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Supporting Information

Highly H_2O Permeable Ionic Liquid Encapsulated Metal-Organic Framework Membranes for Energy-efficient Air-Dehumidification

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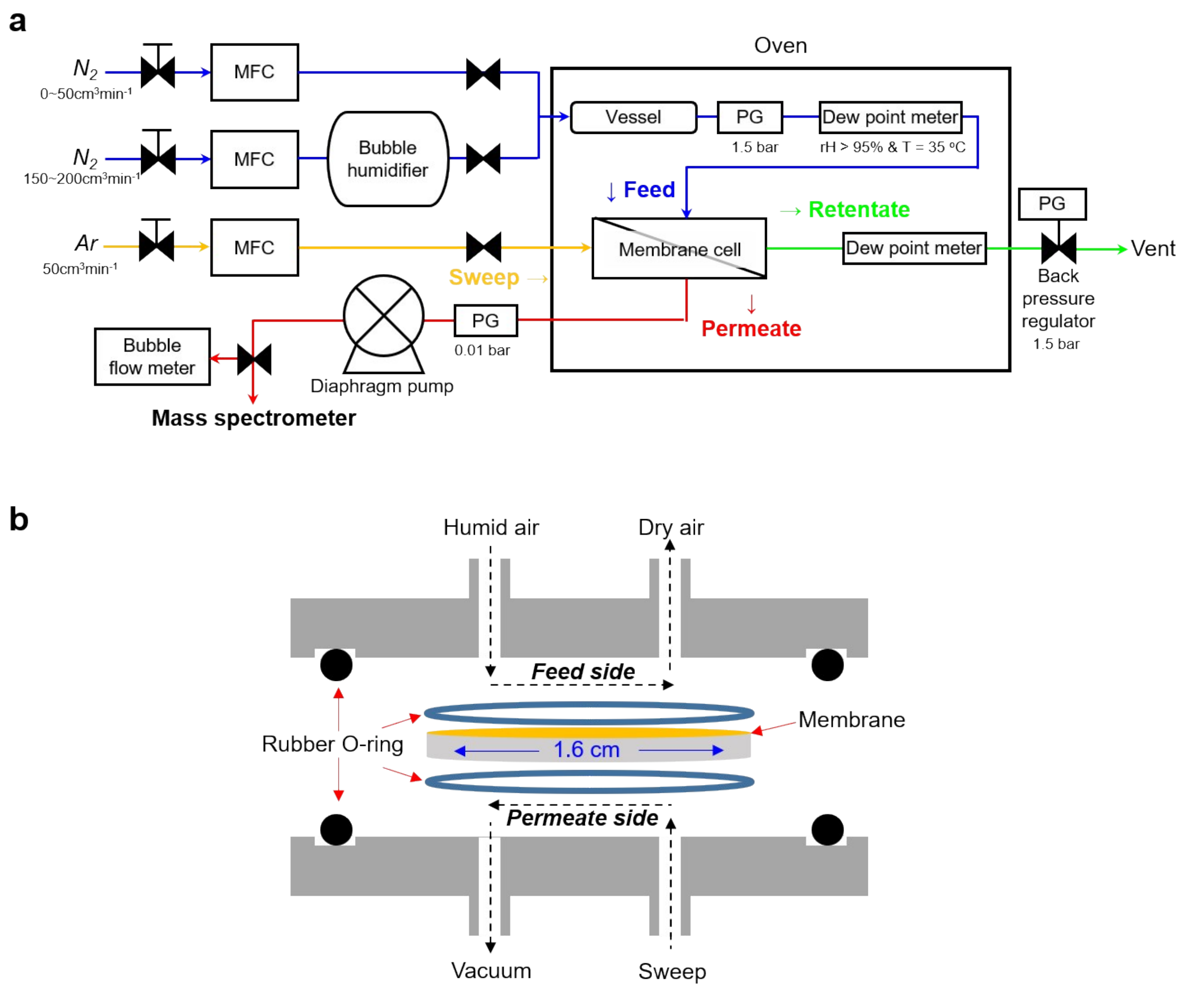

Fig. S1. (a) Schematic diagram of permeation measurement system and (b) design of the membrane test cell.

