Electronic Supplementary Information

CO_2-philic WS_2 laminated membranes with nanoconfined ionic liquid

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Additional Figures and Tables

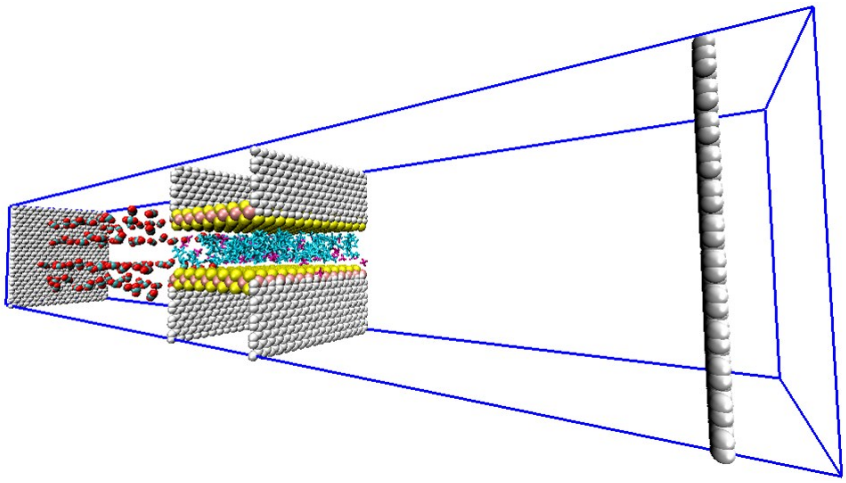

Figure S1 A bichamber system for simulation of gas permeance through the WS_2 SILM. White, yellow, orange, cyan, and red balls denote helium, sulfur, tungsten, carbon, and oxygen, respectively. Green-cyan and magenta stick represent [BMIM] and BF_4 molecules.

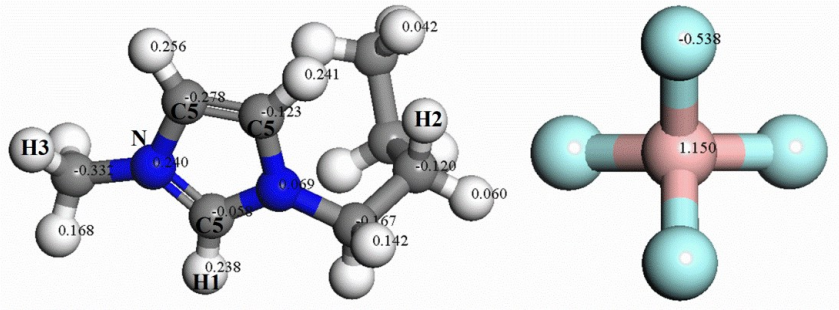

Figure S2 Partial atomic charges on the $[BMIM]^+$ and BF_4^- ions. Atoms are labelled for the bonded-term parameters.

