SUPPORTING INFORMATION

Synthesis of zeolitic imidazolate framework membrane using temperature-switching synthesis strategy for gas separation

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1. Comparison of membranes prepared using the temperature-switching method

Table S1. Single gas permeances (25 ℃ and 1.0 bar) as well as the ideal separation factors on the ZIF-9 membranes using the temperature-switching method with different synthesis times

Membrane	First step		Second step		Total time	Permeance (10^{-6} mol m^{-2} s^{-1} Pa^{-1})				Ideal separation factor		
	T (℃)	t (h)	T (℃)	t (h)	t (h)	H_2	CO_2	N_2	CH_4	H_2/CO_2	H_2/N_2	H_2/CH_4
M1	80	48	130	48	96	7.43	0.50	2.29	1.44	14.71	3.24	5.16
M2	80	24	130	24	48	9.02	0.95	4.49	2.17	9.45	2.01	4.16
M3	80	24	130	48	72	7.51	0.71	3.24	1.62	10.57	2.32	4.64
M4	80	48	130	24	72	8.21	0.84	3.75	1.88	9.76	2.19	4.37

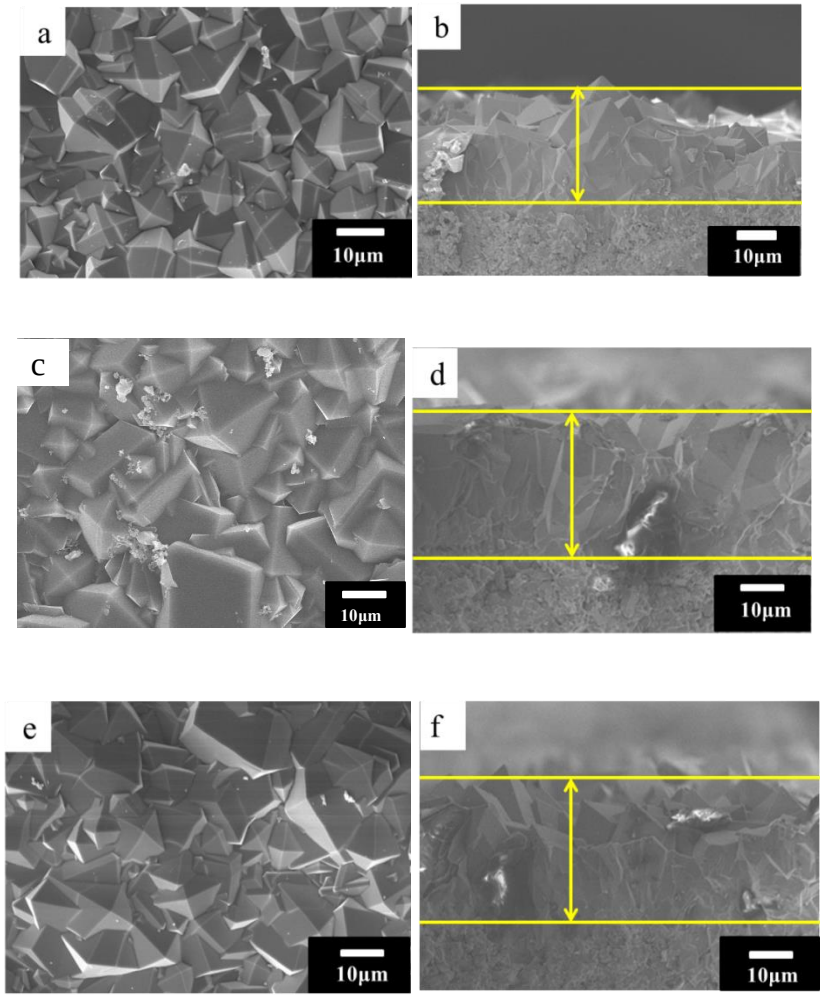


Figure S1 Top view and cross-sectional SEM images of ZIF-9 membranes with different synthesis times: M2 (a, b), M3 (c, d) and M4 (e, f).

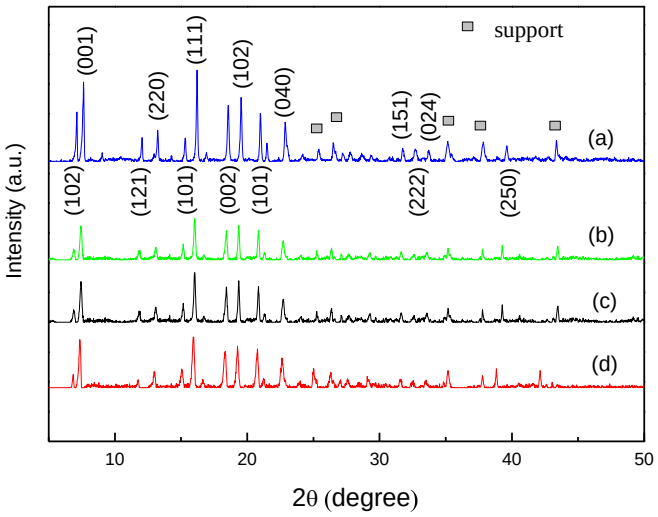


Figure S2 PXRD patterns of the ZIF-9 membranes (a-d: M1-M4)