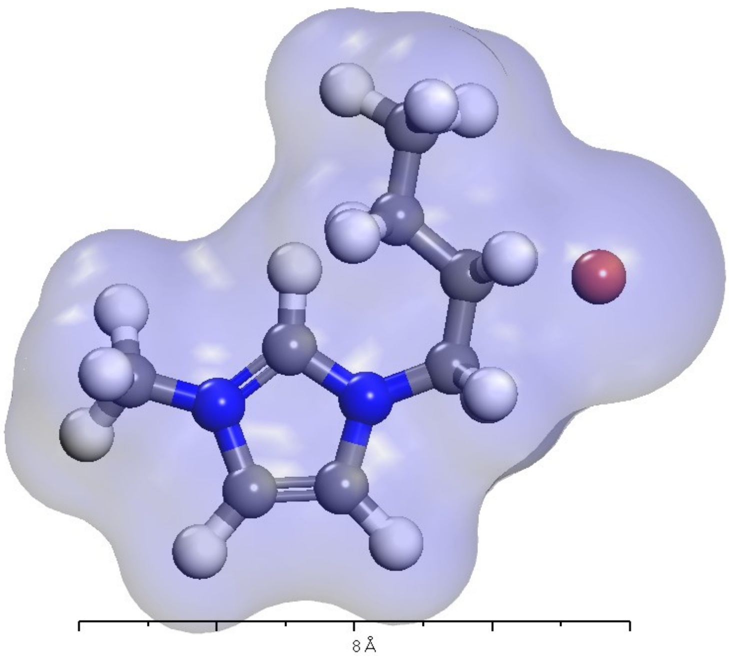

Fig. S2. Structure of [C_4MIM][Br] with an isosurface created by using the Connolly surface method.

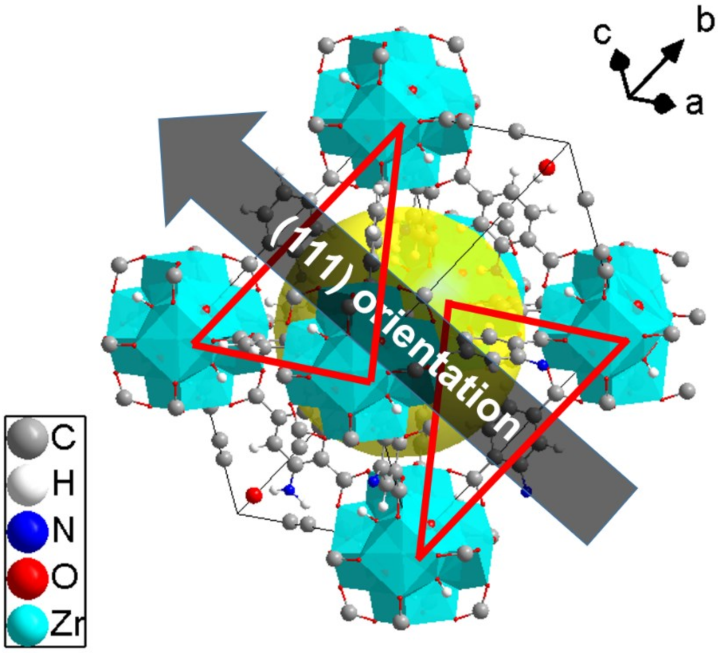

Fig. S3. Illustration of the octahedral cage of UiO-66-NH_2 and the indication of (111) orientation.