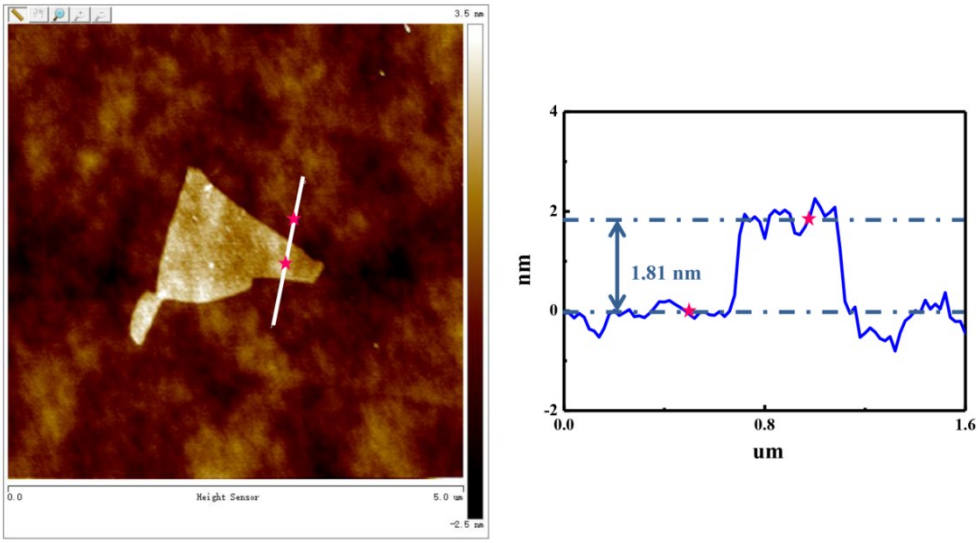

Figure S3 AFM image of WS_2 sheet. Lateral dimension is scaled between two star signs, as shown in right side. The WS_2 sheet is 1.8 nm in thickness. The sharply drop in the height profile is due to silicon substrate.

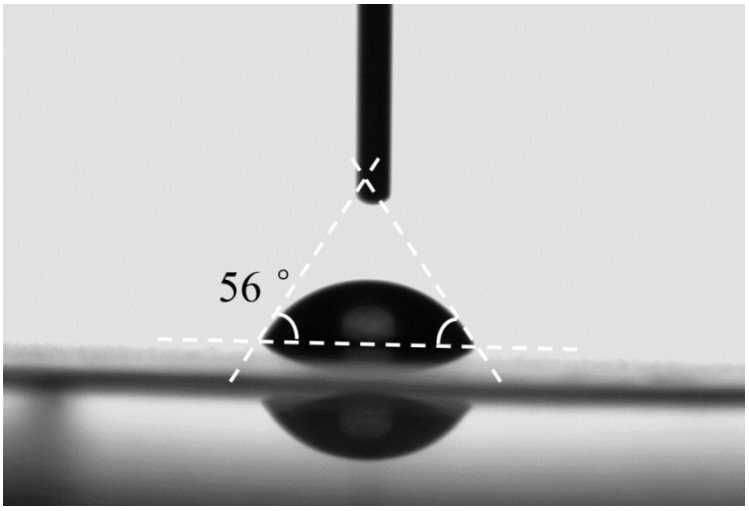

Figure S4 The contact angle image of [BMIM][BF_4] on WS_2 membrane surface.

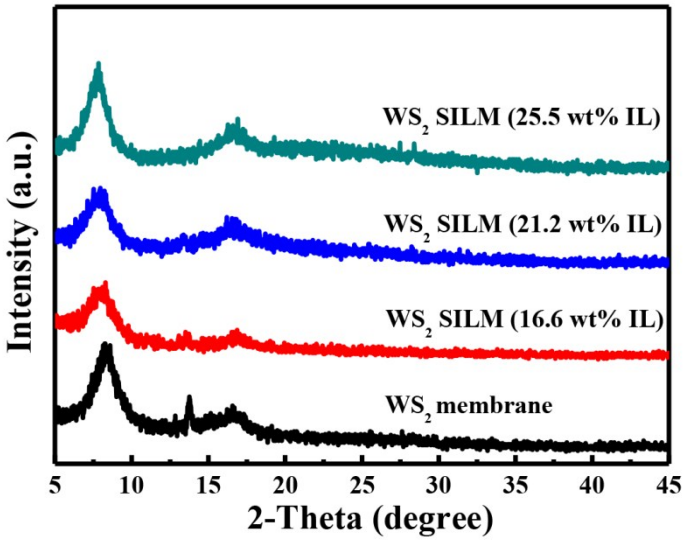

Figure S5 XRD patterns of WS_2 membrane and a series of WS_2 SILMs (16.6 wt% IL, 21.2 wt% IL, 25.5 wt% IL).