2. TGA analysis of the ZIF-9 membrane

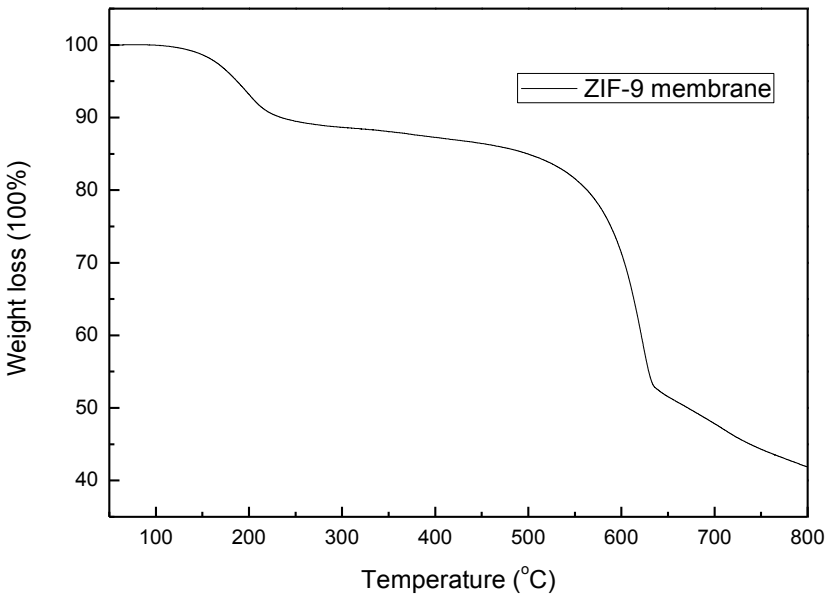


Figure S3 TGA analysis of the ZIF-9 membrane M1

3. Comparison of the characterization of membranes prepared using the conventional synthesis method

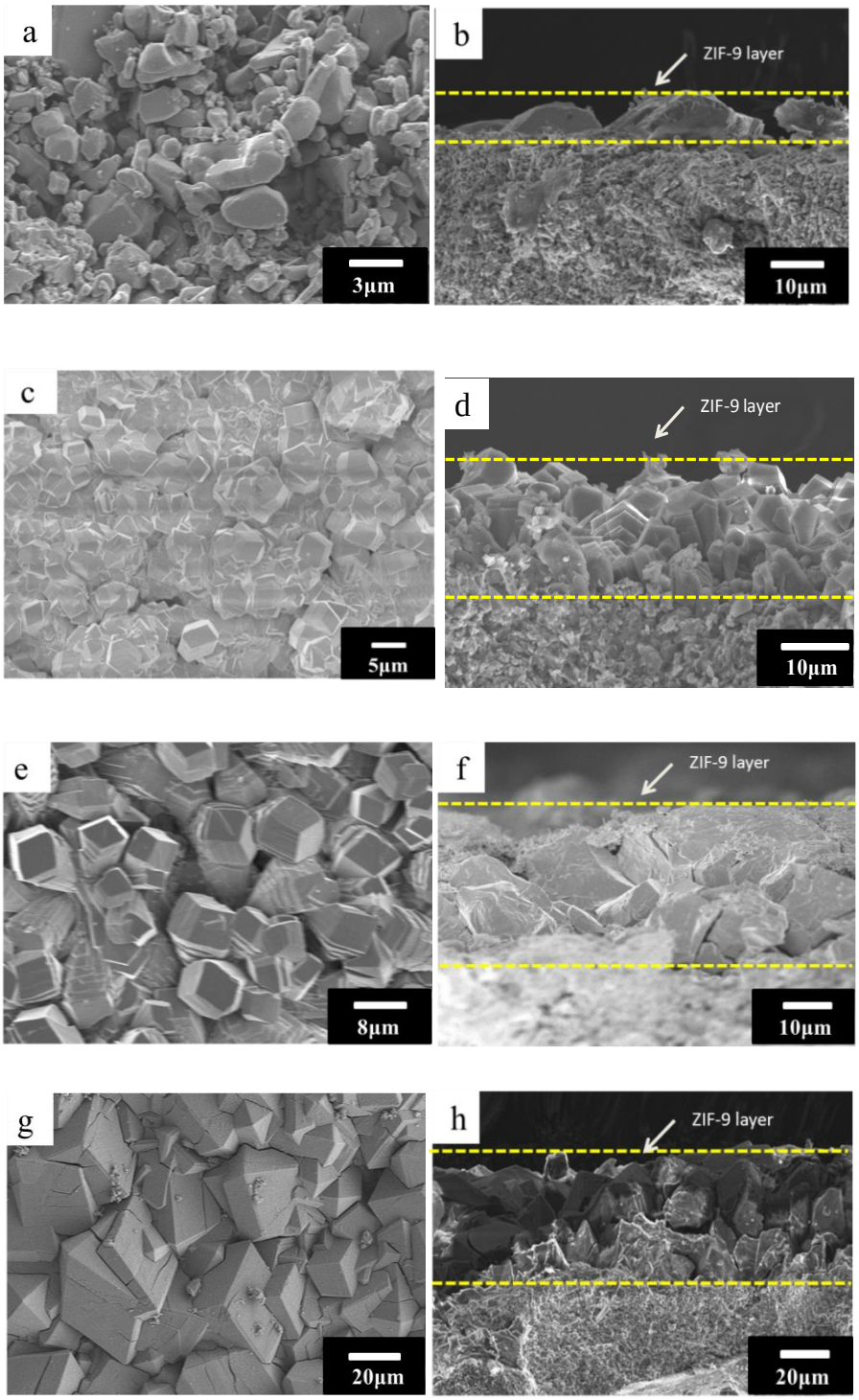


