Supporting Information

Synthesis of Compound-1: A mixture of 2,5-furandicarboxylic acid (0.03g), $In(NO_3)_3 \cdot xH_2O$ (0.03g), 2-(2-aminoethylamino)ethanol (0.01g), concentrated nitric acid (0.02g), dimethylacetamide (1.02g) and methanol (4.10g) was stirred in a 23ml vial for 25 minutes and then heated at 120℃ for 3 days. After cooled to room temperature and washed by methanol, brown crystals were obtained.

Synthesis of Compound-2: A mixture of 2,5-furandicarboxylic acid (0.03g), $In(NO_3)_3 \cdot xH_2O$ (0.015g), 2-(2-aminoethylamino)ethanol (0.01g), concentrated nitric acid (0.02g), dimethylacetamide (4.01g) and methanol (1.02g) was stirred in a 23ml vial for 25 minutes and then heated at 120℃ for 3 days. After cooled to room temperature and washed by methanol, brown crystals were obtained.

Synthesis of Compound-3: A mixture of 2,5-furandicarboxylic acid (0.015g), $In(NO_3)_3 \cdot xH_2O$ (0.03g), 3-amino-5-mercapto-1,2,4-triazole (0.012g), tetramethylurea (1.08g) and water (4.01g) was stirred in a 23ml vial for 25 minutes and then heated at 80℃ for 3 days. After cooled to room temperature and washed by water, clear crystals were obtained.

Measurements:

Powder X-ray diffraction: The experiments were performed on a Bruker D8 Advance X-ray powder diffractometer operating at 40 kV and 40 mA (CuKα radiation, λ =1.5418 Å). The simulated powder XRD pattern was obtained based on the single-crystal data.

Single crystal X-ray diffraction: The measurements were performed on a Bruker APEX II diffractometer with nitrogen-flow temperature controller using graphite-monochromated MoKα radiation (λ = 0.71073 Å), operating in the ω and φ scan mode.

Thermal analysis: The thermogravimetric analysis (TGA) was performed on a TA Instruments TGA Q500 apparatus in the temperature range of 30 °C to 800 °C under N_2 flow at a heating rate of 5 °C/min.

Gas adsorption measurement: The measurements were performed on a Micromeritics ASAP 2020 surface-area and pore-size analyzer.

Table S1.Crystal Data and Structure Refinements for the In-MOFs in this study

Compound reference	Compound-1	Compound-2	Compound-3
Chemical formula	$C_{18}H_{12}In_3O_{19}$•(NO_3)	$C_{12}H_4InO_{10}$•(C_2H_8N)	$C_{12}H_8In_2O_{13}$
Formula Mass	938.70	469.70	589.79
Crystal system	Rhombohedral	Tetragonal	Monoclinic
a/?	13.9508(19)	9.7460(4)	10.7435(6)
b/?	13.9508(19)	9.7460(4)	21.4819(12)
c/?	22.402(7)	31.081(2)	10.7721(7)
α/°	90.00	90.00	90.00
β/°	90.00	90.00	105.1130(10)
γ/°	120.00	90.00	90.00
Unit cell volume/$^?$	3775.9(14)	2952.2(3)	2400.1(2)
Temperature/K	195(2)	195(2)	195(2)
Space group	*R*32	*I*4(1)/*amd*	*P*2(1)/*m*
No. of formula units per unit cell, *Z*	3	4	4
No. of reflections measured	7331	6628	8139
No. of independent reflections	1277	725	4274
R_{int}	0.0502	0.0276	0.0252
Final R_1 values ($I > 2\sigma(I)$)	0.0571	0.0445	0.0279
Final $wR(F^2)$ values ($I > 2\sigma(I)$)	0.1850	0.1263	0.0774
Final R_1 values (all data)	0.0579	0.0461	0.0376
Final $wR(F^2)$ values (all data)	0.1863	0.1285	0.0804
Goodness of fit on F^2	1.035	1.040	1.036

$R_1 = \sum||F_o| - |F_c||/\sum|F_o|$, $wR = \{\sum w[(F_o)^2 - (F_c)^2]^2/\sum w[(F_o)^2]^2\}^{1/2}$.