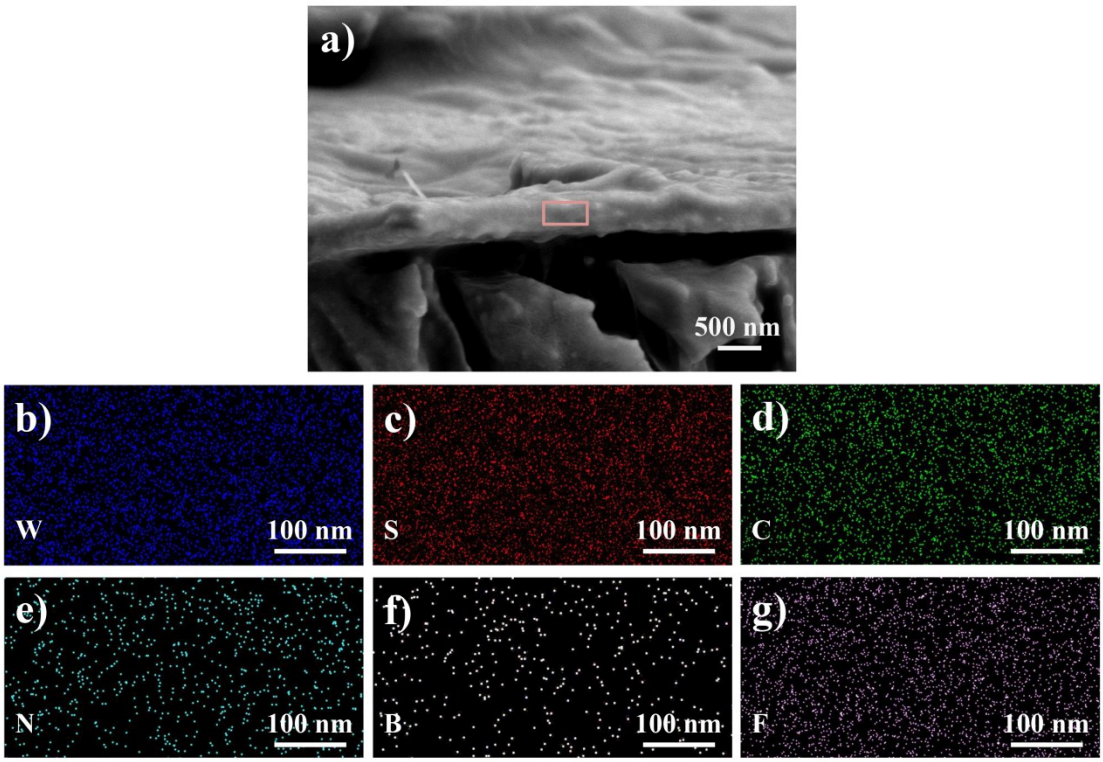

Figure S6 The EDS spectra of WS_2 SILM (25.5 wt% IL): a) cross-section SEM images of WS_2 SILM (25.5 wt% IL), b)-g) EDS mapping images of W, S, C, N, B and F recorded from the marked zone in a), respectively.

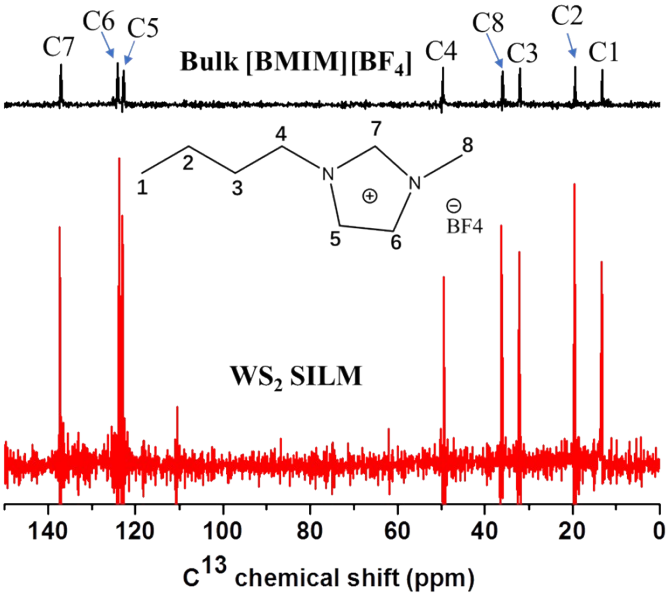

Figure S7 NMR characterization: the ^{13}C spectra of WS_2 SILM (25.5 wt% IL ([BMIM][BF4])) and bulk [BMIM][BF_4].

