Supporting Information

Selective Gas Adsorption in a Magnesium-Based Metal-Organic Framework

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Experimental Details

General Method. All chemicals and solvents used in the syntheses were of reagent grade and were used without further purification.THF was purified by distillation. Infrared spectra were recorded with a Perkin Elmer Spectrum One FT-IR spectrophotometer. UV/vis diffuse reflectance spectra were recorded on a Perkin Elmer Lambda 35 UV/vis spectrophotometer. Elemental analyses were performed on a Perkin Elmer EA 2400 analyzer. Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) were performed under N_2 at a scan rate of 5 °C/min using TGA Q50 and DSC Q10 of TA instruments, respectively. Powder X-ray diffraction (PXRD) data were recorded on a Bruker D5005 diffractometer at 40 kV and 40 mA for Cu Kα (l = 1.54050Å) with a scan speed of 5°/min and a step size of 0.02° in 2θ. An ESR spectrum was recorded on a JEOL JES-TE200.

Synthesis of *N,N,N',N'*-tetracarboxyphenylbenzidine (H_4TCPBDA). The compound was prepared by modifying the method previously reported.[S1] *N,N,N',N'*-Tetraphenylbenzidine (2 g, 4.0 mmol) was dissolved in chloroform (80 mL), and the chloroform solution (20 mL) of Br_2 (0.6 mL, 12.0 mmol) was slowly added to the solution in an ice bath. After stirring the solution at 0 °C for 45 min, the residue was added to a hot EtOH (500 mL) and then cooled to 0 °C until microcrystalline white solid formed. The resulting white precipitate, *N,N,N',N'*-tetrabromophenylbenzidine was filtered, washed with EtOH, and dried under reduced pressure. Yield: 1.86 g (58%). ^{1}H-NMR, ($CDCl_3$): δ 7.5 (d, 4H), 7.4 (d, 8H), 7.1 (d, 4H), 7.0 ppm (d, 8H). To a THF solution (80mL) of *N,N,N',N'*-tetrabromophenylbenzidine (1.86 g, 2.3 mmol), was slowly added 1.6 M *n*-butyllithium in hexane (20 mL) at -78 °C. After 1 h, crushed dry ice was added to the resultant, which produced white precipitates immediately. Acetic acid was added to the greenish yellow suspension in THF for acidification until precipitate was dissolved. The resulting solution was added to mixture of water and MeOH (10:1 v/v). The resulting greenish yellow precipitate (H_4TCPBDA) was filtered, washed with water, and dried under reduced pressure by heating. Yield: 1.25 g (80%). ^{1}H-NMR, (DMSO-d_6): δ 7.8 (d, 8H), 7.7 (d, 4H), 7.4 (d, 4H), 7.1 ppm (d, 8H). FT-IR (KBr pellet): $\nu_{C=O}$, 1688 (s); ν_{C-OH}, 1594 (s) cm^{-1}.

Synthesis of [Mg(TCPBDA)(H_2O)$_2$]·6DMF·6H_2O (1). The DMF (8 mL) solution of *N,N,N',N'*-tetracarboxylphenylbenzidine (H_4TCPBDA) (134 mg, 0.2 mmol) and the H_2O/EtOH (2 mL/ 2 mL) solution of $Mg(NO_3)_2 \cdot 6H_2O$ (156 mg, 0.6 mmol) were mixed in a Teflon vessel within the autoclave, which was heated at 110 °C for 24 h and then cooled to room temperature. The crystal also formed using MeOH instead of EtOH. Yellow plate-shape crystals formed, which were filtered, and washed briefly with DMF. Yield: 106 mg (28.5%). Anal. Calcd for $MgC_{58}H_{82}N_8O_{22}$: C, 54.96; H, 6.52; N, 8.84. Found: C, 54.66; H, 6.70; N, 8.84. FT-IR (Nujol, cm^{-1}): $\nu_{OH(water)}$, 3391 (m, br); $\nu_{C=O(DMF)}$, 1660 (s); $\nu_{OH(water)}$, 1614 (w); $\nu_{O-C=O(carboxylate)}$, 1596 (s). UV/vis (diffuse reflectance spectrum, λ_{max}): 397 nm. Solid luminescence: 478 nm (excitation at 380 nm).

Synthesis of [Mg(TCPBDA)] (SNU-25). Solid **1** was dried in a Schlenk tube at 210 °C under vacuum for 4 h. Anal. Calcd for $MgC_{40}H_{28}N_2O_{10}$: C, 70.14; H, 3.53; N, 4.09. Found: C, 66.58; H, 3.50; N, 3.94. FT-IR (Nujol, cm^{-1}): $\nu_{OH(water)}$, 3349 (w, br); $\nu_{O-C=O(carboxylate)}$, 1595 (s). UV/vis (diffuse reflectance spectrum, λ_{max}): 398 nm. Solid luminescence: 513 nm (excitation at 380 nm).

X-ray Crystallography. Diffraction data of **1** was collected with an Enraf Nonius Kappa CCD diffractometer (MoKα, λ = 0.71073 Å, graphite monochromator). Preliminary orientation matrices and unit cell parameters were obtained from the peaks of the first ten frames and then refined using the whole data set. Frames were integrated and corrected for Lorentz and polarization effects by using DENZO.[S2] The scaling and the global refinement of crystal parameters were performed by SCALEPACK.[S2] No absorption correction was made. The crystal structure was solved by direct method[S3] and refined by full-matrix least-squares refinement using SHELXL-97 computer program.[S4] The positions of all non-hydrogen atoms were refined with anisotropic displacement factors. The hydrogen atoms were positioned geometrically and refined using a riding model. Electron densities of the disordered guest molecules were flattened by using 'SQUEEZE' option of PLATON.[S5] The crystallographic data and selected bond distances and angles are summarized in Tables S1 and S2 in the

Supporting Information**.** CCDC-729639 contains crystallographic data. These data can be obtained free of charge from the Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.

Low-Pressure Gas Sorption Study Gas sorption isotherms of **SNU-25** were measured on a Quantachrome Autosorb-3B instrument up to 1 atm of gas pressure by the static volumetric method. For **SNU-25**, solid **1** was desolvated in a Schlenk tube at 200 °C under vacuum for 1 h, and then reactivated in the gas sorption apparatus at 210 °C for 4 h. All gases used were of 99.999% purity, and the impurity trace water was removed by passing the gases through the molecular sieve column equipped in the gas line. The gas sorption isotherm of **SNU-25** for N_2 was measured at 77 K and 195 K, and those for H_2 and O_2 at 77 K and 87 K. The gas sorption isotherms for CO_2 and CH_4 were measured at 195 K, 273 K and 298 K.

Estimation of Isosteric Heat of H_2 Adsorption. The isosteric heat of H_2 adsorption was estimated for **SNU-25** from the H_2 sorption data measured at 77 K and 87 K, by using a virial-type expression (eq 1).[S6,S7] In eq 1, P is pressure (atm), N is the amount adsorbed H_2 gas ($mg \cdot g^{-1}$), T is temperature (K), and m and n represent the number of coefficients required to adequately describe the isotherms. The parameters a_i and b_i are independent of temperature. The equation was fit with the **R** statistical software package,[S8] and m and n were gradually increased until the contribution of extra added a and b coefficients became statistically insignificant in the overall fit.

$$\ln P = \ln N + \frac{1}{T}\sum_{i=0}^{m} a_i N^i + \sum_{i=0}^{n} b_i N^i \qquad (1)$$

To estimate the values of the isosteric heat of H_2 adsorption, eq 2 was applied, where R is the universal gas constant. The isotherms and fitted virial parameters are presented in Figure S7.

$$Q_{st} = -R\sum_{i=0}^{m} a_i N^i \qquad (2)$$

(S1) S. Dapperheld, E. Steckhan, K.-H. G. Brinkhaus and T. Esch, *Chem. Ber.*, 1991, **124**, 2557-2567.

(S2) Z. Otwinowsky, W. Minor, *Processing of X-ray Diffraction Data Collected in Oscillation Mode, Methods in Enzymology*; D. W. Carter, R. M. Sweet, Eds.:Academic Press: New York, 1996, Vol. 276, pp 307-326.

(S3) G. M. Sheldrick, *Acta Crystallogr.*, 1990, **A46**, 467.

(S4) G. M. Sheldrick, SHELXL97, Program for crystal structure refinement, University of Gottingen, Germany, 1996.