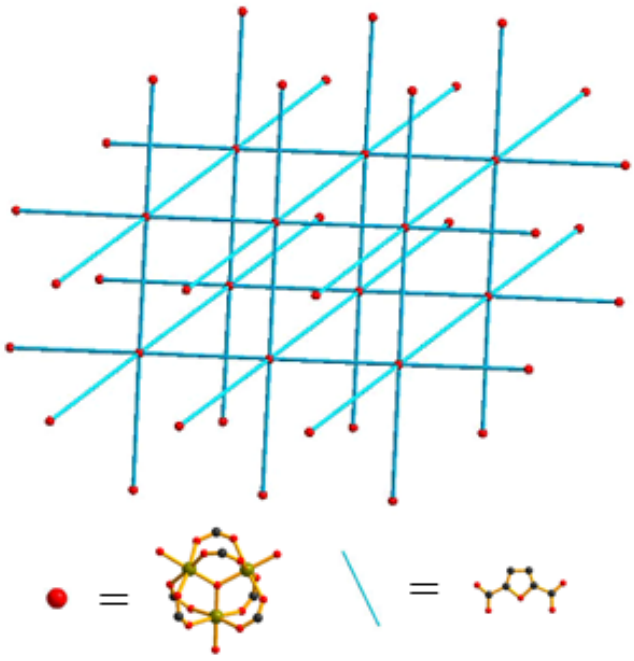


Figure S1. The pcu network formed by indium trimers and FDA.

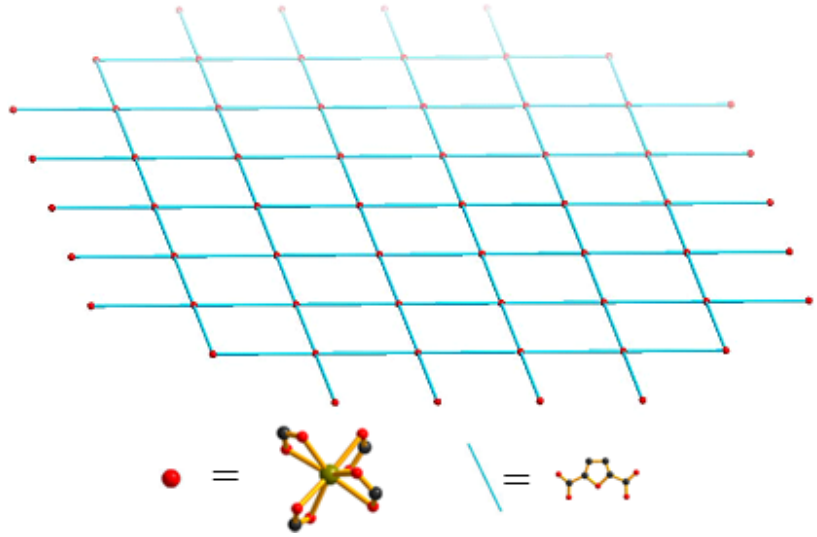
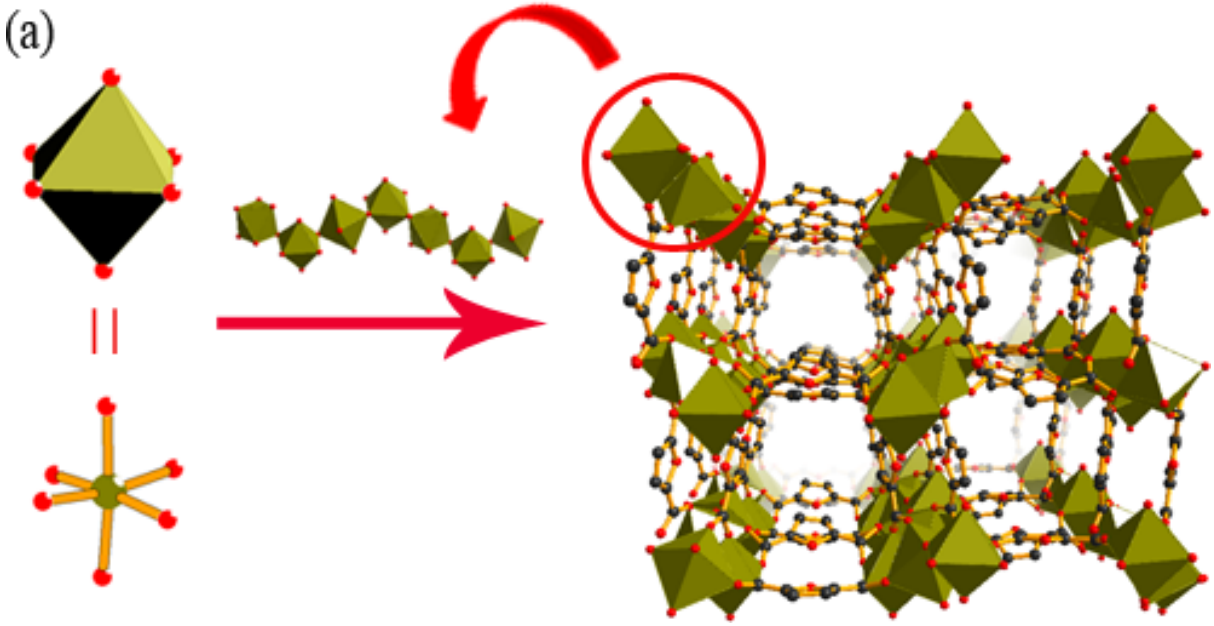