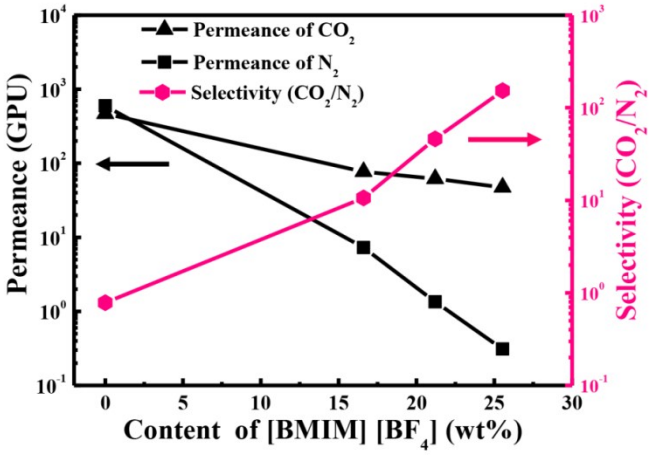

Figure S8 The content of [BMIM][BF_4] influence on the separation performance of WS_2 SILMs.

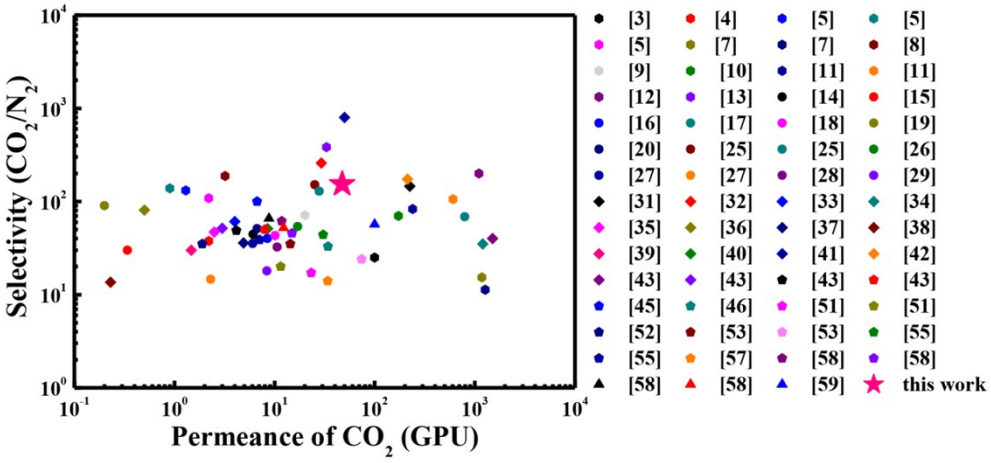

Figure S9 The CO_2/N_2 separation performance of WS_2 SILM (25.5 wt% IL) compared with other materials constructed membranes (see detail in Table S4).