(S5) P. v. d. Sluis and A. L. Spek, *Acta Crystallogr. Sect. A*, 1990, ***46***, 194.

(S6) M. Dinca, A. Dailly, Y. Liu, C. M. Brown, D. A. Neumann and J. R. Long, *J. Am. Chem. Soc.*, 2006, **128**, 16876-16883.

(S7) (a) J. L. C. Rowsell and O. M. Yaghi, *J. Am. Chem. Soc.*, 2006, **189**, 1304-1315; (b) L. Czepirski and J. Jagiello, *Chem. Eng. Sci.*, 1989, **44**, 797–801.

(S8) **R** statistical software package is available online at http://www.r-project.org.

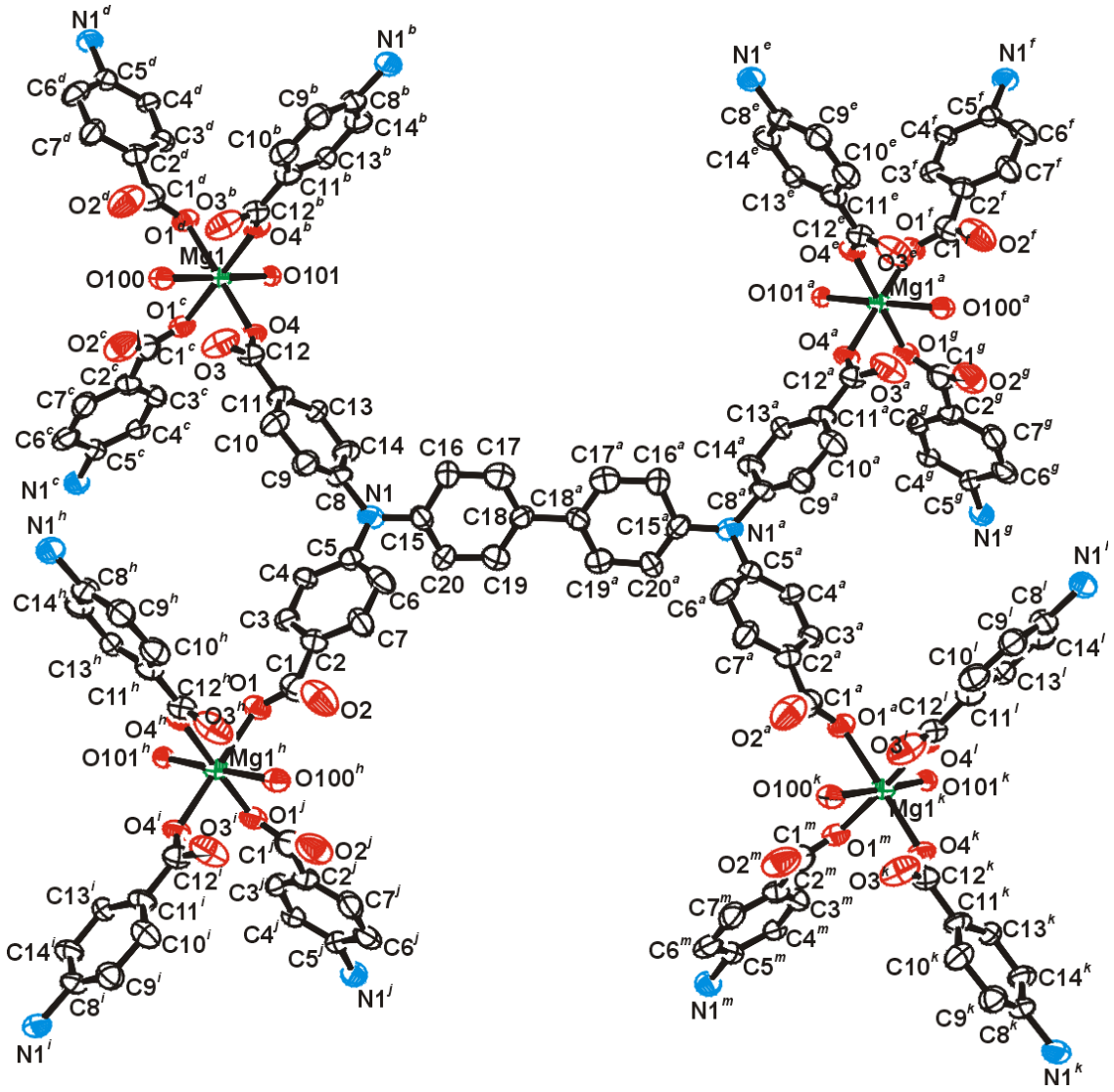

Fig. S1 An ORTEP drawing of **1** with atomic numbering scheme (thermal ellipsoids at 30% probability). Symmetry operation: *a*, x, -y+1/2, z; *b*, -x+1, y, z; *c*, -x+1/2, -y+1, z-1/2; *d*, x+1/2, -y+1, z-1/2; *e*, -x+1, -y+1/2, z; *f*, x+1/2, y-1/2, z-1/2; *g*, -x+1/2, y-1/2, z-1/2; *h*, -x+1/2, -y+1, z+1/2; *i*, x-1/2, -y+1, z+1/2; *j*, -x, y, z; *k*, x-1/2, y-1/2, z+1/2; *l*, -x+1/2, y-1/2, z+1/2; *m*, -x, -y+1/2, z.

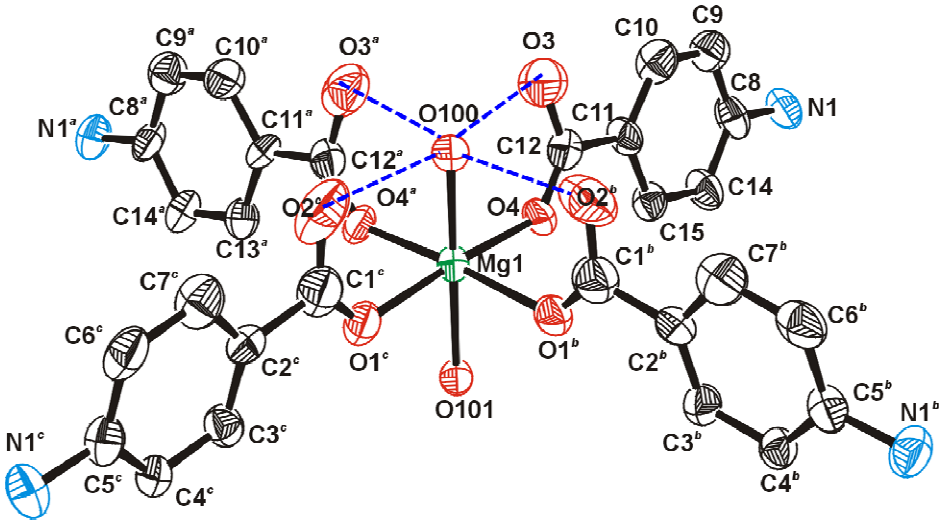

Fig. S2 An ORTEP drawing of **1**, showing the coordination environment around Mg^{II} ion (thermal ellipsoids at 30% probability). Hydrogen bonding interactions between a coordinated aqua ligand and the free carboxylate oxygen atoms are indicated as dotted lines. Symmetry operation: *a*, -x+1, y, z; *b*, -x+1/2, -y+1, z-1/2; *c*, x+1/2, -y+1, z-1/2.

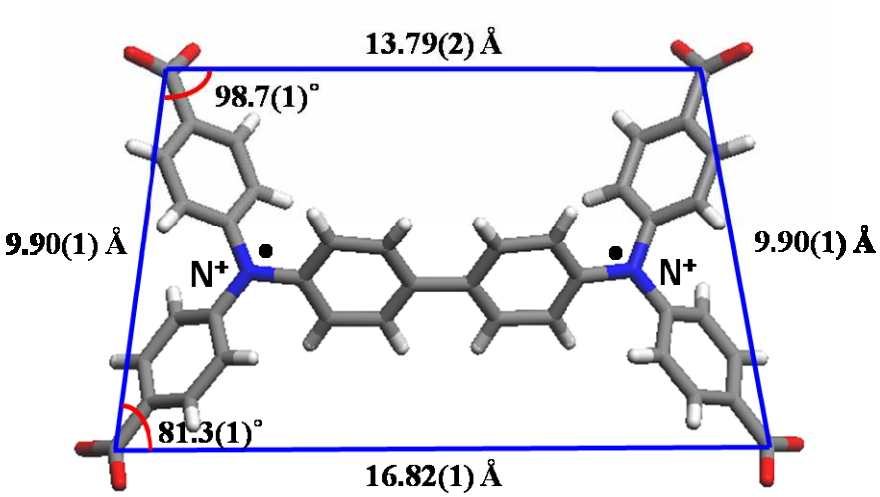

Fig. S3 The trapezoidal shape of $TCPBDA^{2-}$ ligand in **1**.