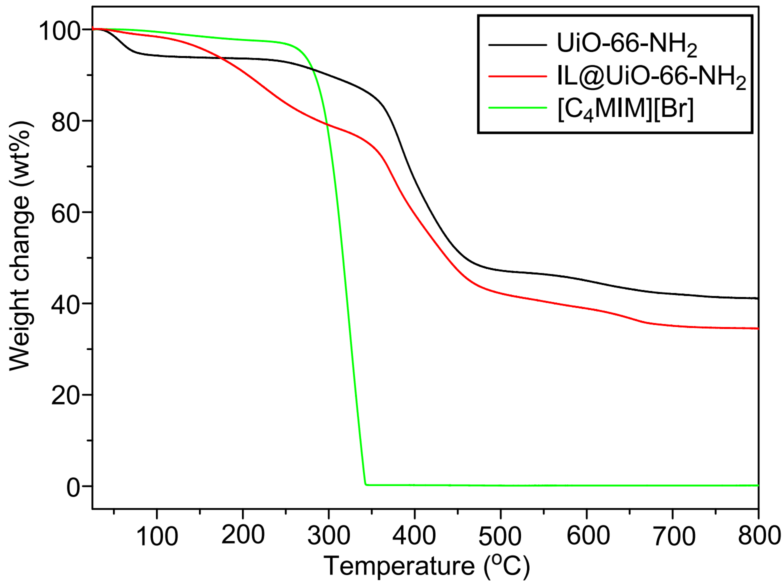

Fig. S4. TGA thermograms of a UiO-66-NH_2 membrane and an IL@UiO-66-NH_2 membrane along with a [C_4MIM][Br] sample under air flow.

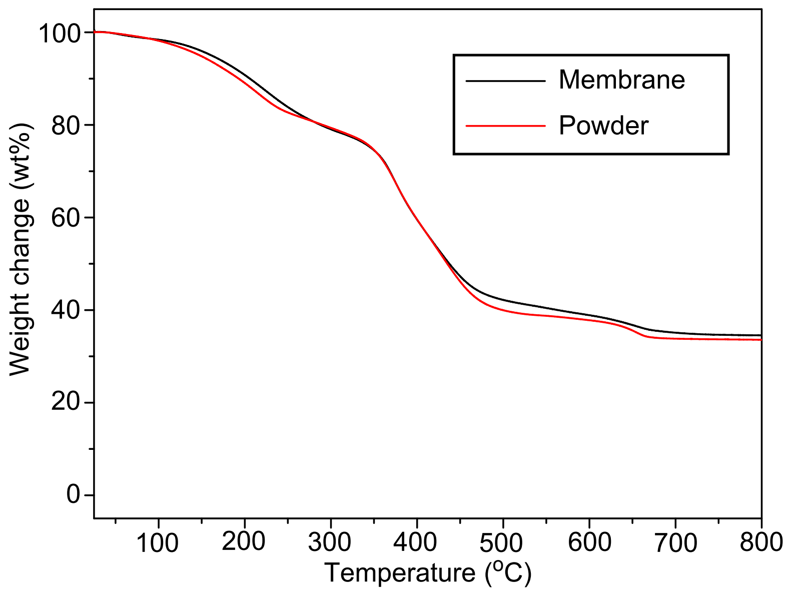

Fig. S5. TGA thermograms of IL@UiO-66-NH_2 membrane and powder samples.

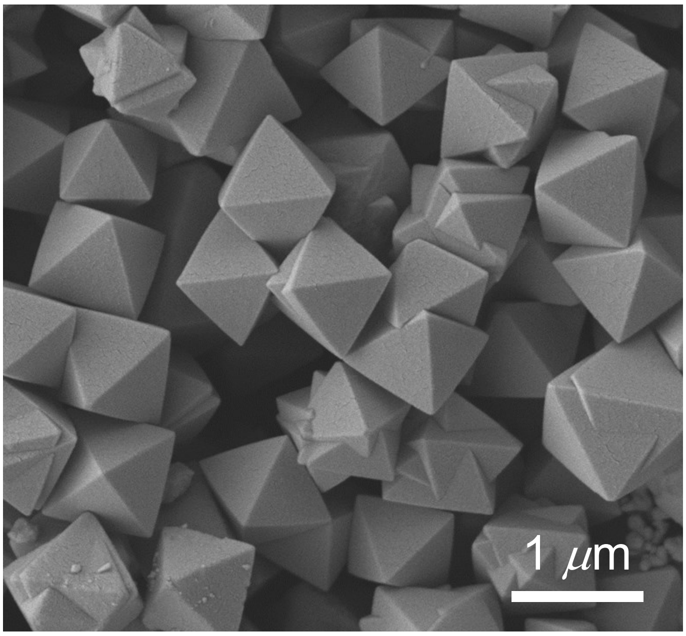

Fig. S6. SEM image of UiO-66-NH_2 particles.