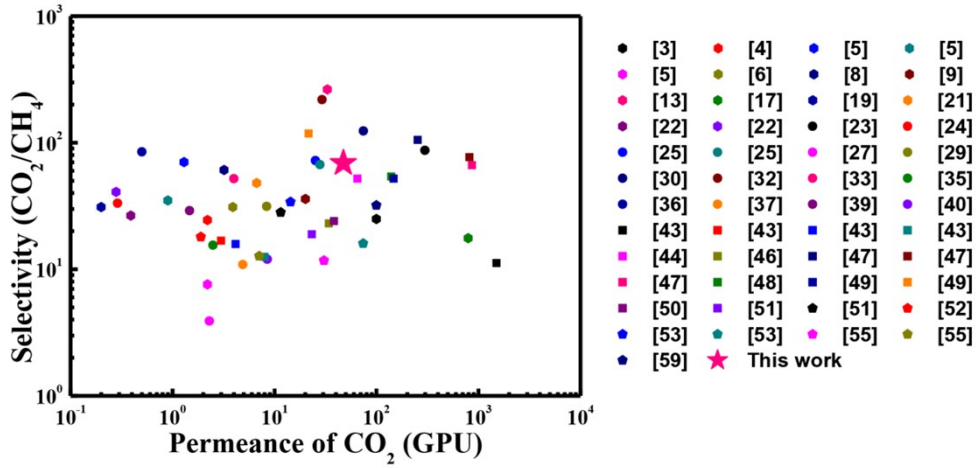

Figure S10 The CO_2/CH_4 separation performance of WS_2 SILM (25.5 wt% IL) compared with other materials (Table S4).

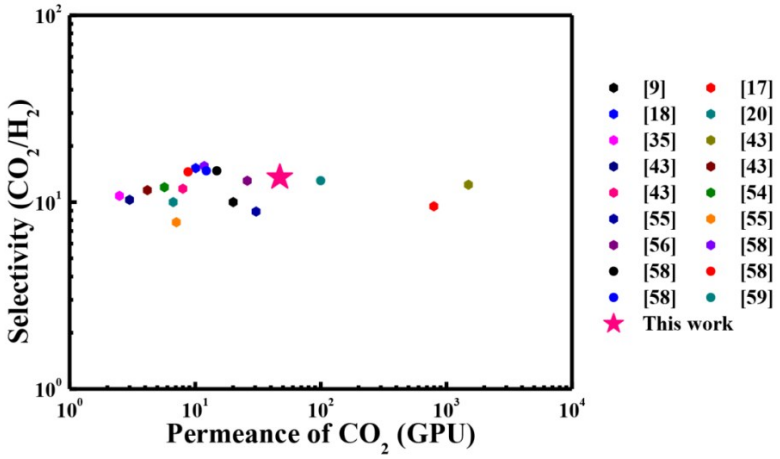

Figure S11 The CO_2/H_2 separation performance of WS_2 SILM (25.5 wt% IL) compared with other materials (Table S4).

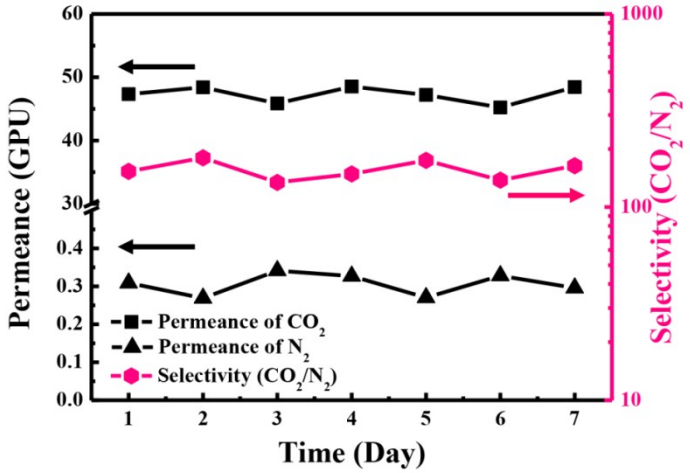

Figure S12 The permeance of CO_2 and N_2 through WS_2 SILM (25.5 wt% IL) for seven days.