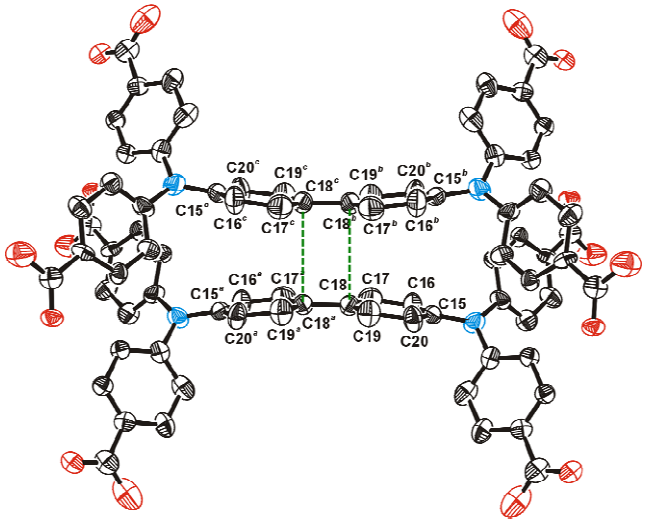

Fig.S4 An ORTEP drawing of **1**, showing the π-π interactions between the phenyl rings of $TCPBDA^{2-}$ belonging to two interpenetrated nets (----). The closest C···C distances are C18···$C18^{b}$, 3.80(1) Å. Symmetry operation: *a*, x, -y+1/2, z; *b*, -x+1/2, y, -z+1/2; *c*, -x+1/2, -y+1/2, -z+1/2.

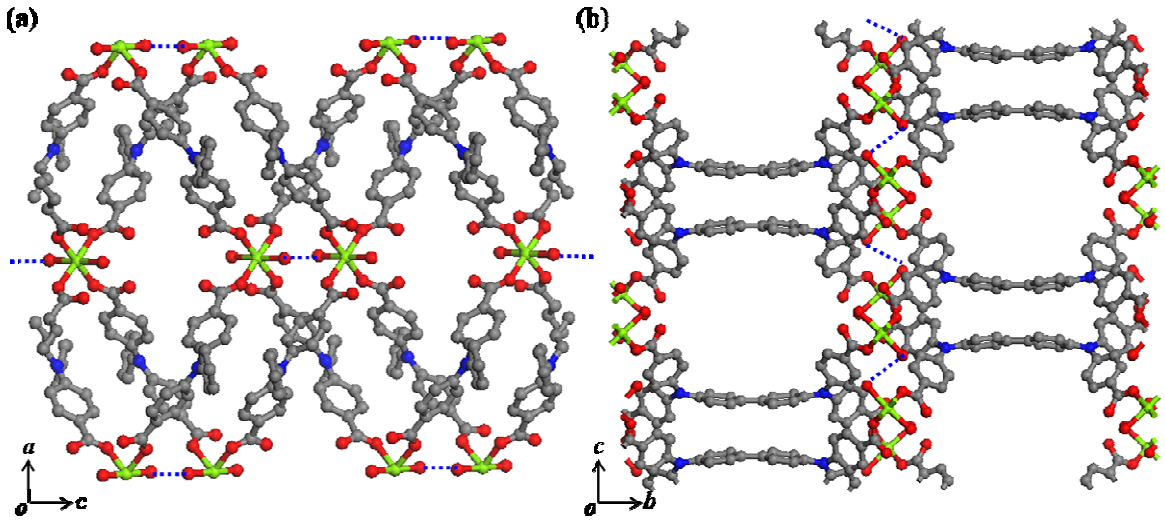

Fig. S5 X-ray crystal structure of **1**, showing the hydrogen bonding interactions between the aqua ligands coordinated to Mg^{II} ions (blue dotted line). The closest O···O distance: O101···$O101^{a}$, 3.23(1) Å. Symmetry operation: *a*, x, -y+1, -z.

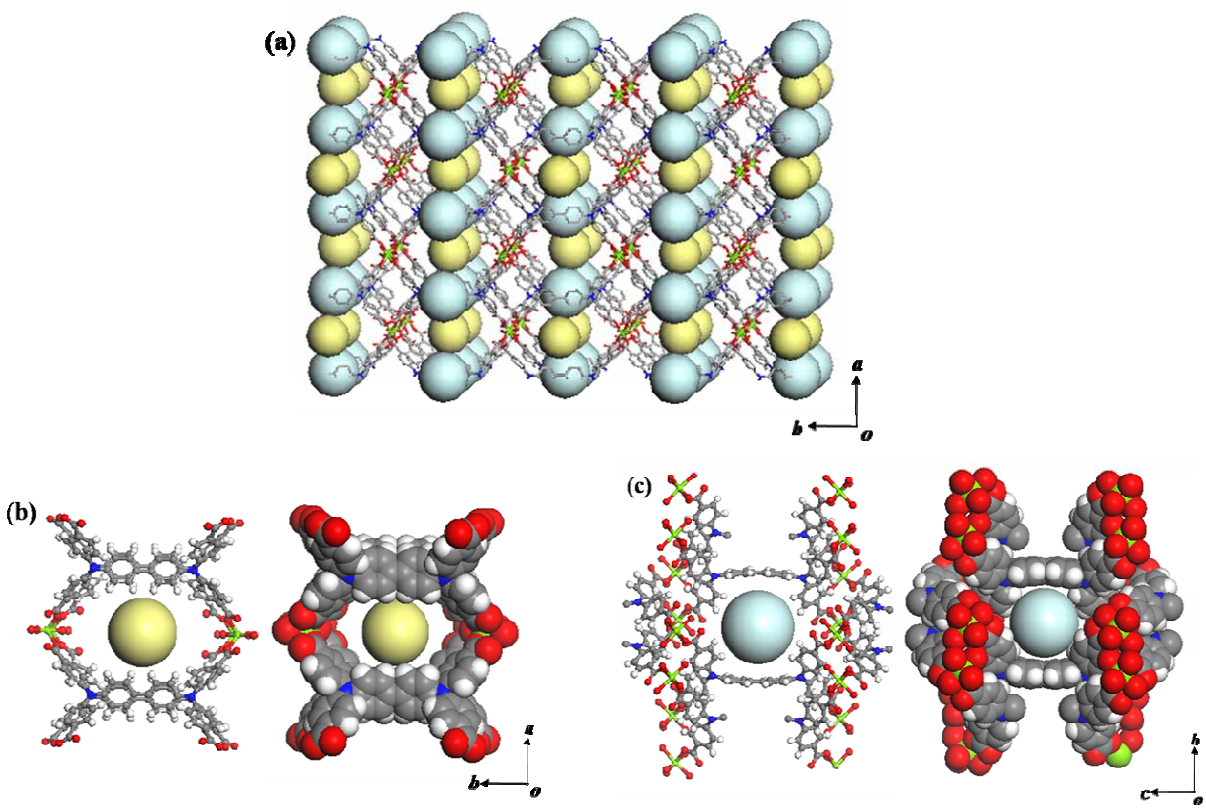

Fig S6 X-ray crystal structure of **1**, showing (a) two kinds of cavities; smaller cavities (yellow, 6.8Å) and larger cavities (blue, 11.6Å). (b) A smaller cavity. (c) A larger cavity. Hydrogen atoms and guest molecules are omitted for clarity. The open spaces represented by two kinds of spheres (yellow and blue) are estimated by inserting the probes of various sizes into the cavities until they scarcely touch the framework by using Material Studio program version 4.1 (Accelrys, San Diego, CA, 2006).