Figure S4 The top view and cross-sectional SEM images of ZIF-9 membranes synthesized in the conventional hydrothermal synthesis method at a constant temperature: M5 (a, b), M6 (c, d), M7 (e, f) and M8 (g, h).

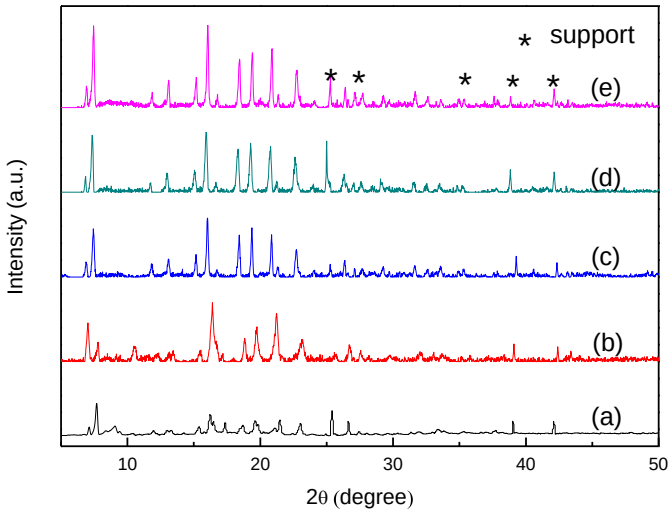
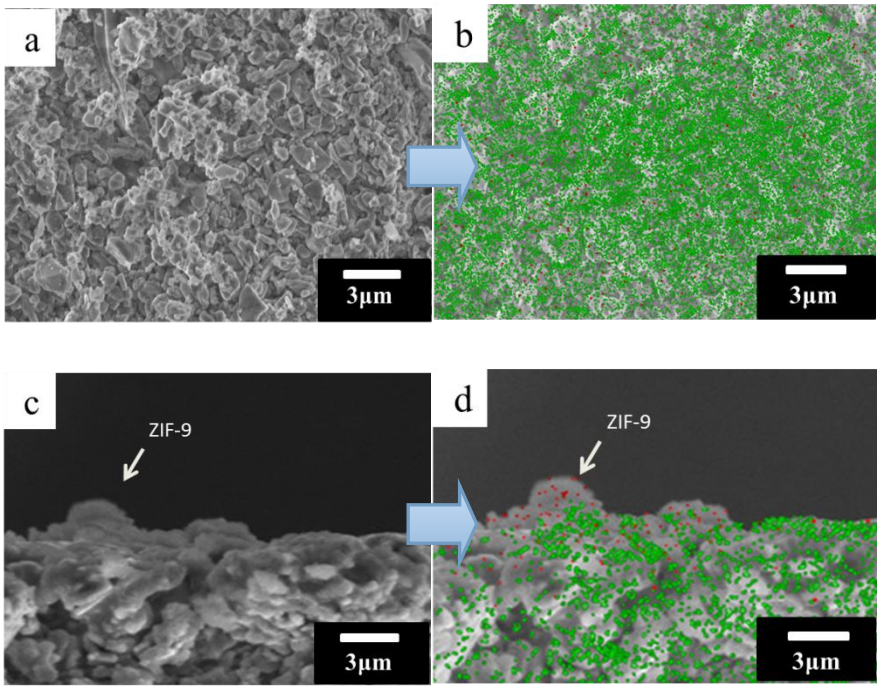