Table S1 Lennard-Jones parameters of helium sheet, WS_2 and IL employed in this work[1]

Element	He	W	S	N	C5	C3
ε (kcal/mol)	0.0203	0.067	0.274	0.170	0.086	0.011
σ (Å)	2.600	3.069	4.035	3.250	3.400	3.400
Element	C2	H1	H2	H3	B	F
ε (kcal/mol)	0.011	0.015	0.0157	0.0157	0.095	0.061
σ (Å)	3.400	2.450	2.650	2.500	3.581	3.118

Table S2 Partial atomic charges and Lennard-Jones parameters for gas molecules

	CO_2		N_2	
	C	**O**	**N**	**Center of Mass**
ε (kcal/mol)	0.0559	0.160	0.0728	0
σ (Å)	2.757	2.565	3.318	0
q / |e|	0.6512	-0.3256	-0.4084	0.8096

Table S3 WS_2 SILMs' obvious FTIR Shift compared with bulk [BMIM][BF_4][2].

[BMIM][BF_4]	**WS_2 SILM (25.5 wt%)**	**Shift of WS_2 SILM (25.5 wt%)**	**WS_2 SILM (21.2 wt%)**	**Shift of WS_2 SILM (21.2 wt%)**	**WS_2 SILM (16.6 wt%)**	**Shift of WS_2 SILM (16.6 wt%)**	**Assignments**
3160.81	3151.16	-9.64	3160.81	0.00	3160.81	0.00	*v* C4-H, *v* C5-H (out-of-phase)
3122.24	3126.09	3.86	3118.38	-3.86	3122.24	0.00	*v* C2-H
2964.10	2958.31	-5.79	2962.17	-1.93	2964.10	0.00	v_{as} CH_2
2939.03	-	-	2937.10	-	2937.10	-1.93	v_{as} CH_2
2877.32	2875.39	-1.93	2873.46	-3.86	2875.39	-1.93	v_s CH_2 (Bu)
1573.65	1560.14	-13.51	1560.15	-13.50	1569.80	-3.86	*v* C=C
1465.66	1457.94	-7.71	-	-	1465.66	0.00	δ_s CH_3
1430.94	1419.37	-11.57	1430.15	-0.79	1430.94	0.00	δ_s CH_3 (Me)
1384.66	-	-	-	-	1386.59	1.93	*w* CH_2
1371.16	1375.02	3.86	1375.02	3.86	-	-	v_{as} C2N1C5, *w* CH_2
1338.38	1340.31	1.93	1340.31	1.93	1340.31	0.00	v N-Bu, N-Me, breathing, *w* CH_2
1284.38	1272.81	-11.57	1284.38	0.00	1286.31	1.93	*v* BF_4, *t* CH_2
1168.67	1164.81	-3.86	1168.67	0.00	1168.67	0.00	*v* N-Bu, N-Me, *r* CH
1114.67	1112.74	-1.93	1114.67	0.00	1116.60	1.93	*r* CH_3, *r* CH_3 (Bu)
1031.75	971.96	-59.78	1016.32	-15.43	1035.60	3.86	*v* C-C, *v* BF
1014.39	966.18	-48.21	1006.68	-7.71	1016.32	1.93	*v* BF_4
846.61	792.61	-54.00	842.75	-3.86	844.68	-1.93	*γ* C4-H, C5-H
752.11	721.26	-30.86	736.69	-15.43	748.26	-3.86	*r* CH_2
696.19	690.40	-5.79	-	-	700.04	3.86	*v* N-Bu, *v* N-Me

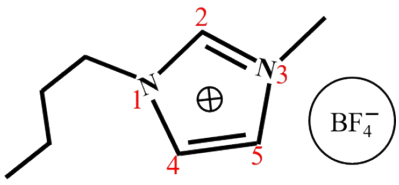

Table S4 WS_2 SILM' Shifts of ^{13}C Chemical Shift compared with bulk [BMIM][BF_4]

	[BMIM][BF_4]	**WS_2 SILM**	**Shift**
C1	-13.167[a]	-13.287	-0.120
C2	-19.421	-19.507	-0.086
C3	-32.038	-32.199	-0.161
C4	-49.648	-49.318	0.330
C5	-124.034	-123.74	0.294
C6	-122.733	-123.013	-0.28
C7	-137.056	-137.316	-0.26
C8	-36.121	-36.214	-0.103

[a] the unit is ppm.