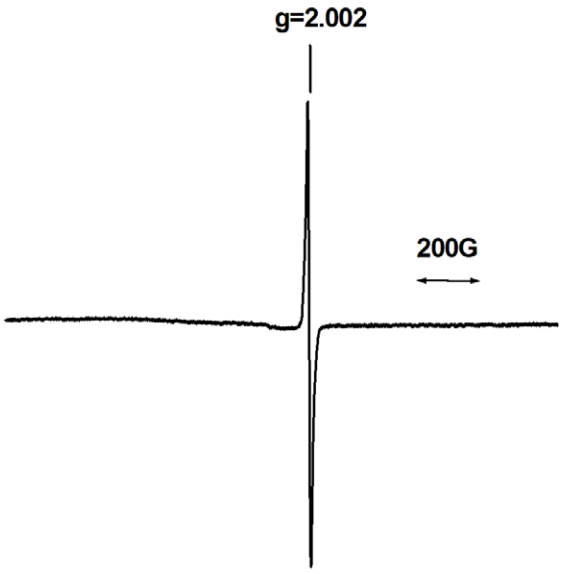

Fig. S7 EPR spectrum (powder sample) for **SNU-25** prepared by drying **1** at 180 °C under vacuum for 2 h. Measured at 173 K. g = 2.004.

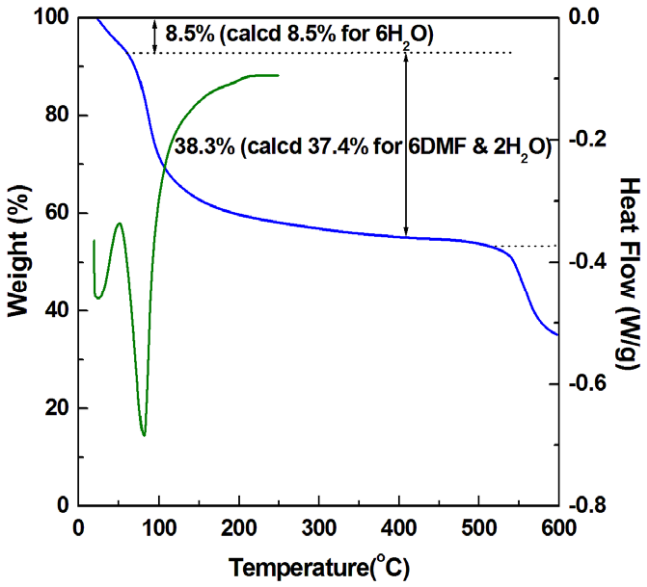

Fig. S8 TGA/DSC trace for **1.**

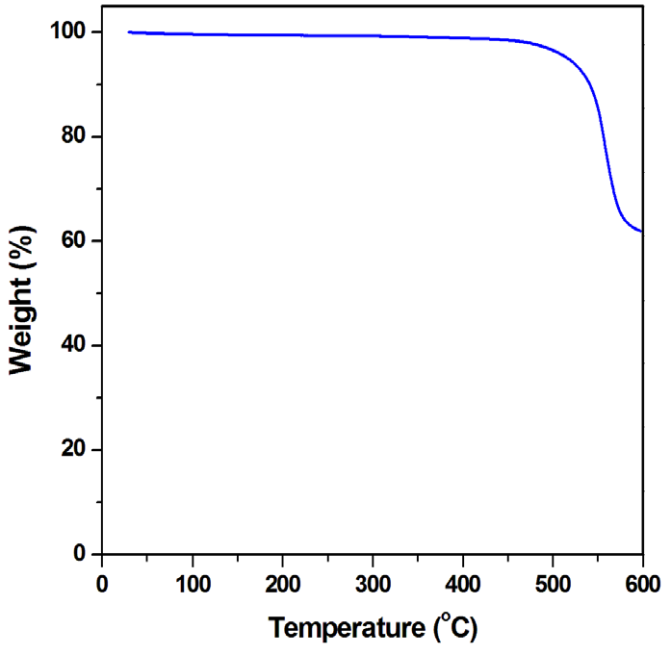

Fig. S9 TGA trace for **SNU-25** (desolvated solid of **1**).

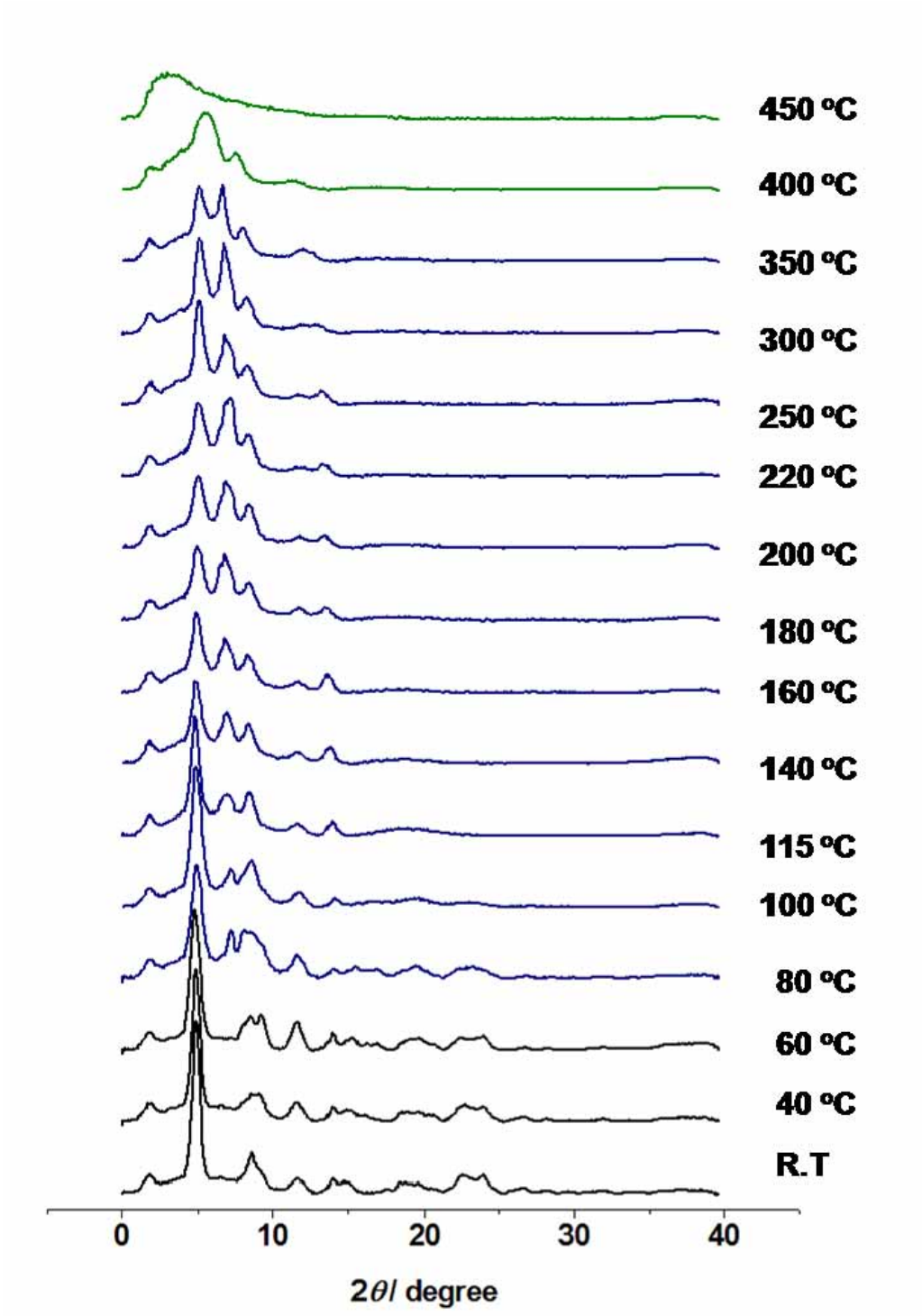

Fig.S10 The PXRD patterns of **1** measured at various temperatures (from R.T. to 450 ^{o}C).