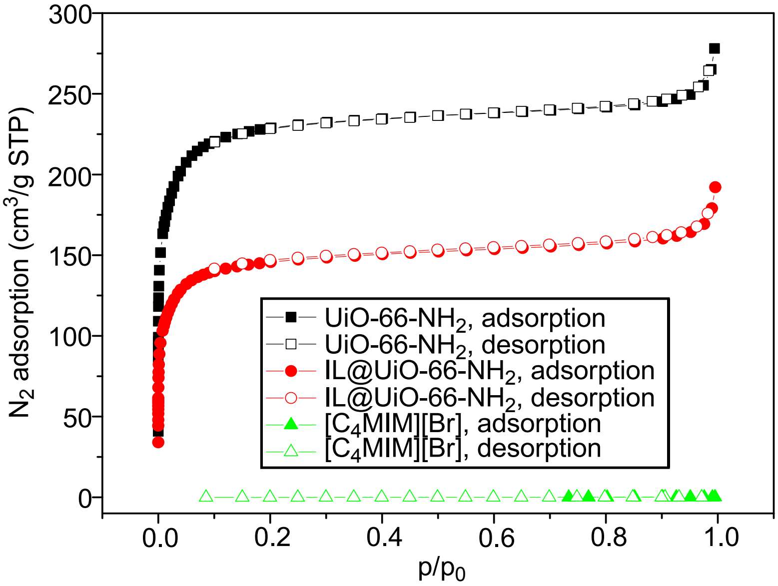

Fig. S7. N_2 adsorption/desorption isotherms at 77 K.

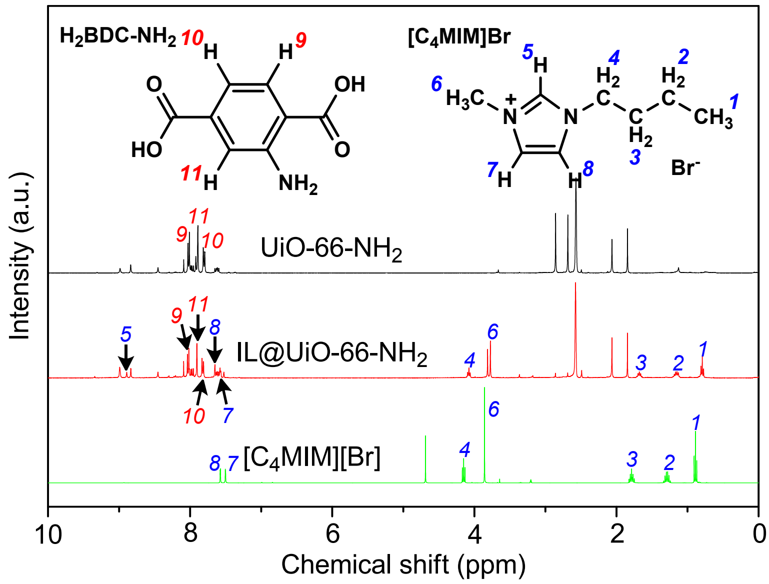

Fig. S8. [1]H NMR spectra of samples dissolved in H_2SO_4 and DMF (1:9, vol) mixture.