Figure S2. The 2D network formed by indium monomers and FDA.



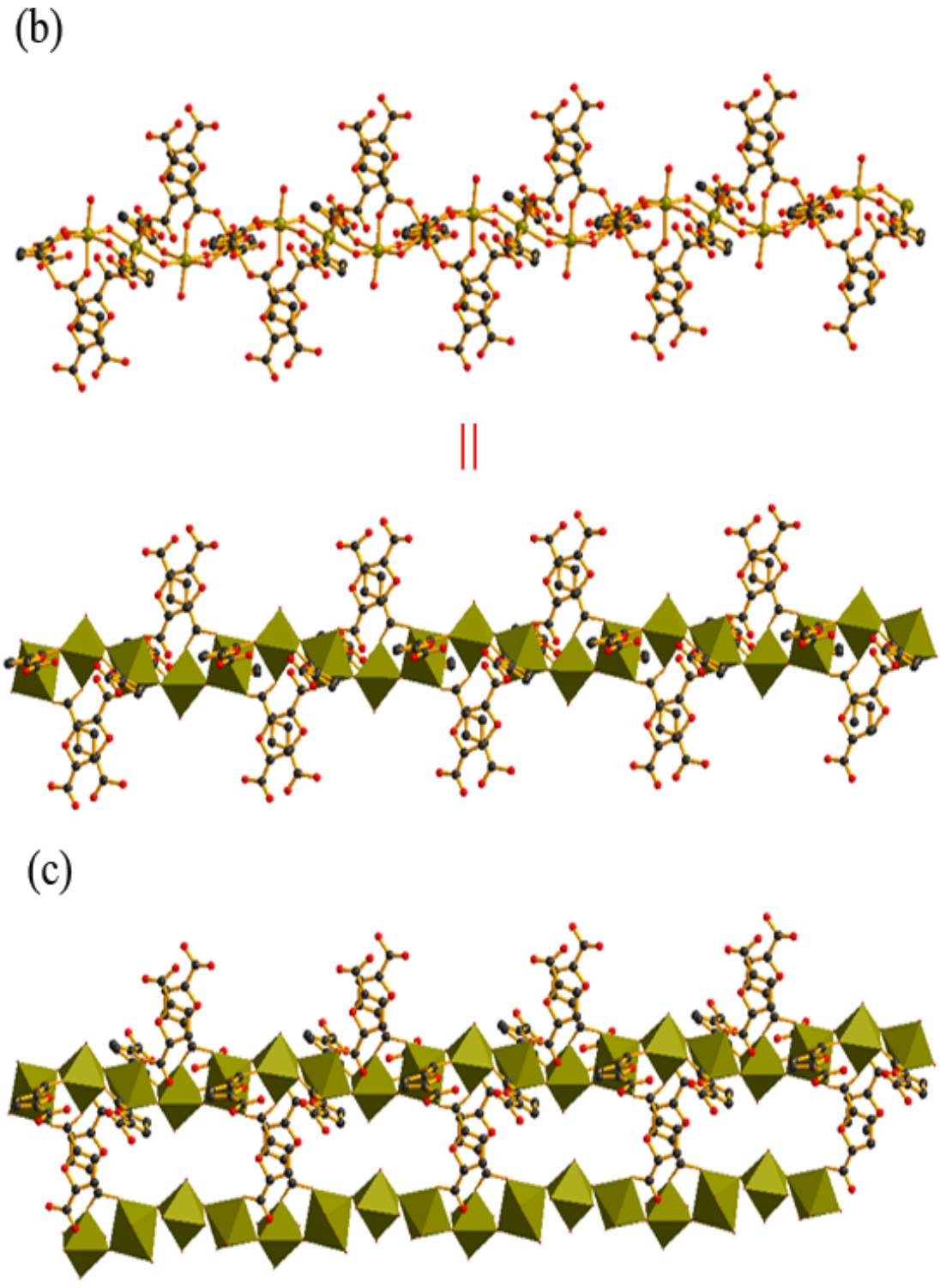


Figure S3. (a) 3D framework, Compound-3, formed by octahedrally coordinated indiums; (b)The construction of chain