Figure S5 PXRD patterns: membranes M5-M8 (a-d) and membrane M1 (e)

4. Structure and morphological characterization of ZIF-9 seeds



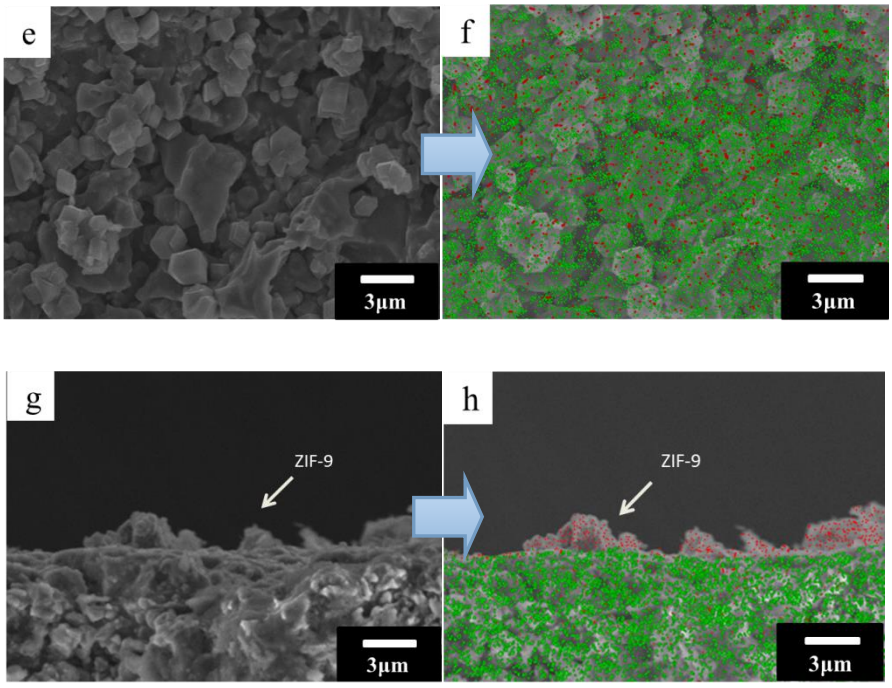


Figure S6 Top view, cross-sectional SEM images and the Energy Dispersive Spectroscopy (EDS) mapping of ZIF-9 seeds prepared for 24 h (a-d) and 48 h(e-h) at 80 ℃ for the first step (red: Co as tracer for ZIF-9; green: Al as the tracer for the support).

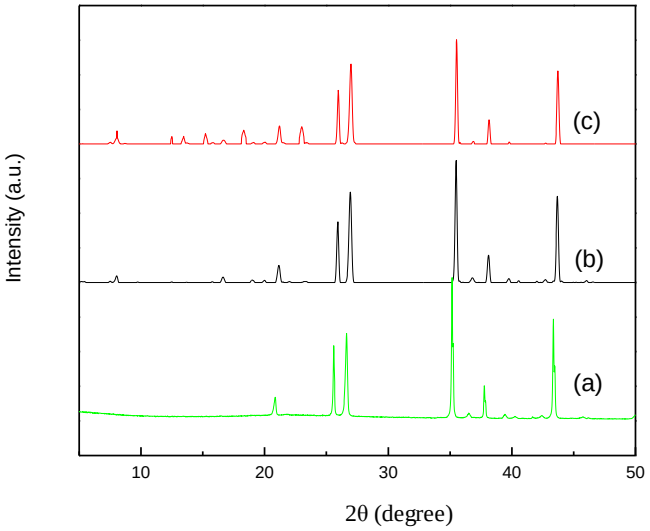


Figure S7 PXRD patterns: α-Al_2O_3 support (a); ZIF-9 seeds growth for 24 h and 48 h at 80 ℃ for the first step (b, c)

5. N_2 adsorption isotherm for α-Al_2O_3 support

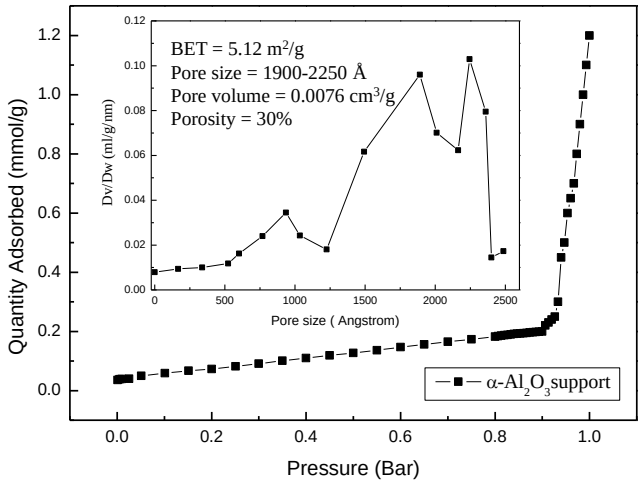


Figure S8 N_2 adsorption isotherm for α-Al_2O_3 support. Inset is the pore size distribution obtained from the adsorption branch.