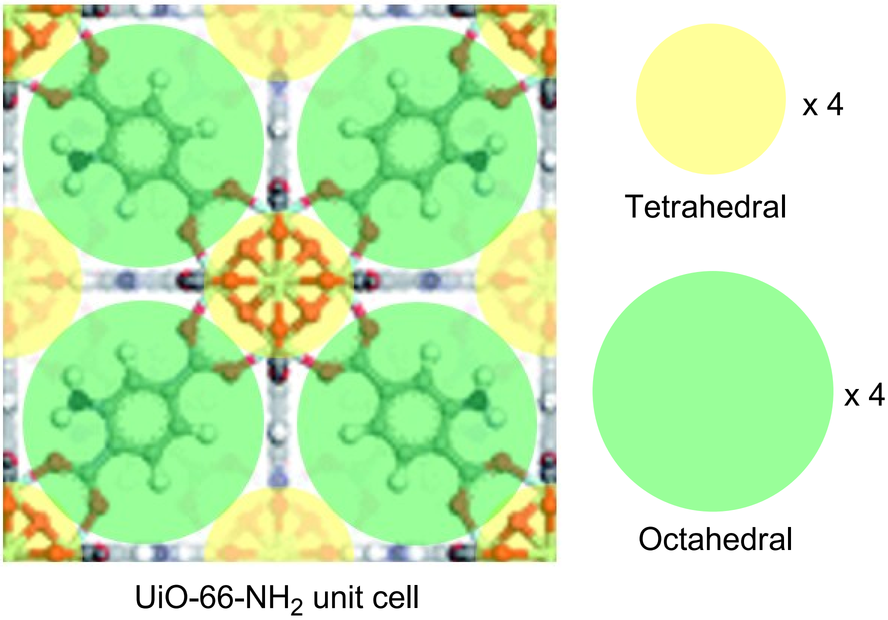

Fig. S9. Illustration of cages of UiO-66-NH_2 unit cell.

Table S1. Heat of sorption (i.e., sorption enthalpy) of water vapor at 1.1 kPa.

UiO-66-NH_2	IL@UiO-66-NH_2	$[C_4MIM][Br]$
-51.76 ± 1.42 kJ mol^{-1}	-54.13 ± 0.85 kJ mol^{-1}	-40.01 ± 11.30 kJ mol^{-1}

Table S2. Effective diffusivity (D_{eff}) of water vapor at 293 K and p/p_o = 0.5.

UiO-66-NH_2	IL@UiO-66-NH_2	$[C_4MIM][Br]$
4.85 ± 0.09 x 10^{-11} cm^2 sec^{-1}	5.98 ± 0.05 x 10^{-11} cm^2 sec^{-1}	1.77 ± 0.63 x 10^{-7} cm^2 sec^{-1}

Diffusion in spherical particle (unipore model)

In the case of UiO-66-NH_2 and IL@UiO-66-NH_2, the fractional uptake of adsorbate diffusing in or out a spherical particle at time t is given as follows;[1]

$$\frac{M_t}{M_\infty} = 1 - \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}e^{\left(-\frac{D_{eff}\,n^2\pi^2 t}{R^2}\right)}. \quad (S1)$$

Where M_t (mmol) and M_∞ (mmol) are moles of adsorbed gas phase molecules at time t and at equilibrium, respectively, and D_{eff} (cm^2/s) is the effective diffusivity, and R (cm) is the radius of particle. In the short time region ($M_t/M_\infty < 0.5$), the kinetic adsorption is essentially linear and Equation S1 can be simplified by [2]

$$\frac{M_t}{M_\infty} \approx \frac{6}{\sqrt{\pi}}\sqrt{\frac{D_{eff}}{R^2}t}. \quad (S2)$$

Diffusion in plane liquid

In case of $[C_4MIM][Br]$, the relative amount of gas uptake diffusing into a plane liquid at time t is expressed by follows;[1]

$$\frac{M_t}{M_\infty} = 1 - \frac{8}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{(2n+1)^2}e^{\left(-\frac{D_{eff}\,(2n+1)^2\pi^2 t}{4L^2}\right)}. \quad (S3)$$

Where L (cm) is the half thickness of plane. For the short time region ($M_t/M_\infty < 0.5$), Equation S3 is approximated by[3]

$$\frac{M_t}{M_\infty} \approx \frac{2}{\sqrt{\pi}}\sqrt{\frac{D_{eff}}{L^2}t}. \quad (S4)$$