by octahedrally coordinated indiums in Compound-3; (c) The connection between two chains.

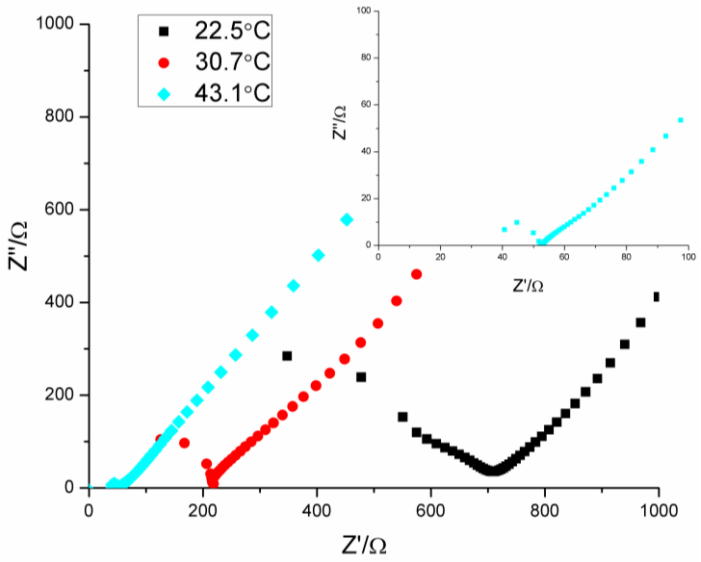
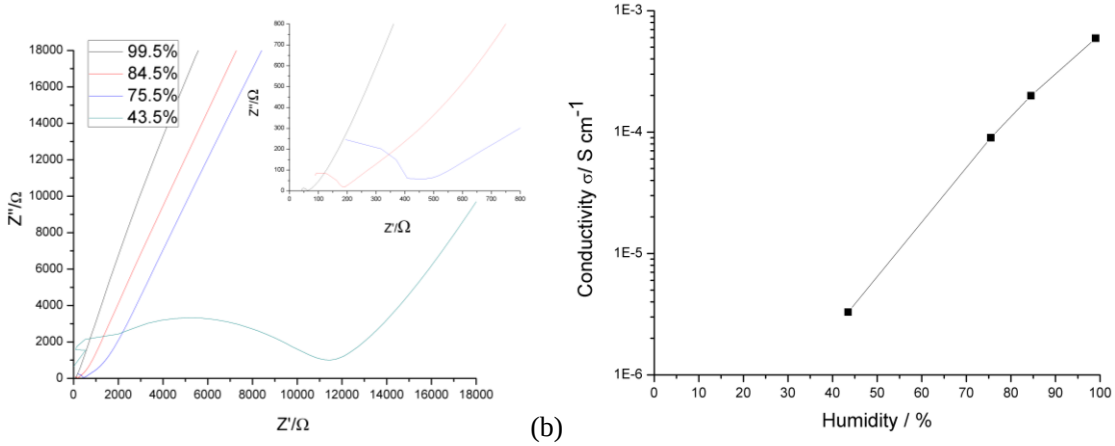
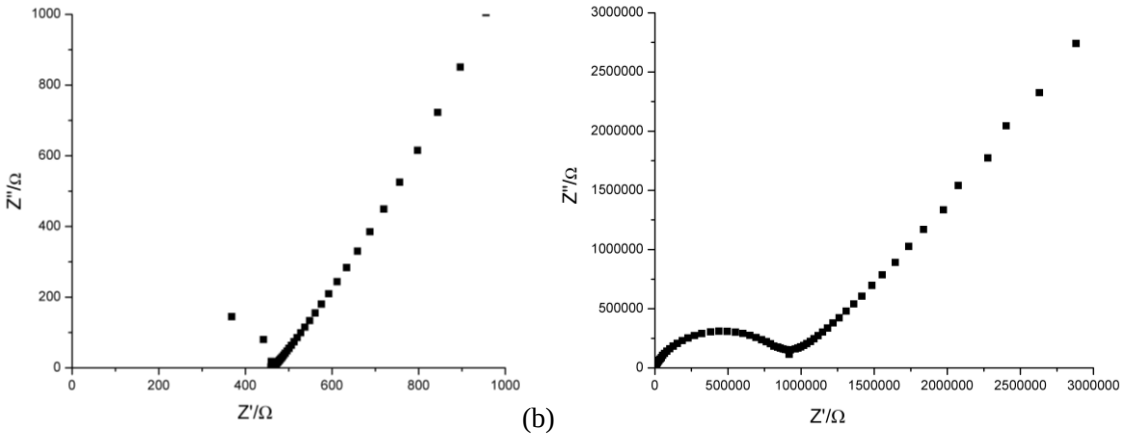