6. Single gas adsorption and desorption for ZIF-9 membrane

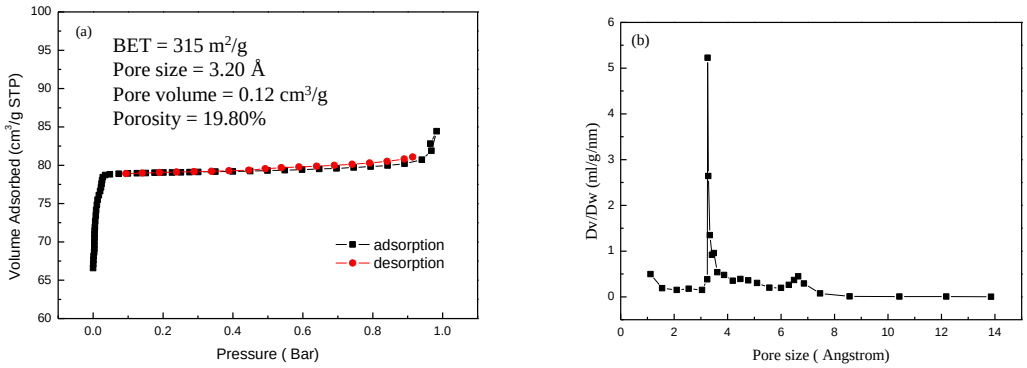


Figure S9 N_2 adsorption and desorption (a) as well as the pore size distribution (b) for ZIF-9 membrane M1

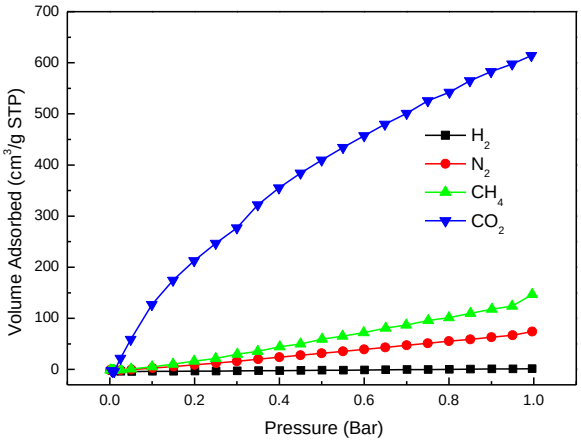


Figure S10 Single gas adsorption at 298 K for ZIF-9 membrane M1

7. Comparison of permeation properties of membranes prepared using the two methods

Table S2 Single gas permeances (25 ℃ and 1.0 bar) as well as the ideal separation factors on ZIF-9 membranes prepared using two methods

Membrane	Reaction conditions		Permeance (10^{-6} mol m^{-2} s^{-1} Pa^{-1})				Ideal separation factor		
	T (℃)	t (h)	H_2	CO_2	N_2	CH_4	H_2/CO_2	H_2/N_2	H_2/CH_4
M1	80,130	96	7.43	0.50	2.29	1.44	14.71	3.24	5.16
M5	80	96	11.35	3.81	6.04	5.54	2.98	1.88	2.05
M6	90	96	10.62	3.07	5.23	3.85	3.46	2.03	2.76
M7	110	96	9.86	1.96	4.01	3.28	5.02	2.46	3.01
M8	130	96	8.71	1.35	2.84	2.04	6.43	3.07	4.26

8. Gas permeance and ideal separation factor as a function of testing temperature.

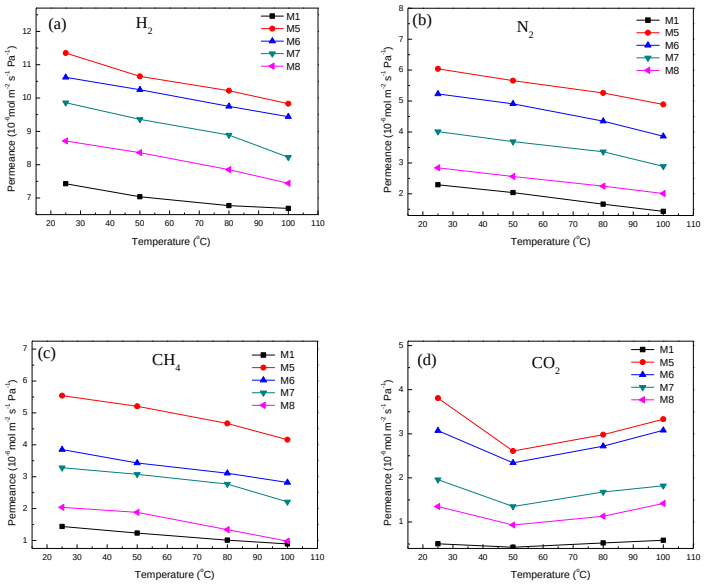


Figure S11 The permeances for single gases as a function of testing temperature in membranes M1 and M5-M8 at 1.0 bar of trans-membrane pressure drop.