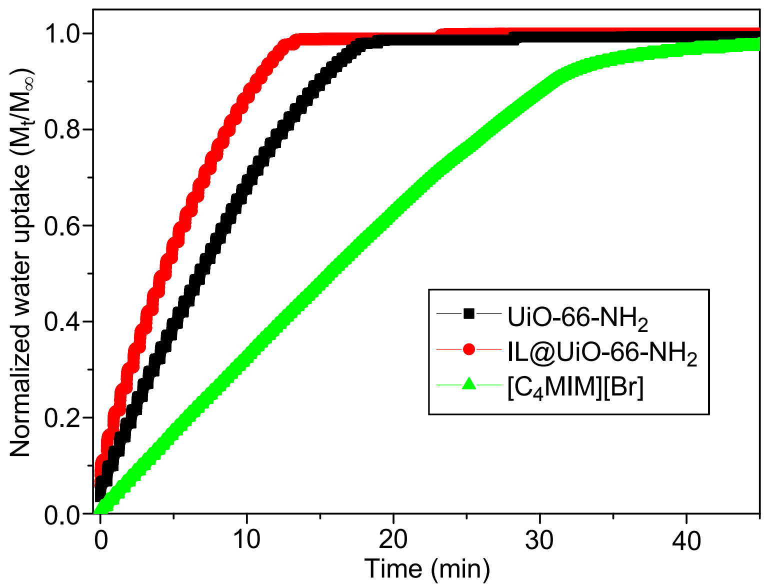

Fig. S10. Kinetics of water vapor adsorption at 295 K and 50% RH for overall dosing.

Table S3. Comparison of H_2O/N_2 separation performances.

Sample	Temperature (°C)	Thickness (μm)	H_2O permeance (mol m^{-2} s^{-1} Pa^{-1})	H_2O permeability (Barrer)	H_2O/N_2 separation factor	Ref.
PEO-PBT	30	30	9.57×10^{-7}	85500	40500	4
PAN	30	12.7	7.9×10^{-9}	300	1875000	5
PEBAX®1074	30	78	8.58×10^{-7}	200000	104167	6
SPEEK	30	51	3.22×10^{-6}	490000	350000	
TMSC	23	1.2	1.34×10^{-6}	4808	11000	7
PVA/LiC/TiO_2	24	n/a	4.57×10^{-7}	n/a	450	8
PVA/TiO_2	24	n/a	5.11×10^{-7}	n/a	5781	
PVA/LiCl	24	180	5.58×10^{-7}	300000	2800	9
Pebax@1657/GO	21	5.7	1.68×10^{-6}	28632	80000	10
PSf/Si-TFN	30	0.4	7.37×10^{-7}	881	501	11
PSf/$D^{1.0}T^{0.2}cTiO_2^{0.2}$	30	0.226	4.49×10^{-7}	303	486	12
ABn-NH-TFN	30	0.31	9.41×10^{-7}	872	913	13
PBI/TiO2	22	20	1.19×10^{-6}	71000	3100000	14
PA/GT	30	n/a	9.45×10^{-7}	n/a	910	15
Zeolite NaA	32	2	6.8×10^{-6}	40664	178	16
GO	30.8	6	1.01×10^{-5}	182000	10000	17
GLX-cross-linked GO		1.64	1.68×10^{-5}	82380	2800000	18
[C_2MIM][Tf_2N]	31	132	2.1×10^{-7}	82883	3843	19
[C_2MIM][BF_4]	31	132	3.5×10^{-7}	138138	16300	
[N_{4111}][Tf_2N]	31	132	1.9×10^{-7}	74989	3290	
TEG	30	47	5.7×10^{-8}	8010	2500	20
PEG-400	22	67	5×10^{-8}	10017	2000	
[C_2MIM][DCA]	25	11.5	1.6×10^{-6}	55016	> 1000	21
[C_2MIM][ESU]	25	10.3	1.5×10^{-6}	46196	> 1000	
[C_2MIM][BF_4]	25	9.9	1.2×10^{-6}	35521	> 1000	
TEG	25	12.8	7.2×10^{-7}	27556	> 1000	
UiO-66-NH_2	22	~ 2	$6.10 \pm 1.52 \times 10^{-5}$	440000 ± 120000	312 ± 68	This work
IL@UiO-66-NH_2	22	~ 2	$2.36 \pm 0.59 \times 10^{-4}$	1580000 ± 110000	1564 ± 218	
[C_4MIM][Br]	22	~ 58	$1.57 \pm 0.50 \times 10^{-6}$	270000 ± 90000	4206 ± 1182	