Figure S4. Nyquist plots of the pellet sample Compound-1 at various temperatures and 99.5% RH condition, S=1.327cm^2, L＝0.097cm. The inset shows Nyquist plot of the pellet sample Compound-1 at 43.1 ℃ and 99.5% RH condition.



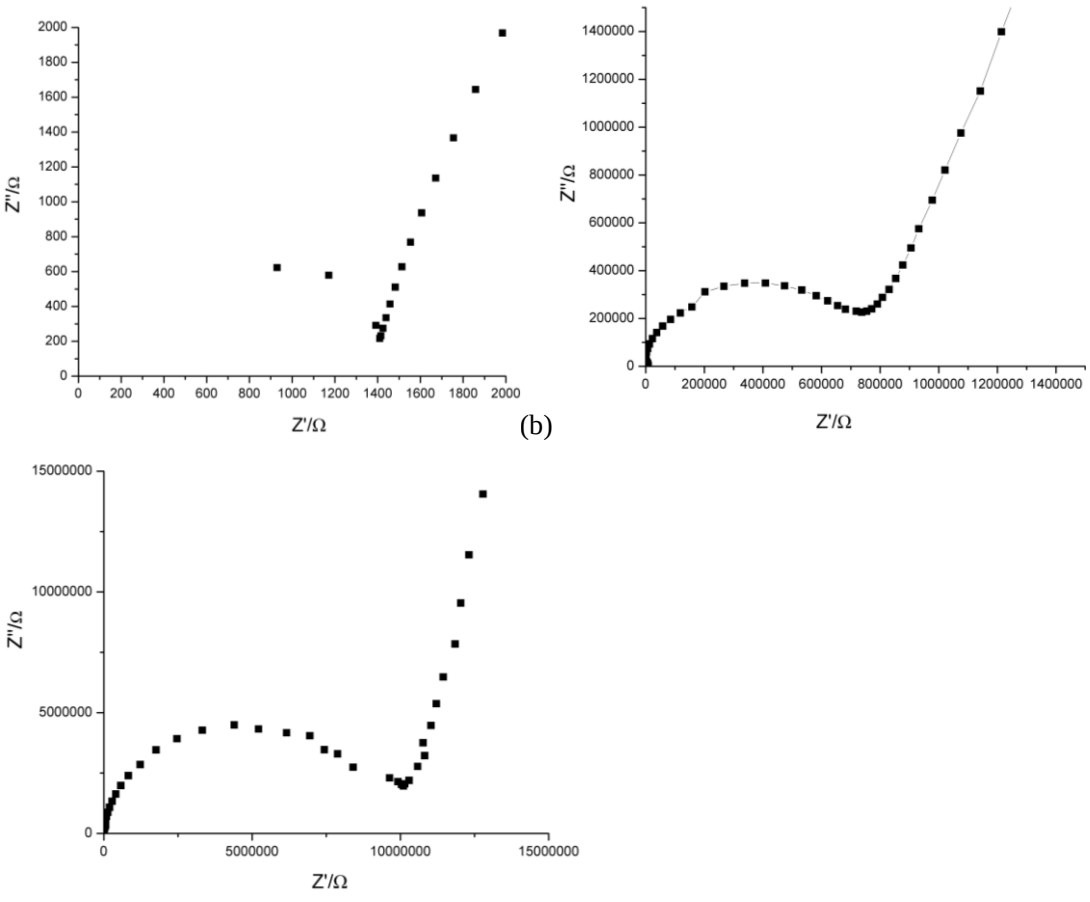
(a) (b)

