Supplementary Materials

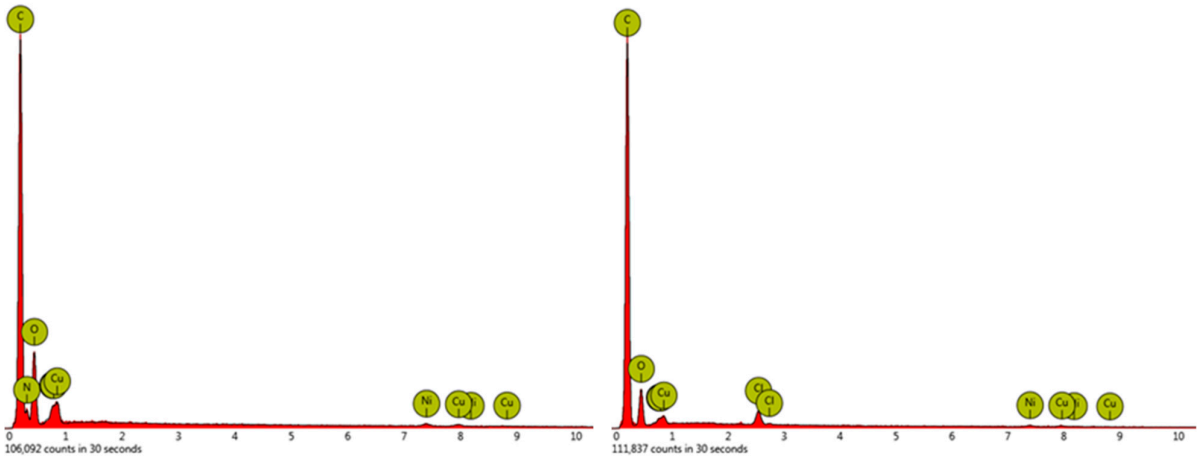
1. Membrane preparation

Table SI-1. List of MMMs prepared

MMM names	Weight Ratio PIM-1:MOF	Filler g	Polymers g	MOF loading (wt%)	MOF loading (vol%) [a)]
PEBAX/CuNi	10:1	0,030	0,300	9,1	9,4
	10:2	0,060	0,300	16,7	16,0
	10:3	0,090	0,300	23,1	20,9
PEEK-WC/CuNi	10:1	0,030	0,300	9,1	18.8
	10:2	0,060	0,300	16,7	31.6
	10:3	0,090	0,300	23,1	41.0

[a)] Volume fraction calculated from bulk density equal to 1,25 g cm^{-3} for PEEK-WC and 1,14 g cm^{-3} for Pebax®1657 and 0,54 g cm^{-3} for CuNi-MOF.

2. Membrane characterization



(a) (b)

Fig. S-1 EDX of Pebax®1657/CuNi-MOF (A) and PEEK-WC/CuNi-MOF (B) at an accelerating voltage of 15Kv.

