fiber	13.8	8.98×10^{-8}	15.89	11.75	Solvothermal synthesis; methanol	Heating, long-time, reproducibility	[38]
	α-Al_2O_3 tube	5	8.21×10^{-8}	33.1	28.7	Solvothermal synthesis; water	Long-time	[39]
ZIF-7	PSf hollow fiber	2.4	2.2×10^{-9}	34.6	35.1	Microfluidic synthesis; ethanol	Pressure	[18]
	Al_2O_3 disc	2	4.5×10^{-8}	14.7	20.7	Solvothermal synthesis; DMF	Long-time	[40]
	α-Al_2O_3 disc	1.5	7.4×10^{-7}	6.29	6.73	Microwave synthesis; DMF	-	[41]
ZIF-9	α-Al_2O_3 tube	10	1.85×10^{-7}	7.6	8.9	Solvothermal synthesis; DMF	Long-time, heating	[42]
	α-Al_2O_3 tube	4	1.13×10^{-7}	14.7	8.2	Solvothermal synthesis; DMF	Long-time	[43]
	α-Al_2O_3 disc	50	7.43×10^{-6}	5.16	3.24	Solvothermal synthesis; DMF	-	[44]
UiO-66	Al_2O_3 disc	1.6	4.1×10^{-7}	4.2	6.4	Solvothermal synthesis; DMF-	Long-time	[45]

							HAc		
		Al_2O_3 hollow fiber	2	7.2×10^{-7}	6.4	22.4	Solvothermal synthesis; DMF	-	[46]
		α-Al_2O_3 tube	6	5.4×10^{-7}	11.4	8.2	Solvothermal synthesis; DMF-HAc	-	[47]
UiO-66-NH_2		α-Al_2O_3 hollow fiber	5.7	1.39×10^{-7}	21.7	3.13	Solvothermal synthesis; DMF-HAc	Long-time, heating, ultrasonic, hydrothermal, reproducibility	[48]
		α-Al_2O_3 hollow fiber	3	1.57×10^{-7}	18.9	-	Solvothermal synthesis; DMF-HAc	Heating	[49]
$Zn_2(bim)_4$		ZnO disc	1.8	1.25×10^{-8}	9.2	-	Solvothermal synthesis; toluene	Heating	[50]
Polymer membrane	PLA	-	25	3.35×10^{-10}	10	5	-	-	[51]
	PPSU/PBNPI	-	40-50	3.38×10^{-10}	5.05	-	-	-	[52]
	PIM-1	-	65	1.74×10^{-8}	5.9	8.5	-	-	[53]
	PIM-NH_2	-	67	7.25×10^{-9}	6.8	10.8	-	-	[53]
	PIM-deBOC	-	80	7.12×10^{-9}	6.7	10	-	-	[53]
	PIM-T-BOC	-	71	6.13×10^{-10}	26	36	-	-	[53]
	PIM-1	-	44	1.46×10^{-8}	7.99	10	-	Long-time	[54]
	PIM-1-550	-	34	9.63×10^{-9}	4.54	5	-	Long-time	[54]
	PIM-EA-TB	-	129	2.02×10^{-8}	11.1	14.8	-	-	[55]
	PI	-	40-60	6.45×10^{-9}	15.5	12.9	-	-	[56]

	CTB1-DMN	-	48	9.04×10^{-9}	13.5	17	-	-	[57]
MMMs membrane	ZnO/ZIF-8	-	-	3.29×10^{-8}	16.6	13.1	-	Heating	[58]

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