Figure S5. (a) Nyquist plots of the pellet sample Compound-1 at 22.5℃ and various RH condition, S=1.327cm^2, L＝0.050cm. The inset shows Nyquist plots of Compound-1 at 22.5℃ and 43.5% RH, 75.5% RH and 84.5% RH; (b) Humidity dependence of conductivity at 25 ℃ for the pellet sample Compound-1.



(a) (b)

Figure S6. Nyquist plots of the pellet sample Compound-2 at 22.5°C and 99.5% RH (a) and 84.5% RH (b), S=1.327cm^2, L＝0.114cm.



(a) (b)

(c)

Figure S7. Nyquist plots of the single crystal sample Compound-2 at 22.5°C and 99.5% RH (a), 84.5% RH (b) and 75.5% RH (c), S=4.57×10^{-5}cm^2, L＝0.0099cm.

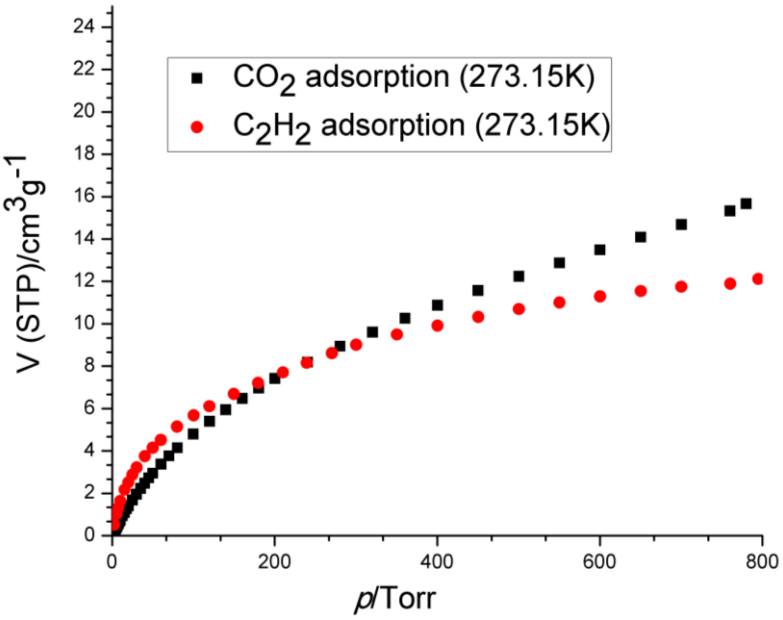


Figure S8. CO_2 and C_2H_2 adsorption isotherms of Compound-1.

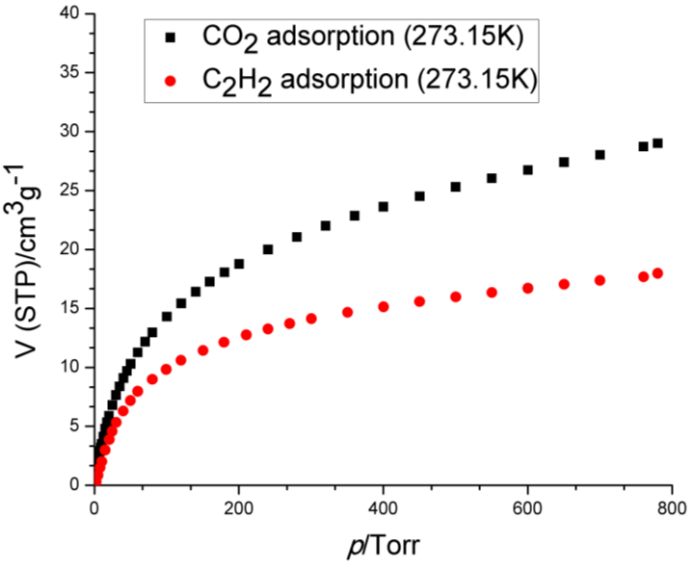


Figure S16. CO_2 and C_2H_2 adsorption isotherms of Compound-3.