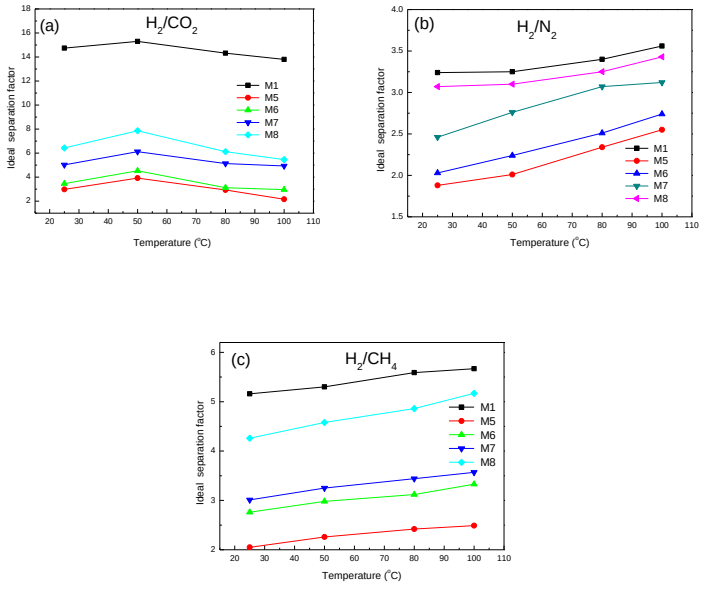


Figure S12 The ideal separation factors for single gases as a function of testing temperature in membranes M1 and M5-M8 at 1.0 bar of trans-membrane pressure drop.

9. Gas permeance and ideal separation factor as a function of pressure drop

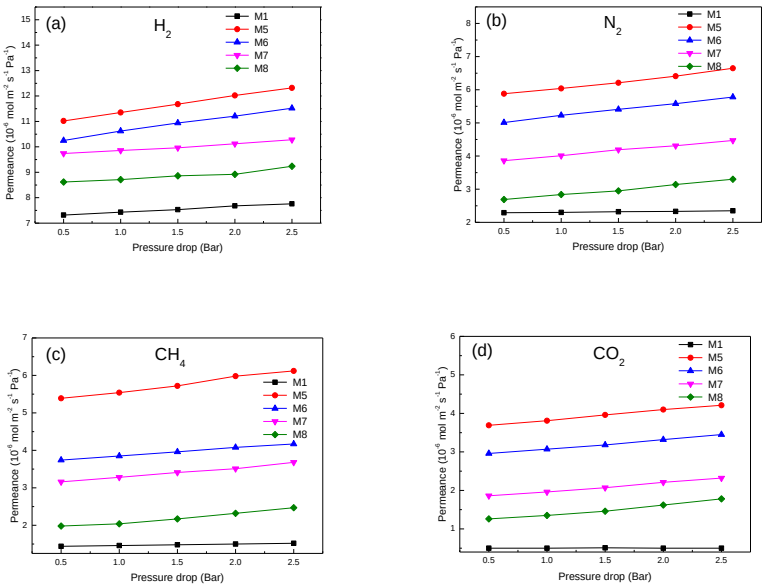


Figure S13 The permeances for single gases as a function of trans-membrane pressure drop in membranes M1 and M5-M8 at room temperature.

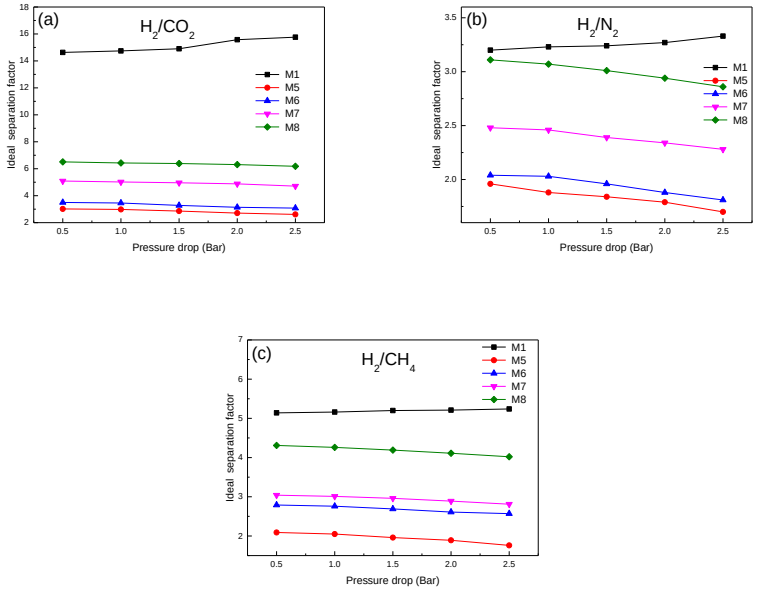


Figure S14 The ideal separation factors for single gases as a function of trans-membrane pressure drop in membranes M1 and M5-M8 at room temperature.