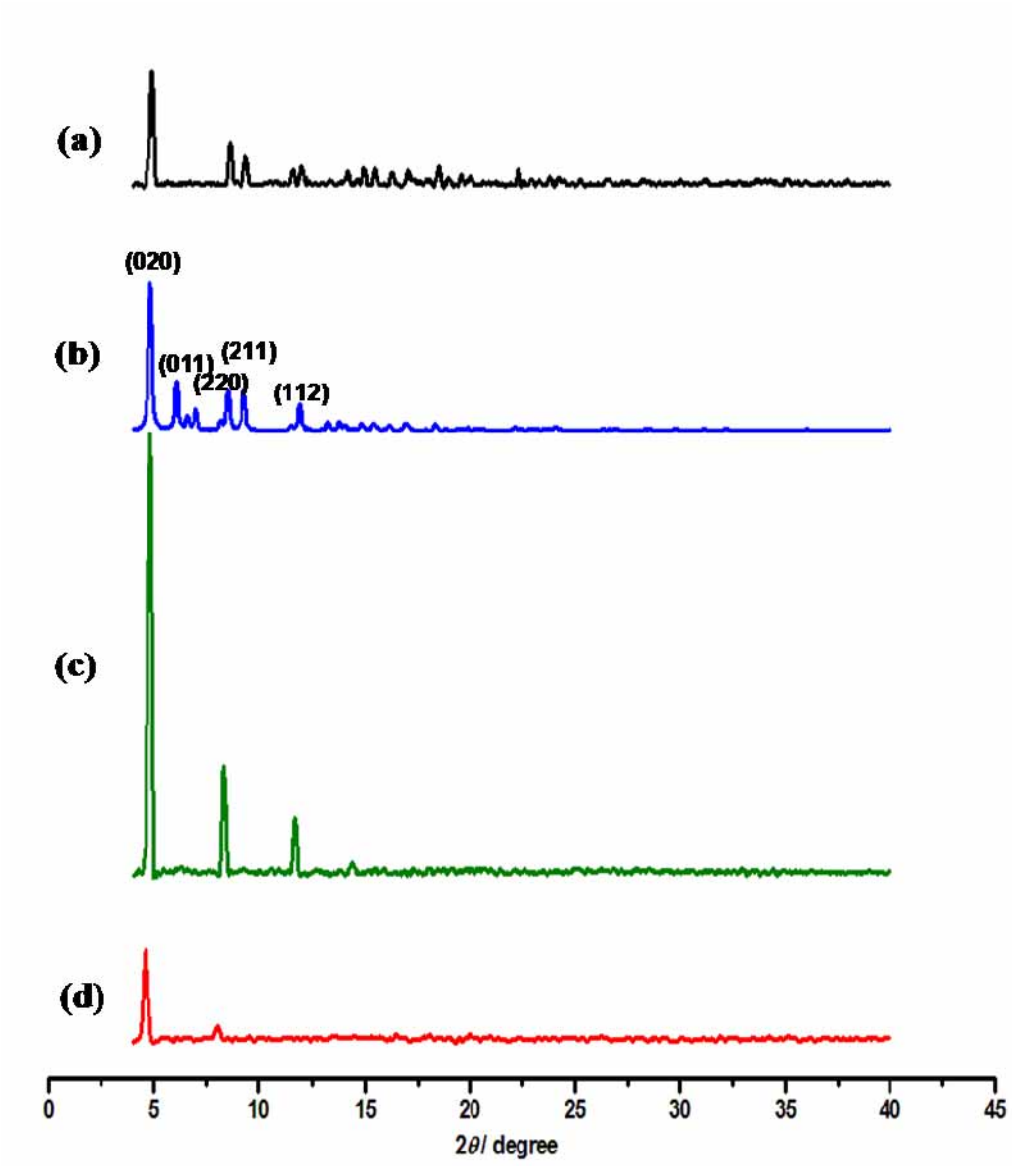

Fig. S11 The PXRD patterns of **1** and **SNU-25**. (a) **1** as prepared, (b) simulated pattern derived from the X-ray crystal data of **1**, (c) desolvated solid **SNU-25** obtained by heating **1** at 210 °C under vacuum for 4 h, (d) the solid isolated after **SNU-25** was exposed to the vapor of DMF/EtOH/H_2O (28:0.5:6.8, v/v) at 38 °C for 3 days.

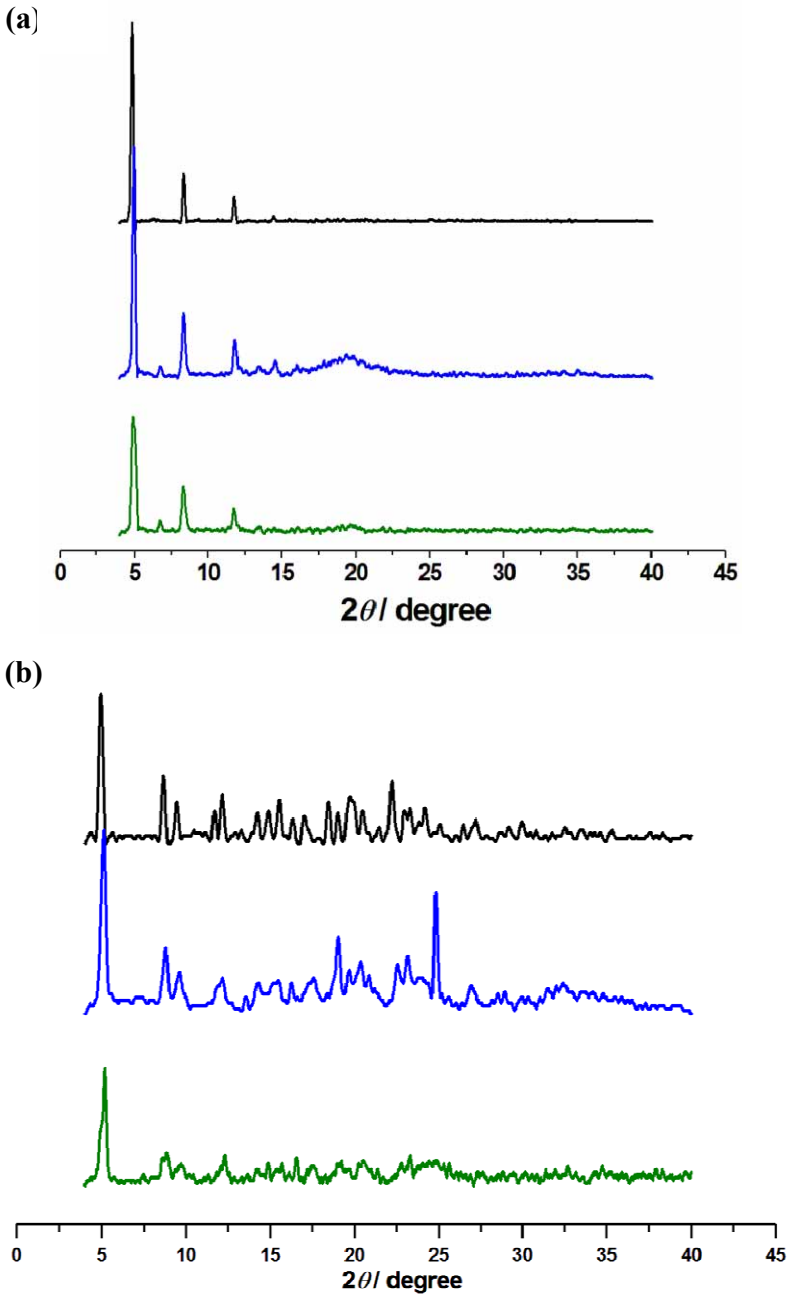

Fig. S12 The PXRD patterns of (a) **SNU-25** (black) and (b) **1** as synthesized (black), and those measured after exposure to air for 1 day (blue) and for 7 days (green).

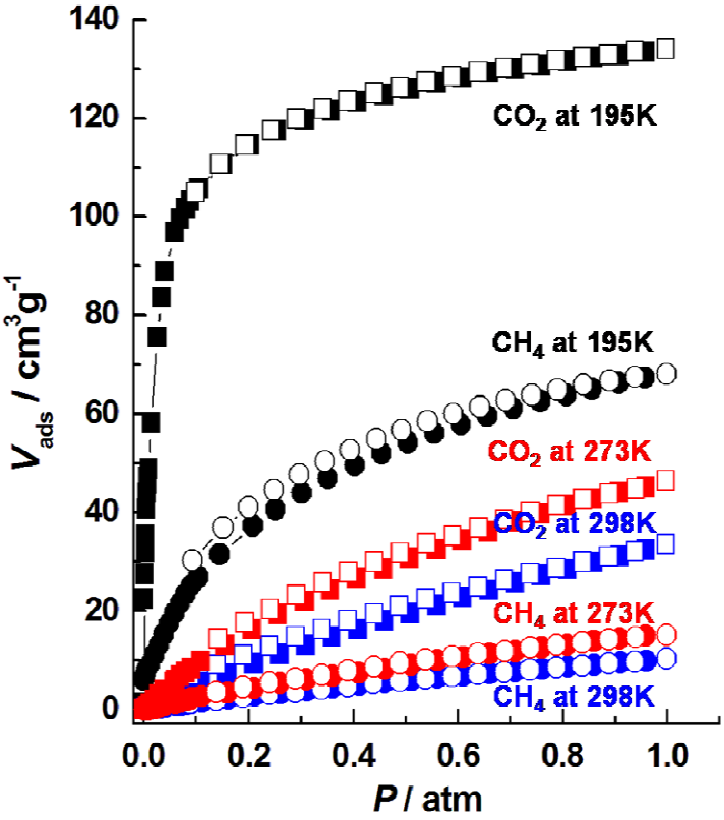

Fig. S13 The CO_2 (square) and CH_4 (circle) adsorption isotherms of **SNU-25** measured at 195 K (black), 273 K (red), and 298 K (blue).