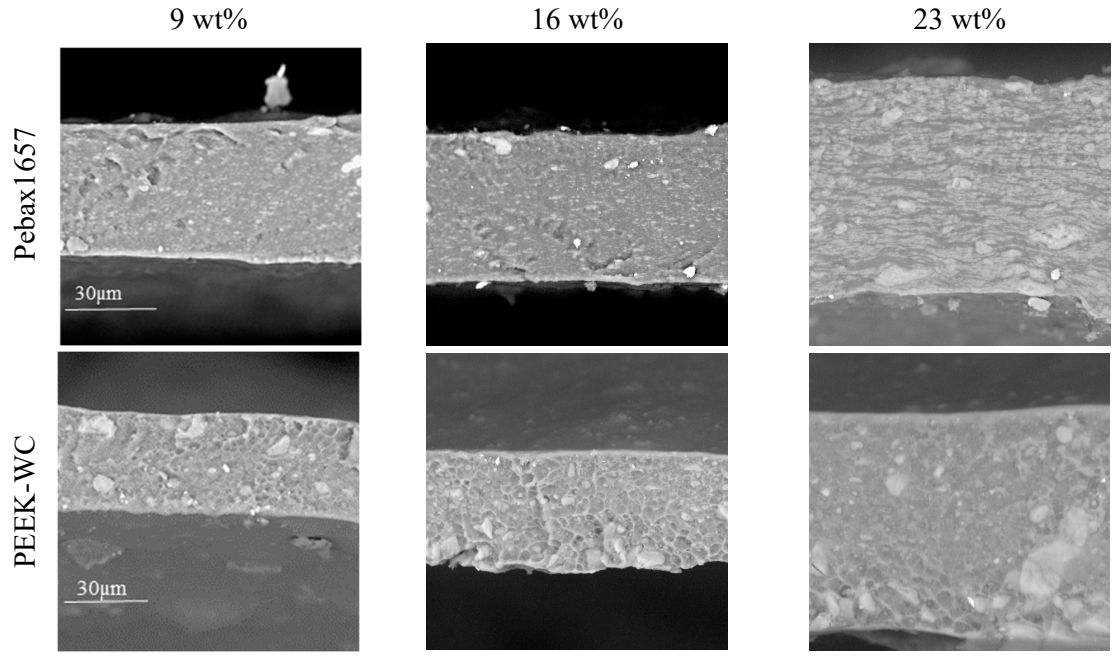


Fig. S-2. SEM images of cross section for MMMs of Pebax®1657/CuNi-MOF and PEEK-WC/CuNi-MOF with different MOF loadings at a magnification of 2,500 x and an accelerating voltage of 15 kV. The indicated scale bar is identical for all membranes.

3. Gas permeation

3.1 Pure gas permeation measurements

An overview of the pure gas permeability, solubility and diffusion coefficients, and respective selectivity for Pebax1657 CuNi MMMs is given in

Table S-2 Pure gas permeability, solubility and diffusion coefficients, and respective selectivity for neat Pebax1657 and Pebax1657/CuNi MMMs.

CuNi	Permeability [Barrer]						α (Px/Py)				
[wt%]	N_2	O_2	CO_2	CH_4	H_2	He	CO_2/N_2	CO_2/CH_4	O_2/N_2	H_2/CH_4	H_2/N_2
0	1.3	3.3	76.2	4.1	7.8	5.2	58.6	18.6	2.5	1.9	6.0
9	1.2	3.2	71.9	3.8	7.7	5.0	61.3	18.8	2.7	2.0	6.5
17	1.2	2.9	56.6	3.3	8.0	5.4	49.2	17.3	2.5	2.4	6.9
23	1.4	3.8	70.9	3.8	11.5	8.0	51.9	18.5	2.7	3.0	8.4
CuNi	Dx [10-12 m2s-1]						α (Dx/Dy)				
[wt%]	N_2	O_2	CO_2	CH_4	H_2	He	CO_2/N_2	CO_2/CH_4	O_2/N_2	H_2/CH_4	H_2/N_2
0	74	85	51	40	484	1005	0.7	1.3	1.1	12.1	6.5
9	36	52	34	23	343	511	0.9	1.5	1.4	14.9	9.5
17	14	25	16	10	206	297	1.1	1.6	1.8	20.8	14.4
23	8	20	13	7	197	273	1.5	2.0	2.3	30.1	23.4
CuNi	Sx [cm3(STP) cm-3 bar-1]						α (Sx/Sy)				
[wt%]	N_2	O_2	CO_2	CH_4	H_2	He	CO_2/N_2	CO_2/CH_4	O_2/N_2	H_2/CH_4	H_2/N_2
0	0.0	0.0	1.1	0.1	0.0	0.00	85.0	14.6	2.2	0.2	0.9
9	0.0	0.0	1.6	0.1	0.0	0.01	66.1	12.9	1.9	0.1	0.7
17	0.1	0.1	2.7	0.2	0.0	0.01	44.0	10.7	1.4	0.1	0.5
23	0.1	0.1	4.1	0.4	0.0	0.02	34.0	9.4	1.2	0.1	0.4

An overview of the pure gas permeability, solubility and diffusion coefficients, and respective selectivity for PEEK-WC CuNi MMMs is given in Table S-3.

Table S-3 Pure gas permeability, solubility and diffusion coefficients, and respective selectivity for neat PEEK-WC and PEEK-WC/CuNi MMMs.