Table S5 Separation performance of WS_2 SILM (25.5 wt% IL) compared with other materials.

Materials	Permeance (GPU)	ΔP (bar)	α CO_2/N_2	α CO_2/CH_4	α CO_2/H_2	Ref
BTPP-HCl-240	100	1	25	25	-	3
PI + acid-treated MWCNTs	1.7-2.2	1	37.7	24.5	-	4
PEG crosslinked with [TETA][Tfa] 80% free PEGDME	1.3	2	131.6	70.3	-	5
PEG cross-linked with A-amine 40% free PEGDME	0.9	2	138.3	35	-	5
PEG cross-linked with A-amine 80% free PEGDME	2.2	2	108.5	7.6	-	5
6FDA-DAM-ZIF-11	3.9	4		31	-	6
$(PDAC/PSS)_{25}(GO/GO)_{40.5}$	1175	-	15.33	-	-	7
$(PDAC/PSS)_{25}(GO/GO)_{20.5}$	1269	-	11.3	-	-	7
Pebax/Ag/50%[BMIM][BF_4]	3.2	10	187.5	61	-	8
IL-Pebax 165/ZIF-8 Mixed Matrix Membranes	20.1	2	71	36	10	9
DNMDAmTMC/PS	173	1	70	-	-	10
PVAm-EDA/PS	239	1	83	-	-	11
PVAm-EDA/PS	607	1	106	-	-	11
PVAm/zeolite-γ/PES	1100	0.1	200	-	-	12
AF-MWNTs/PVA-POS/PSF	33	15	384	264	-	13
AbS-PEG	6.1	1	44.36	-	-	14
Aminosilane Cross-linked PEG	0.34	0.69	30	-	-	15
PEG–silica	8.45	2.02	40	-	-	16
Polyactive	795	1	68.6	17.6	9.5	17
Pebax-PEGDME50	6.1-10.1	0.3	43	-	15.2	18
PU100	0.2	10	90	31	-	19
PTT-b-PEO	2.9-6.7	0.3	51	-	10	20
Pebax 1657/PDA	5.4-6.7	2.02	-	48	-	21
PC	0.39	3	-	26.6	-	22
PC/PEG-300 (95:5)	0.28	3	-	40.9	-	22
[TESPMIM][BF_4] modified membrane ILM-4	298.5	2	-	87		23
ETS-10/Matrimid	0.01-0.29	8	-	33.3	-	24
[DMAPAH][EOAc]/PES	25.2	0.025	151.4	72.1	-	25
[DMAPAH][TFA]/PES	27.9	0.02	128.9	67	-	25
[Emim][$B(CN)_4$]/PES	17	0.35	53.7	-	-	26
[Emim][CF_3SO_3]/PES	6.05	0.2	35.4	-	-	27
[P66614][Cl]/PES	2.3	0.2	14.6	3.9	-	27

