

Supporting Information

**Hierarchical N-doped Carbon as CO₂ Adsorbent with High CO₂ Selectivity
from Rationally Designed Polypyrrole Precursor**

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Methods

Synthesis of 4-aminobutyric acid. 4-(pyrrol-1-yl)butanoic acid is synthesized according to previous work by Gracia *et al.*¹ In a typical synthesis, 4-aminobutyric acid (10.0 g, 97 mmol), H₂O (144 ml), sodium acetate (NaOAc, 8.0 g, 97.5 mmol), acetic acid (AcOH, 48 ml) and 1,2-dichloroethane (144 ml) were heated together at 90 °C in N₂. 2,5-dimethoxytetrahydrofuran (12.6 ml, 97.2 mmol) was added to the mixture, which was vigorously stirred at 90 °C for 16 h. The mixture was cooled, and the organic layer was removed. The aqueous layer was extracted with dichloromethane three times (CH₂Cl₂, 3 x 20 ml). The combined organic extracts were washed with water (2 x 200 ml), dried with anhydrous magnesium sulphate (MgSO₄), filtered and the solvent was removed under a reduced pressure using a rotor-vap. The crude product was dissolved in CH₂Cl₂ (20 ml) and extracted repeatedly with saturated aqueous NaHCO₃ solution. The combined basic extracts were made acidic with aqueous HCl solution and again extracted with CH₂Cl₂ (3 x 20 ml). The organic phase was dried (MgSO₄) and the solvent was removed under a reduced pressure to give 4-(pyrrol-1-yl)butanoic acid in 64% yield. ¹H NMR (400 MHz, CDCl₃, δ , ppm) 6.64 (t, J = 2.1 Hz, ArH, 2H), 6.15 (t, J = 2.1 Hz, ArH, 2H), 3.95 (t, J = 6.8 Hz, NCH₂-, 2H), 2.32 (d, J = 7.3 Hz, -CH₂COOH, 2H), 2.08 (q, J = 6.9 Hz, -CH₂CH₂CH₂-, 2H). The ¹H NMR spectrum is shown in Fig. S20.

Dynamic Column Breakthrough Experiments. The dynamic CO₂ capacity was evaluated using a custom-built dynamic gas breakthrough system. A mixed gas of 90 vol% N₂ and 10 vol% CO₂ was obtained by regulating the flow rates of each gas with mass flow controllers. The total flow rate of the mixed gas was kept at 30 cm³ min⁻¹. The packed-bed column was a vertical stainless steel tube with an inner diameter of 0.45 cm. The sorbent sample was held on a porous stainless steel filter, which was cut to fit tightly inside the stainless steel tube. The typical sample size is within the range of 15-30 mg. In specific, the column parameters for the SU-MAC-500 tests are as follows: bed length, L = 0.4 cm; bed voidage, ε = 0.4; superficial gas velocity at inlet, u = 3.145 cm s⁻¹. The column is heated using controlled Thermolyne heating tape. The sorbent sample was heat treated at 200 °C in a helium stream for at least 2

hours prior to measurements. Regeneration was performed by purging N₂ at 25 °C for 30 min between subsequent cycles. The effluent gas was analyzed using an Extrel Max300-LG mass spectrometer. In a test under humid conditions, the N₂ stream was bubbled through a stainless steel column with water before mixing with CO₂. The bubbling process was allowed to equilibrate for at least 20 minutes prior to analysis. The water concentration in the gas stream is approximately 3 vol% assuming water saturation vapor pressure (100% humidity) at 298 K.

Inductively coupled plasma mass spectrometry (ICP-MS). Potassium (K) ICP-MS was performed and the concentration of K in the samples of SU-MAC-500, 600 and 800 were found to be 1.04%, 2.12% and 0.56%, respectively.

Density Measurements. Helium buoyancy experiments were carried out using a Rubotherm magnetic suspension balance. The sample was heated at 200 °C under vacuum for 8 hours prior to the measurements. The experiments were performed at 200 °C to minimize helium adsorption. The true mass and skeletal volume of the sample were obtained by fitting measured weight (*i.e.*, apparent weight) as a function of the helium density. The results for empty basket and basket with sample are shown in Figure S2. The skeletal density of SU-MAC-500 was found to be 1.07 g cm⁻³. The bulk density of the sample can be estimated using the following equation:

$$\rho_{bulk} = \frac{m}{V_s + V_p}$$

where m and V_s are the mass and skeletal volume, respectively, measured using helium buoyancy experiments; and V_p is the total pore volume measured using nitrogen adsorption at 77 K and a relative pressure of 0.999, which is 0.47 cm³ g⁻¹. The resulting bulk density is 0.71 g cm⁻³.