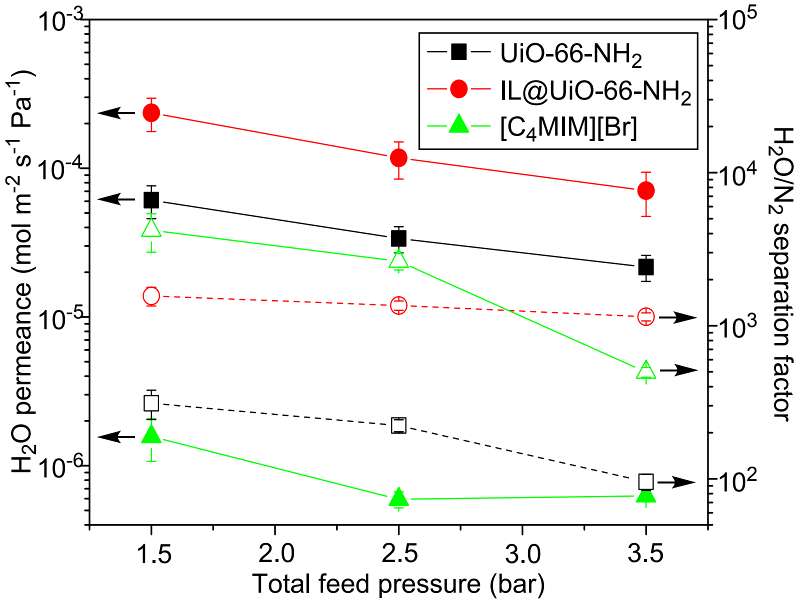

Fig. S11. H_2O/N_2 separation performances of samples under various feed pressure at 22 °C.

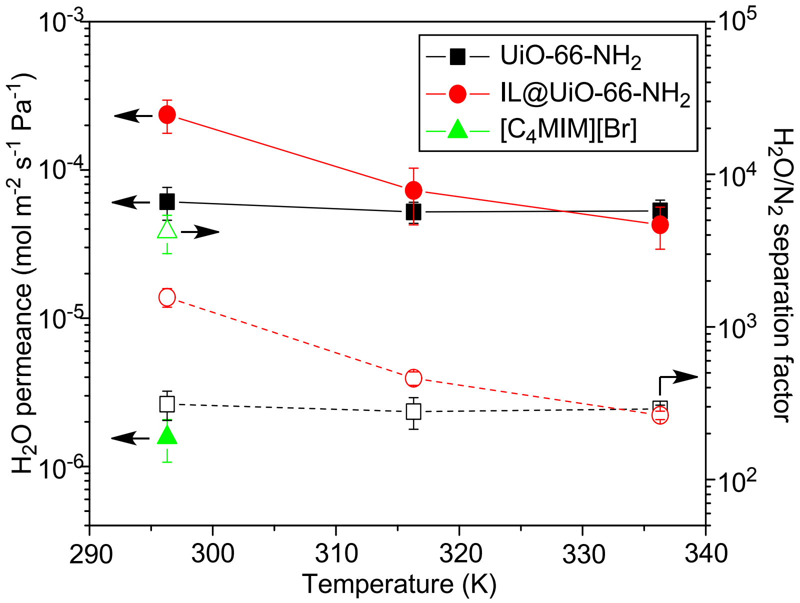

Fig. S12. H_2O/N_2 separation performances of samples under various temperatures at 1.5 bar of feed pressure.

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