10. Reproducibility of membranes prepared using the temperature-switching synthesis method

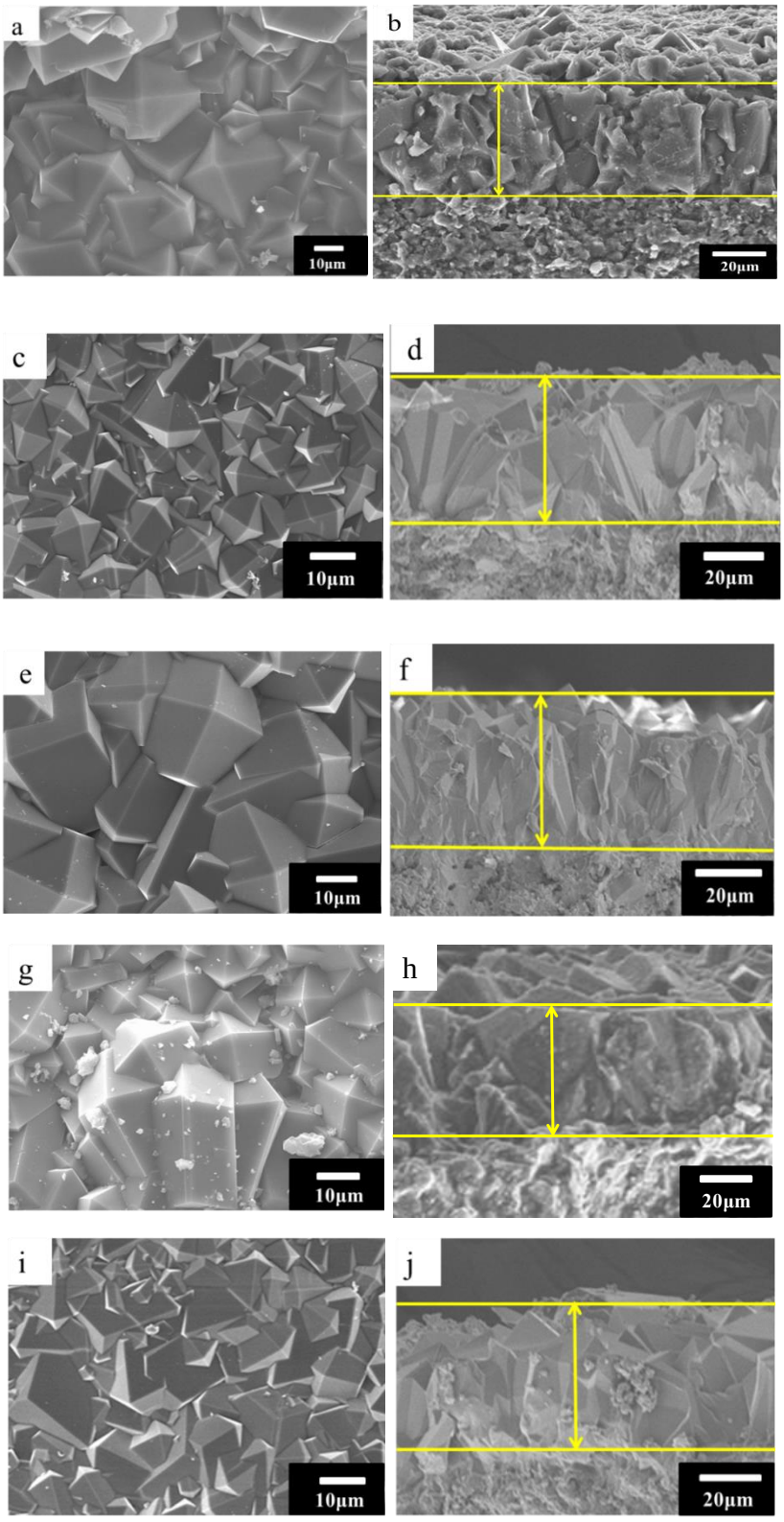


Figure S15 The top view (a, c, e, g and i) and cross-sectional (b, d, f, h and j) SEM images of five ZIF-9 membranes parallelly prepared using the temperature-switching synthesis method

Table S3 Single gas permeances and ideal separation factors at 25 ℃ and 1.0 bar of 5 tested ZIF-9 membranes showing the reproducibility of membrane preparation and testing.