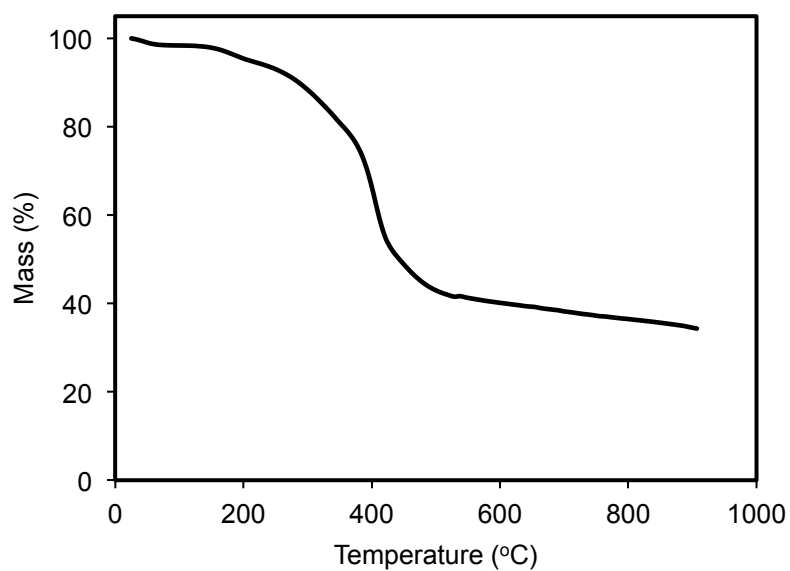


Figure S1. Thermogravimetric (TG) curve of as-synthesized polymer with a 5 °C min⁻¹ rate up to 900 °C.

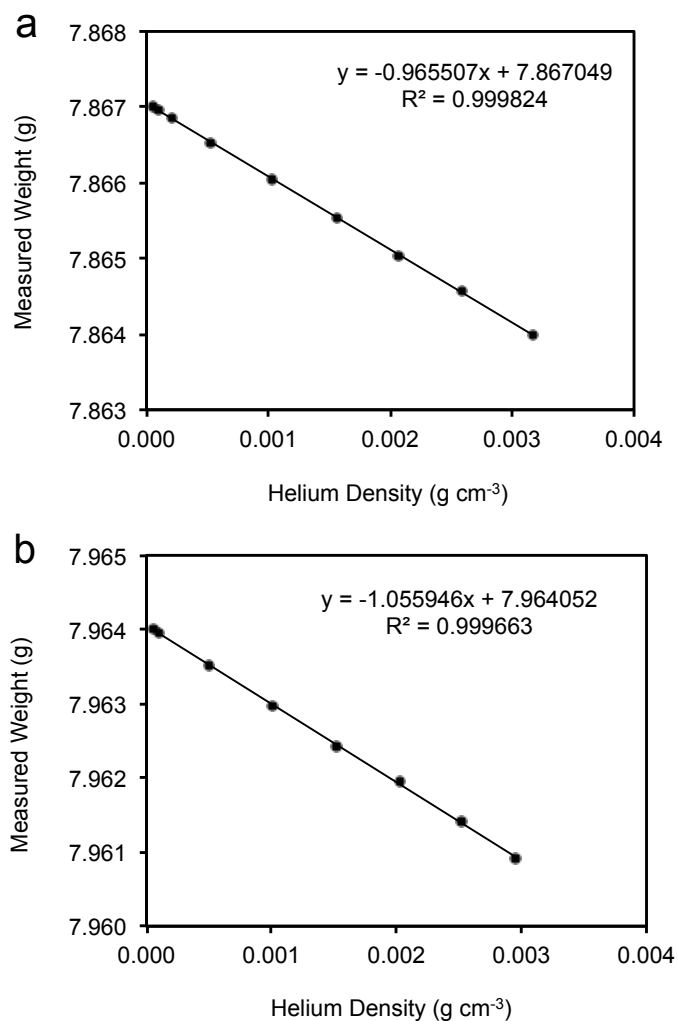


Figure S2. Helium buoyancy data at 200 °C for (a) empty basket and (b) basket with the SU-MAC-500 sample. The true mass and volume can be determined from the y-intercept and slope of the linear fit, respectively. The basket has a mass of 7.867049 g and a volume of 0.965507 cm³, while the sample has a mass of 0.097003 g and a skeletal volume of 0.090439 cm³.