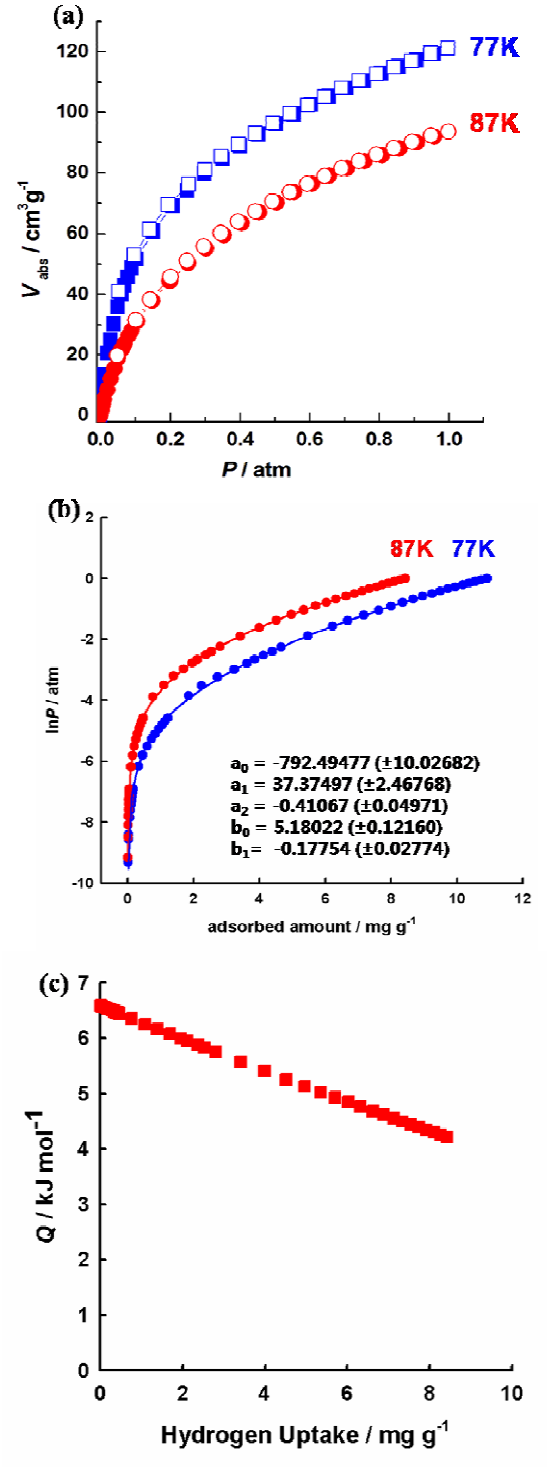

Fig. S14 (a) The H_2 adsorption isotherms of **SNU-25** measured at 77 K (blue) and 87 K (red). (b) Virial fit of the H_2 adsorption isotherms. (c) Isosteric heats of H_2 adsorption. Filled shape, adsorption; open shape, desorption.

Table S1. Crystallographic Data for 1. [squeezed data for the guest solvent molecules]

Empirical formula	$C_{40}H_{28}MgN_2O_{10}$
crystal system	Orthorhombic
space group	*Imma*
fw	720.95
a, Å	25.16(2)
b, Å	36.592(18)
c, Å	15.7272(18)
V, Å^3	14479(14)
Z	8
ρ_{calcd}, gcm^{-3}	0.661
temp, K	298(2)
λ, Å	0.71073
μ, mm^{-1}	0.056
GOF (F^2)	0.879
F(000)	2992
reflections collected	31218
independent reflections	6243 [R(int) = 0.1586]
completeness to θ_{max}, %	93.6
data/parameters/restraints	6243 / 244 / 0
θ range for data collection,	2.97 - 25.25
diffraction limits (*h*, *k*, *l*)	$-29 \le h \le 29$
	$-41 \le k \le 41$
	$-15 \le l \le 18$
Refinement method	Full-matrix least-squares on F^2
R_1, wR_2 [$I > 2\sigma(I)$]	0.1109,[a] 0.2728[b]
R_1, wR_2 (all data)	0.2456,[a] 0.3181[b]
largest peak, hole, eÅ^{-3}	1.284, -0.288

[a] $R = \Sigma||F_o| - |F_c||/\Sigma|F_o|$. [b] $wR(F^2) = [\Sigma w(F_o^2 - F_c^2)^2/\Sigma w(F_o^2)^2]^{1/2}$ where $w = 1/[\sigma^2(F_o^2) + (0.1660P)^2 + (0.00)P]$, $P = (F_o^2 + 2F_c^2)/3$

Table S2. Bond distances (Å) and angles (°) of **1.**

Mg(1)-O(1)	2.141(4)	C(5)-C(6)	1.370(7)
Mg(1)-O(4)	2.047(4)	C(6)-C(7)	1.392(7)
Mg(1)-O(100)	2.104(5)	C(8)-C(9)	1.358(7)
Mg(1)-O(101)	2.091(5)	C(8)-C(14)	1.371(7)
O(1)-C(1)	1.289(7)	C(9)-C(10)	1.406(7)
O(2)-C(1)	1.246(7)	C(10)-C(11)	1.383(7)
O(3)-C(12)	1.312(7)	C(11)-C(12)	1.508(8)
O(4)[a]-C(12)	1.196(8)	C(11)-C(13)	1.393(7)
N(1)-C(5)	1.433(6)	C(13)-C(14)	1.383(7)
N(1)-C(8)	1.435(6)	C(15)-C(16)	1.389(8)
N(1)-C(15)	1.423(7)	C(15)-C(20)	1.361(7)
C(1)-C(2)	1.478(8)	C(16)-C(17)	1.443(8)
C(2)-C(3)	1.358(7)	C(17)-C(18)	1.348(8)
C(2)-C(7)	1.407(7)	C(18)-C(18)[b]	1.472(11)
C(3)-C(4)	1.385(6)	C(18)-C(19)	1.396(7)
C(4)-C(5)	1.363(6)	C(19)-C(20)	1.376(7)
O(100)-Mg(1)-O(101)	174.3(2)	O(4)[a]-C(12)-C(11)	123.7(7)
O(100)-Mg(1)-O(1)	90.19(16)	C(1)-C(2)-C(3)	126.0(6)
O(100)-Mg(1)-O(4)	92.08(17)	C(1)-C(2)-C(7)	116.4(6)
O(101)-Mg(1)-O(1)	85.71(15)	C(2)-C(3)-C(4)	121.9(5)
O(101)-Mg(1)-O(4)	91.81(17)	C(2)-C(7)-C(6)	120.7(5)
O(1)[c]-Mg(1)-O(4)	176.25(18)	C(3)-C(2)-C(7)	117.5(5)
O(1)-Mg(1)-O(4)	88.89(15)	C(3)-C(4)-C(5)	120.3(5)
O(1)-Mg(1)-O(1)[c]	88.1(2)	C(4)-C(5)-C(6)	119.8(5)
O(4)-Mg(1)-O(4)[c]	94.0(2)	C(5)-C(6)-C(7)	119.8(5)
Mg(1)-O(1)-C(1)	127.9(4)	C(8)-C(9)-C(10)	119.2(5)
Mg(1)-O(4)-C(12)[d]	134.2(4)	C(8)-C(14)-C(13)	119.1(6)
C(5)-N(1)-C(8)	120.1(5)	C(9)-C(8)-C(14)	121.4(5)
C(5)-N(1)-C(15)	117.2(4)	C(9)-C(10)-C(11)	120.4(5)
C(8)-N(1)-C(15)	122.4(4)	C(10)-C(11)-C(12)	124.8(6)
N(1)-C(5)-C(4)	120.7(5)	C(10)-C(11)-C(13)	118.1(5)
N(1)-C(5)-C(6)	119.5(5)	C(11)-C(13)-C(14)	121.3(5)
N(1)-C(8)-C(9)	117.3(6)	C(12)-C(11)-C(13)	117.1(6)
N(1)-C(8)-C(14)	121.0(6)	C(15)-C(16)-C(17)	118.2(6)
N(1)-C(15)-C(16)	118.8(6)	C(15)-C(20)-C(19)	121.2(6)
N(1)-C(15)-C(20)	122.2(5)	C(16)-C(15)-C(20)	118.8(6)
O(1)-C(1)-O(2)	124.0(5)	C(16)-C(17)-C(18)	122.9(6)
O(3)-C(12)-O(4)[a]	126.5(6)	C(17)-C(18)-C(18)[b]	121.9(4)