Number	Permeance(10^{-6} mol m^{-2} s^{-1} Pa^{-1})				Ideal separation factor		
	H_2	CO_2	N_2	CH_4	H_2/CO_2	H_2/N_2	H_2/CH_4
1	7.43	0.50	2.29	1.44	14.74	3.24	5.16
2	7.35	0.48	2.22	1.41	15.31	3.31	5.21
3	7.52	0.54	2.26	1.50	13.93	3.33	5.01
4	7.46	0.52	2.25	1.46	14.35	3.32	5.11
5	7.38	0.51	2.20	1.42	14.47	3.35	5.20

11. Characterization of ZIF-9 membrane based on ZrO_2 support

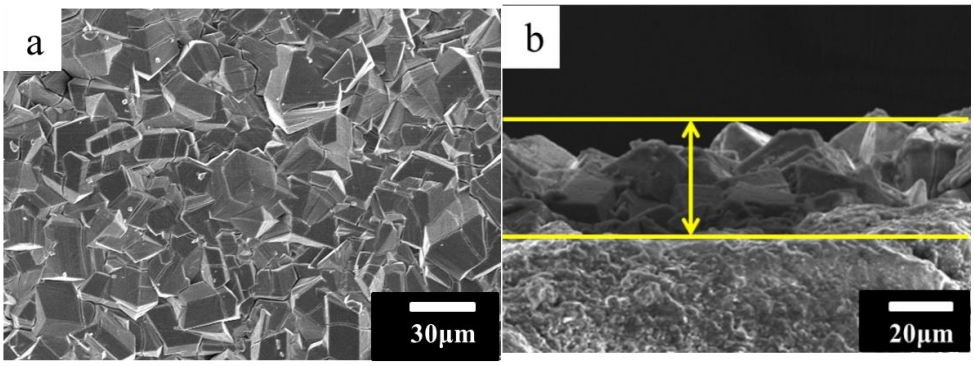


Figure S16 Top view (a) and cross-section (b) SEM images of as-synthesized ZIF-9 membrane based on the ZrO_2 support

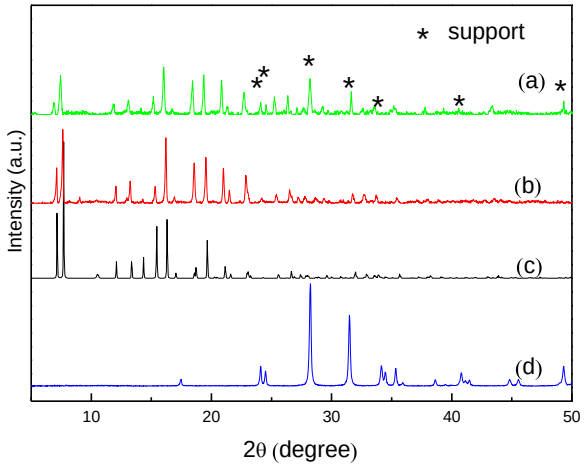


Figure S17 PXRD patterns of as-synthesized ZIF-9 membrane based on the ZrO_2 support: (a) ZIF-9 membrane; (b) ZIF-9 powder; (c) simulation of ZIF-9; (d) the bare ZrO_2 support

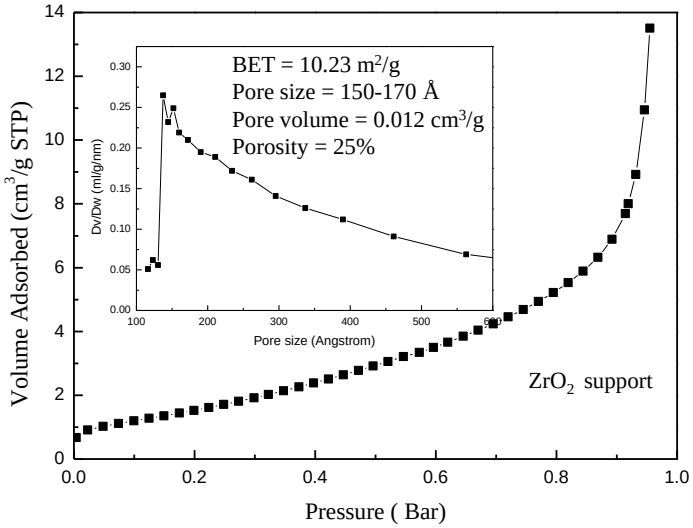


Figure S18 N_2 adsorption isotherm for ZrO_2 support. Inset is the pore size distribution obtained from the adsorption branch.