Table S5 continued

[Emim][Ac]/PVDF	10.6	0.45	32.4	-	-	28
$[C_2mim][Tf_2N]_{0.75}[Ac]_{0.25}$/PVDF	8.4	1	18	31.4	-	29
$[C_3NH_2mim]$ $[CF_3SO_3]$/PTFE	74.3	1.01		123.8	-	30
[Emim][Gly]/PTFE	224.1	0.1	145.5	-	-	31
$[N_{2224}]_2$[maleate]/PES	24.7-29.3	0.1	258.2	218.5	-	32
Matrimids/NHs-20	2.8-4.0	2	61	52	-	33
UTFC-MMMs	1200	3.5	35	-	-	34
Pebax®/PEG50	1.5-2.5	0.6	47	15.5	10.8	35
Matrimid-CNTs/GO-5/5	0.4-0.5	2	81	84.6	-	36
PEO-ran-PPO-T6T6T	4.9	4	36	10.9	-	37
PEG cross-linked with ILs	0.2-0.23	0.81	13.6	-	-	38
PEG-MOF	1.47	1	30	29	-	39
PETA/PEGMEA	3.4-8.5	2	51	12	-	40
APS/MS/Al_2O_3(S)	50	1.5	800	-	-	41
PVAm/PVA	212	2	174	-	-	42
PEO-PBT/PEG-DBE (PAN-PDMS)	1500	0.3	40	11.2	12.4	43
PEO-PBT	3	0.3	51.5	16.8	10.3	43
PEO-PBT+PEG	4.16	0.3	48.7	15.8	11.6	43
PEO-PBT+PEG-BE	8	0.3	50.1	12.5	11.8	43
PETEDA-PVA/PES	65.1	2	-	52	-	44
AAIL-based DN gel membranes	6.7	0.1	100	-	-	45
9,10-Diisopropyltriptycene-Based KAUST-PI Membranes	23.9-34.1	2	33	23	-	46
M-3-20-750	253.3	1	-	105	-	47
M-3-100-750	817.8	1	-	76.8	-	47
M-2-20-750	864.9	1	-	66.4	-	47
6FDA/BPDA-DAM	81-139.9	2	-	54	-	48
6FDA/mPDA/DABA -CMSMs	147.5	6.9	-	52	-	49
6FDA/mPDA/DABA-CMSMs	21.6	6.9	-	118	-	49
4 wt%-ns-CuBDC@PIM-1	32.8-38.3	3	-	24	-	50
6FDA-CADA1-450	23.2	2.02	17.2	18.9	-	51
6FDA-CADA1-425	11.5	2.02	20	28.1	-	51
PIL-IL 75	1.9	1	35	18	-	52
AO-PIM-1	11.5-14.4	2	35	34	-	53
PIM-1	59.2-74	2	24	16	-	53
poly(ethylene glycol) acrylate	5.7	4.5	-	-	12	54
LPPEOMASQ82	30.6	3	43.9	11.7	8.9	55
LPPEOMASQ64	7.1	3	38.8	12.7	7.8	55

Table S5 continued						
poly(ethylene glycol)dimethyl incorporated to PEG	13-26	3.55	-	-	13	56
(bicycloheptenyl)ethylterminated polydimethylsiloxane (PDMSPNB) membranes	27.2-34	0.4	14	-	-	57
SIPN-3-7-10	9.5-11.8	3.55	61.4	-	15.6	58
SIPN-7-3-10	11.9-14.9	3.55	45.7	-	14.7	58
SIPN-0-10-10	7.1-8.8	3.55	65.9	-	14.5	58
SIPN-5-5-10	9.9-12.3	3.55	52.1	-	14.7	58
GO membranes under wet-conditions	100	0.3	57	32	13	59
WS_2 SILM (25.5 wt% IL)	**47.3**	**0.4**	**153.21**	**68.81**	**13.56**	**This work**

Table S6. Gas separation performance of WS_2 SILM (25.5 wt% IL) compared with other [BMIM][BF_4]-based membranes.

[BMIM][BF_4]-based membranes	**CO_2 permeance (GPU)**	**Selectivity (CO_2/N_2)**	**Selectivity (CO_2/CH_4)**	**Ref.**
Neat [BMIM][BF_4]	17	5	4.8	60
[BMIM][BF_4]/Cu	25	11	11	60
[BMIM][BF_4] / (Cu NPs/KIT-6) 1/0.1	51	16	23	61
[BMIM][BF_4]-PES	11.5	50	-	62
0.1Water/[BMIM][BF_4]-PES	13.8	60	-	62
0.15Water/[BMIM][BF_4]-PES	13.3	47	-	62
[BMIM][BF_4]/AgO	14.1	28.2	-	63
[BMIM][BF_4]/CuO	52.4	21	-	64
[BMIM][BF_4]/$LiBF_4$	13.36	8.4	8.25	65
[BMIM][BF_4]/cyanuric chloride	19.2	11	10.7	66
WS_2 SILM (25.5 wt%)	**47.3**	**153.21**	**68.81**	**This work**

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