O(1)-C(1)-C(2)	117.3(7)	C(17)-C(18)-C(19)	115.8(5)
O(2)-C(1)-C(2)	118.7(6)	C(18)[b]-C(18)-C(19)	122.2(4)
O(3)-C(12)-C(11)	109.7(7)	C(18)-C(19)-C(20)	122.6(6)

Symmetry transformations used to generate equivalent atoms: *a*, -x+1/2, -y, z+1/2; *b*, x, -y+1/2, z; *c*, -x+1, y, z; *d*, -x+1/2, -y, z-1/2.

Table S3. The H_2 adsorption and desorption data at 77 K and 87 K for **SNU-25**.

P (atm)	H_2 at 77 K	P (atm)	H_2 at 77 K	P (atm)	H_2 at 87 K	P (atm)	H_2 at 87 K
9.06E-05	0.1082	0.999	121.2403	1.06E-04	0.0713	0.999	93.6133
1.91E-04	0.2367	0.949	120.0483	2.06E-04	0.12	0.949	92.3462
2.91E-04	0.3659	0.896	118.0743	3.05E-04	0.172	0.895	90.3582
3.93E-04	0.5075	0.844	115.9309	4.08E-04	0.2264	0.842	88.2122
4.94E-04	0.6403	0.793	113.7081	5.06E-04	0.2786	0.792	86.179
5.96E-04	0.773	0.743	111.4473	6.09E-04	0.334	0.742	83.9455
6.98E-04	0.9043	0.693	109.0644	7.08E-04	0.3873	0.692	81.6127
7.99E-04	1.0284	0.643	106.5421	8.05E-04	0.4432	0.643	78.9751
8.99E-04	1.1481	0.593	103.8894	9.12E-04	0.4987	0.593	76.407
9.96E-04	1.2347	0.544	101.0003	1.00E-03	0.5482	0.543	73.5857
2.01E-03	2.551	0.495	97.9086	2.04E-03	1.0976	0.493	70.579
0.003	3.9378	0.446	94.5301	0.003	1.6296	0.444	67.3057
0.00408	5.2729	0.395	93.0935	0.00407	2.1887	0.394	63.9912
0.00509	6.4234	0.347	89.2577	0.00503	2.7022	0.345	60.2179
0.00619	7.5922	0.301	84.8662	0.00608	3.2468	0.296	55.8665
0.00709	8.5213	0.243	78.4527	0.00719	3.8202	0.248	51.1288
0.0082	9.5791	0.201	73.0413	0.00809	4.2644	0.201	45.7901
0.00912	10.4131	0.147	64.9286	0.00907	4.7409	0.143	38.3153
0.0101	11.3026	0.0972	55.3239	0.0102	5.2635	0.102	31.5795
0.0194	14.5865	0.0459	40.7343	0.0207	8.5478	0.048	19.8715
0.0312	21.3235			0.0303	12.1035		
0.0399	25.8707			0.0409	15.453		
0.0503	30.6042			0.0507	18.8851		
0.0594	34.1888			0.0616	21.8025		
0.0691	38.4322			0.0698	23.5537		
0.0798	41.5377			0.0815	26.3753		
0.09	44.3039			0.0899	28.1361		
0.104	47.4974			0.106	31.146		
0.152	57.2603			0.149	37.9138		
0.205	66.9252			0.199	44.3818		
0.247	72.7618			0.251	50.2335		
0.298	78.5039			0.303	55.1686		
0.349	83.7486			0.354	59.4532		
0.401	88.2688			0.405	63.421		
0.452	92.4436			0.455	66.9977		
0.503	96.1678			0.506	70.212		
0.554	99.6371			0.556	73.6148		
0.605	102.7208			0.607	76.4296		
0.656	105.3922			0.657	79.1558		
0.706	108.3023			0.708	81.513		
0.756	110.7946			0.758	83.9135		
0.807	113.206			0.809	85.9377		
0.857	115.4987			0.858	88.0457		
0.907	117.5543			0.908	90.1283		
0.958	119.4652			0.959	91.9535		
0.999	121.2403			0.999	93.6133		

Table S4. The N_2 adsorption and desorption data at 77 K for **SNU-25**.

P (atm)	N_2 at 77 K	P (atm)	N_2 at 77 K
8.71E-05	1.086	0.937	38.9584
2.08E-04	1.7189	0.892	36.6019
3.14E-04	2.8788	0.841	34.8251
4.11E-04	3.6796	0.791	33.1156
5.07E-04	3.7055	0.741	31.3742
5.08E-04	3.8373	0.691	29.7812
7.08E-04	5.4132	0.641	28.1233
7.68E-04	5.5641	0.591	26.4435
9.28E-04	5.5814	0.541	24.9174
1.00E-03	5.5933	0.491	23.276
0.00218	5.749	0.441	21.763
0.00303	5.8819	0.391	20.3122
0.00407	6.0227	0.341	18.8132
0.00521	6.1688	0.291	17.353
0.00629	6.3126	0.241	15.9185
0.00734	6.4541	0.191	14.4935
0.00839	6.5943	0.141	13.1755
0.00944	6.7337	0.0909	11.7946
0.0133	6.9029	0.04	10.3107
0.0224	6.9647		
0.0322	7.2675		
0.0422	7.5114		
0.0522	7.7404		
0.0621	7.9789		
0.0724	8.1373		
0.0821	8.3734		
0.0921	8.5898		
0.11	8.95		
0.159	9.7667		
0.209	10.709		
0.259	11.6987		
0.309	12.658		
0.36	13.5489		
0.409	14.618		
0.459	15.7703		
0.509	16.7827		
0.559	17.8931		
0.609	19.0727		
0.659	20.2106		
0.709	21.3659		
0.754	30.8577		
0.807	33.096		
0.858	35.0644		
0.908	37.0742		
0.937	38.9584		

Table S5. The O_2 adsorption and desorption data at 77 K for **SNU-25**.

P (atm)	O_2 at 77 K	P (atm)	O_2 at 77 K
8.76E-05	22.3006	0.207	233.2921
1.89E-04	26.1697	0.151	229.7483
2.89E-04	63.6338	0.0967	225.2538
3.91E-04	66.0138	0.0486	218.1869
4.95E-04	67.6107		
5.44E-04	68.1793		
6.96E-04	70.3876		
7.99E-04	71.2321		
8.17E-04	71.3631		
0.00101	73.2545		
0.00201	152.8076		
0.00309	157.7362		
0.00401	160.7079		
0.00521	163.5841		
0.00618	165.3641		
0.00717	186.7907		
0.01	198.7448		
0.0147	208.1176		
0.0142	206.3631		
0.0213	209.921		
0.0309	213.8952		
0.0416	216.7773		
0.0504	218.5445		
0.0599	220.4879		
0.0704	222.1107		
0.0808	223.4952		
0.091	224.7086		
0.109	226.4524		
0.156	230.1245		
0.207	233.2921		

Table S6. The CO_2 adsorption and desorption data at 195 K, 273 K, and 298 K for **SNU-25**.