CuNi	Permeability [Barrer]					α (Px/Py)				
[wt%]	N_2	O_2	CO_2	CH_4	H_2	CO_2/N_2	CO_2/CH_4	O_2/N_2	H_2/CH_4	H_2/N_2
0	0,24	1,20	6,06	0,20	13,43	25,45	30,59	5,04	67,80	56,40
9	0,43	2,34	13,38	0,39	-	31,35	34,70	5,48	-	-
17	0,90	4,13	33,70	0,71	69,00	37,44	47,46	4,59	97,18	76,67
23	1,32	6,51	47,84	1,00	68,01	36,17	47,84	4,92	68,01	51,42
CuNi	Dx [10-12 m2s-1]					α (Dx/Dy)				
[wt%]	N_2	O_2	CO_2	CH_4	H_2	CO_2/N_2	CO_2/CH_4	O_2/N_2	H_2/CH_4	H_2/N_2
0	0,50	2,09	0,60	0,14	134	1,19	4,38	4,13	982,27	266,17
9	3,32	9,88	2,31	0,61	-	0,70	3,79	2,98	-	-
17	4,22	12,41	3,18	0,70	1200	0,75	4,55	2,94	1714,29	284,36
23	4,30	16,68	3,55	0,95	731	0,83	3,74	3,88	770,17	170,34
CuNi	Sx [cm3(STP) cm-3 bar-1]					α (Sx/Sy)				
[wt%]	N_2	O_2	CO_2	CH_4	H_2	CO_2/N_2	CO_2/CH_4	O_2/N_2	H_2/CH_4	H_2/N_2
0	0,35	0,43	7,59	1,09	0,07	21,46	6,99	1,22	0,07	0,21
9	0,10	0,18	4,35	0,47	-	45,09	9,16	1,84	-	-
17	0,16	0,25	7,94	0,76	0,04	49,62	10,43	1,56	0,06	0,27
23	0,23	0,29	10,11	0,79	0,07	43,78	12,81	1,27	0,09	0,30

3.2 Mixed gas permeation measurements

Table S-4 Mixed gas permeabilities and selectivities of PEEK-WC/CuNi 23 wt% membrane using binary mixture CO_2/CH_4 (35/65) at pressure of 1-6 bar

	Permeability (Barrer)		Selectivity
Total Pressure	**CH_4**	**CO_2**	**CO_2/CH_4**
1	0.83	45.4	55.1
2	0.84	42.3	50.1
3	0.87	40.1	46.3
4	0.82	39.1	47.5
5	0.77	37.5	48.8
6	0.72	36.0	49.7
5	0.74	36.8	49.9
4.5	0.77	38.6	49.9
3.5	0.83	40.8	49.0
2.5	0.91	43.7	48.1
1.5	1.02	46.9	45.8
1	1.09	48.4	44.2

Table S-5 Mixed gas permeabilities and selectivities of PEEK-WC/CuNi 23 wt% membrane using binary mixture CO_2/N_2 (15/85) at pressure of 1-6 bar

	Permeability (Barrer)		Selectivity
Total Pressure	**N_2**	**CO_2**	**CO_2/N_2**
1	1.08	51.2	47.4
2	1.12	49.7	44.2
3	1.08	47.6	44.0
4	1.00	45.7	45.7
5	0.97	44.0	45.6
6	0.93	42.0	45.1
5	0.94	42.9	45.6
4.5	0.99	44.5	45.1
3.5	1.05	46.2	44.0
2.5	1.06	48.6	45.9
1.5	1.19	51.2	43.1
1	1.14	52.1	45.5

Table S-6 Mixed gas permeabilities and selectivities of Pebax®1657/CuNi 23 wt% membrane using binary mixture CO_2/CH_4 (35/65) at pressure of 1-6 bar

	Permeability (Barrer)		Selectivity
Total Pressure	**CH_4**	**CO_2**	**CO_2/CH_4**
1	2.99	63.3	21.2
2	3.27	63.2	19.3
3	3.31	63.5	19.2
4	3.36	63.6	19.0
5	3.41	64.0	18.8
6	3.59	70.8	19.7
5	3.62	71.0	19.6
4.5	3.62	67.5	18.6
3.5	3.60	67.9	18.9
2.5	3.56	68.2	19.2
1.5	3.54	68.3	19.3
1	3.51	68.8	19.6

Table S-7 Mixed gas permeabilities and selectivities of Pebax/CuNi 23 wt% membrane using binary mixture CO_2/N_2 (15/85) at pressure of 1-6 bar

	Permeability (Barrer)		Selectivity
Total Pressure	**N_2**	**CO_2**	**CO_2/N_2**
1	1.18	67.9	57.4
2	1.22	68.5	56.0
3	1.22	67.9	55.6
4	1.22	69.2	56.8
5	1.23	68.3	55.4
6	1.25	68.5	54.8
5	1.25	68.5	54.6
4.5	1.28	68.7	53.6
3.5	1.27	69.9	55.0
2.5	1.31	69.4	53.1
1.5	1.31	70.2	53.5
1	1.33	72.0	54.3