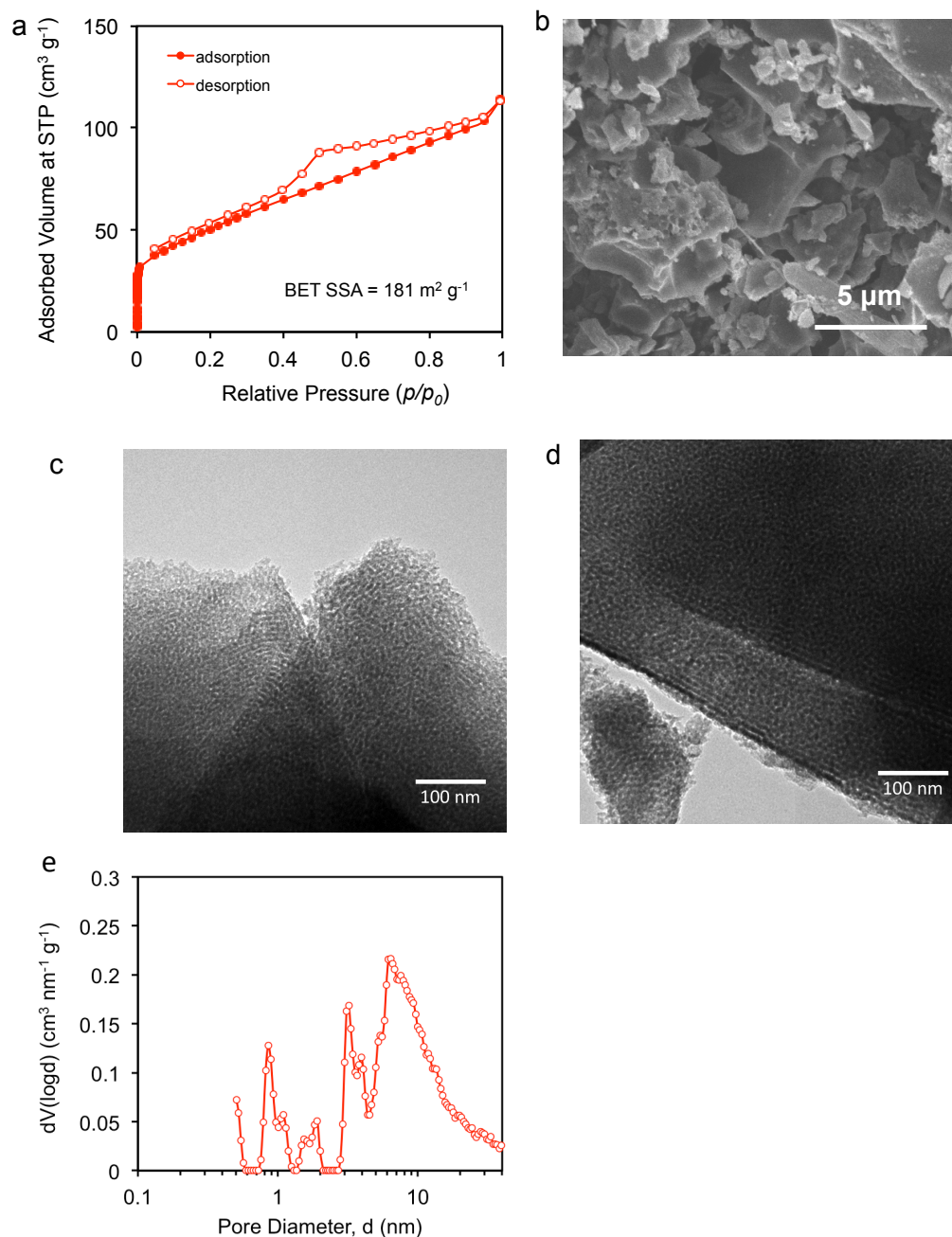


Figure S3. Characterizations of the carbon synthesized using a similar method with an exception that the soft template is changed to Pluronic F-127, which has a longer hydrophobic chain: (a) N_2 adsorption/desorption isotherms at 77 K, BET specific surface area (SSA) = $181 \text{ m}^2 \text{g}^{-1}$; (b) Scanning electron microscopic (SEM) image; (c) and (d) transmission electron microscopic (TEM) images, which suggest a disordered worm-like mesopore morphology; (e) pore size distribution based upon the N_2 isotherm with a major mesopore peak at 6.2 nm