P (atm)	CO_2 at 195 K	P (atm)	CO_2 at 195 K	P (atm)	CO_2 at 273 K	P (atm)	CO_2 at 273 K	P (atm)	CO_2 at 298 K	P (atm)	CO_2 at 298 K
5.16E-04	0.008	0.999	134.0512	0.00102	0.0095	0.999	46.3758	0.00205	0.0187	0.999	33.4433
6.22E-04	0.0109	0.94	133.4808	0.00202	0.0251	0.941	44.8296	0.00304	0.0599	0.939	32.0591
7.22E-04	0.0127	0.889	132.8244	0.00304	0.042	0.89	43.8036	0.00406	0.1036	0.889	30.9782
8.23E-04	0.0145	0.839	132.2849	0.00407	0.0639	0.84	42.5979	0.00508	0.1474	0.839	29.8655
9.19E-04	0.0194	0.789	131.6278	0.005	0.1028	0.79	41.4517	0.00605	0.1901	0.789	28.6871
0.00153	1.1752	0.739	130.9171	0.00634	0.2192	0.74	40.0109	0.00703	0.2341	0.739	27.465
0.00225	22.4078	0.69	130.154	0.00705	0.2938	0.69	38.5161	0.00939	0.3478	0.689	26.2014
0.00304	27.3535	0.64	129.301	0.00927	0.4835	0.641	36.9052	0.0103	0.3873	0.639	24.898
0.00407	31.8408	0.59	128.3719	0.0101	0.5868	0.591	35.2023	0.0131	0.5756	0.589	23.6085
0.00503	35.3592	0.54	127.3525	0.0127	0.9025	0.541	33.5608	0.0227	1.0708	0.539	22.3033
0.00679	40.8486	0.491	126.2062	0.0208	1.7081	0.491	31.7782	0.0324	1.561	0.489	20.9037
0.00801	43.981	0.441	124.9348	0.0305	2.8048	0.441	29.8834	0.0422	2.063	0.44	19.4512
0.00878	45.7914	0.391	123.4314	0.0405	3.8228	0.392	27.86	0.0609	2.9509	0.39	17.9337
0.00937	47.0496	0.342	121.7912	0.0587	5.4208	0.342	25.5889	0.0628	3.0769	0.34	16.3398
0.0102	48.8571	0.292	119.794	0.0621	5.8557	0.292	23.0846	0.0725	3.5395	0.29	14.6626
0.0151	58.0909	0.243	117.4366	0.0714	6.6967	0.242	20.3768	0.0825	4.0129	0.24	12.8578
0.0273	75.5166	0.194	114.4974	0.0814	7.5262	0.193	17.5068	0.0925	4.4694	0.19	11.0028
0.0352	83.573	0.146	110.5262	0.0916	8.2611	0.143	14.2301	0.111	5.2875	0.14	9.0223
0.0421	88.8153	0.1	104.7746	0.11	9.6915			0.16	7.3145	0.0902	6.8973
0.0604	96.6696			0.156	13.0469			0.21	9.2667		
0.0712	99.4927			0.206	16.2348			0.26	11.1348		
0.0817	101.5668			0.257	19.2082			0.31	12.9293		
0.09	103.0725			0.307	21.829			0.36	14.6803		
0.107	105.6312			0.357	24.2664			0.41	16.3342		
0.152	110.56			0.407	26.5779			0.46	17.9791		
0.204	114.4852			0.458	28.6706			0.51	19.5878		
0.255	117.3932			0.508	30.6662			0.56	21.1196		
0.307	119.6777			0.559	32.4758			0.61	22.6324		
0.358	121.5577			0.609	34.3577			0.66	24.1484		
0.408	123.1766			0.659	36.1026			0.71	25.6583		
0.459	124.5943			0.708	38.0249			0.76	27.195		
0.509	125.9023			0.758	39.7117			0.81	28.5263		
0.559	127.0878			0.809	41.3499			0.86	29.7863		
0.609	128.1429			0.859	42.7364			0.91	31.0836		
0.66	129.1104			0.909	44.0452			0.96	32.3767		
0.71	129.9517			0.96	45.1488			0.999	33.4433		
0.76	130.7719			0.999	46.3758						
0.81	131.5629										
0.86	132.2473										
0.91	132.7709										
0.96	133.4572										
0.999	134.0512										

Table S7. The CH_4 adsorption and desorption data at 195 K, 273 K, and 298 K for **SNU-25**.

P (atm)	CH_4 at 195 K	P (atm)	CH_4 at 195	P (atm)	CH_4 at 273 K	P (atm)	CH_4 at 273 K	P (atm)	CH_4 at 298 K	P (atm)	CH_4 at 298 K
6.50E-04	5.9868	0.999	68.0912	0.00103	-1.00E-02	0.999	15.0902	0.00406	0.0054	0.999	10.2249
6.57E-04	5.9699	0.939	67.4486	0.00207	4.00E-04	0.94	14.6779	0.00511	0.027	0.939	9.6547
7.34E-04	5.9608	0.89	66.6964	0.00308	0.003	0.889	14.1158	0.00615	0.0439	0.888	9.2595
8.26E-04	5.9544	0.84	65.8896	0.00412	0.0057	0.838	13.5296	0.00709	0.0543	0.838	8.8888
9.29E-04	5.9507	0.791	64.8686	0.0051	0.0086	0.789	13.0561	0.00954	0.1074	0.788	8.5081
0.00104	5.9488	0.741	63.8317	0.0061	0.0113	0.739	12.5698	0.0102	0.1014	0.738	8.06
0.002	6.3241	0.691	62.7842	0.0071	0.0189	0.69	11.8009	0.0133	0.1589	0.688	7.5747
0.00301	6.7302	0.642	61.4356	0.00923	0.0414	0.639	11.2736	0.0231	0.2921	0.638	7.1116
0.00403	7.1105	0.592	60.0395	0.01	0.0568	0.589	10.6497	0.0329	0.4449	0.589	6.6609
0.005	7.4164	0.542	58.5319	0.013	0.1481	0.539	10.067	0.043	0.5776	0.538	6.1938
0.00609	7.7629	0.493	56.8127	0.0227	0.3198	0.489	9.388	0.0617	0.8432	0.489	5.7029
0.00715	8.053	0.444	54.9031	0.0322	0.6237	0.44	8.6232	0.063	0.825	0.439	5.1928
0.00868	8.5284	0.394	52.7688	0.0423	0.8057	0.39	7.8782	0.0728	0.9621	0.389	4.6988
0.00959	8.7725	0.345	50.4384	0.0609	1.1998	0.34	7.0161	0.0828	1.0983	0.339	4.1679
0.0121	9.0782	0.297	47.753	0.0626	1.3363	0.29	6.207	0.0928	1.2324	0.289	3.6047
0.021	11.0408	0.248	44.6634	0.0723	1.5593	0.24	5.4699	0.112	1.5151	0.239	3.0401
0.03	13.136	0.2	41.0847	0.0822	1.8439	0.19	4.5642	0.161	2.1472	0.189	2.4661
0.0396	15.3478	0.152	36.9013	0.0924	2.0921	0.14	3.5766	0.211	2.7198	0.139	1.8626
0.0496	17.5057	0.0937	30.219	0.111	2.5708			0.261	3.3	0.0891	1.1829
0.06	19.6032			0.16	3.3642			0.311	3.854		
0.0705	21.5436			0.21	4.3409			0.361	4.4256		
0.0809	23.3509			0.26	5.1822			0.411	4.921		
0.0912	25.0347			0.31	6.032			0.461	5.414		
0.106	26.8894			0.36	6.7582			0.511	5.9233		
0.146	31.4613			0.41	7.5178			0.561	6.4088		
0.208	37.3392			0.46	8.2596			0.611	6.8698		
0.251	40.621			0.51	9.0812			0.661	7.3149		
0.301	43.9338			0.56	9.9122			0.711	7.7917		
0.352	46.9748			0.61	10.6052			0.761	8.2842		
0.403	49.6644			0.66	11.3785			0.811	8.6951		
0.454	52.0299			0.71	11.884			0.861	9.0524		
0.505	54.1997			0.76	12.5681			0.911	9.4346		
0.555	56.1426			0.81	13.1497			0.961	9.8071		
0.606	57.9514			0.86	13.8248			0.999	10.2249		
0.656	59.6475			0.91	14.3323						
0.706	61.1569			0.96	14.9128						
0.757	62.5727			0.999	15.0902						
0.808	63.8678										
0.857	65.0392										
0.908	66.132										
0.958	67.1777										
0.999	68.0912										