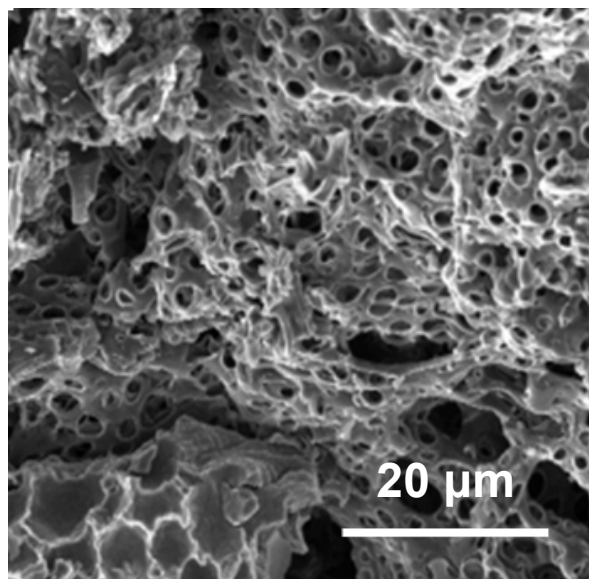


Figure S4. Scanning electron microscopy (SEM) of the SU-MC material synthesized at pH = 1. Foam-like morphology is obtained under this condition.

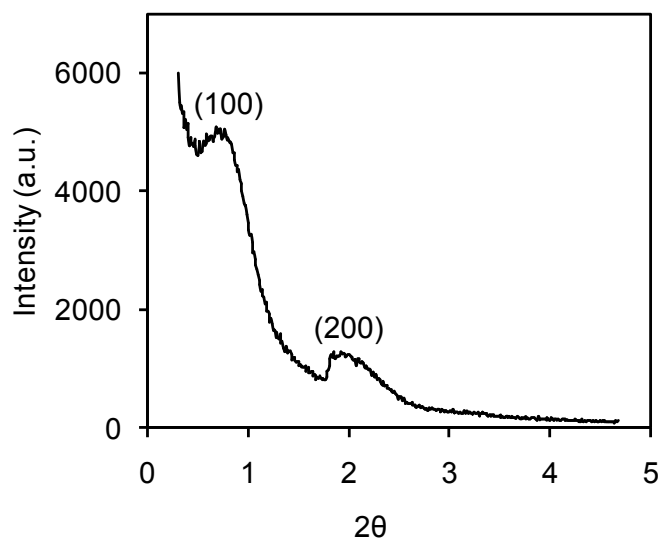


Figure S5. Small-angle X-ray diffraction (XRD) pattern of SU-MC. The existence of the (100) and (200) peaks are clearly observed, which further supports the presence of 2-dimensional hexagonal arrays. The corresponding d_{100} -spacing based on this data is 8.4 nm.

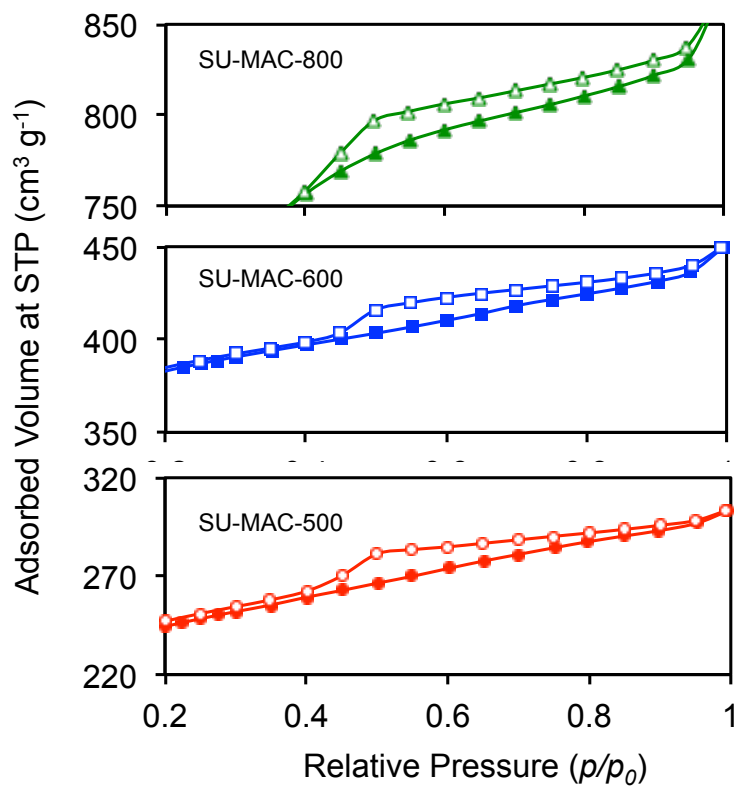


Figure S6. Zoom-in views of the N₂ adsorption (solid symbols) and desorption (open symbols) for the SU-MAC samples showing the hysteresis, which is indicative of mesoporosity. These hysteretic loops can be classified as type H4 hysteresis according to the IUPAC classifications². The forced closure of the hysteresis loop occurs for N₂ at 77 K in the relative pressure range of 0.40 – 0.45 is due to the tensile strength effect³.

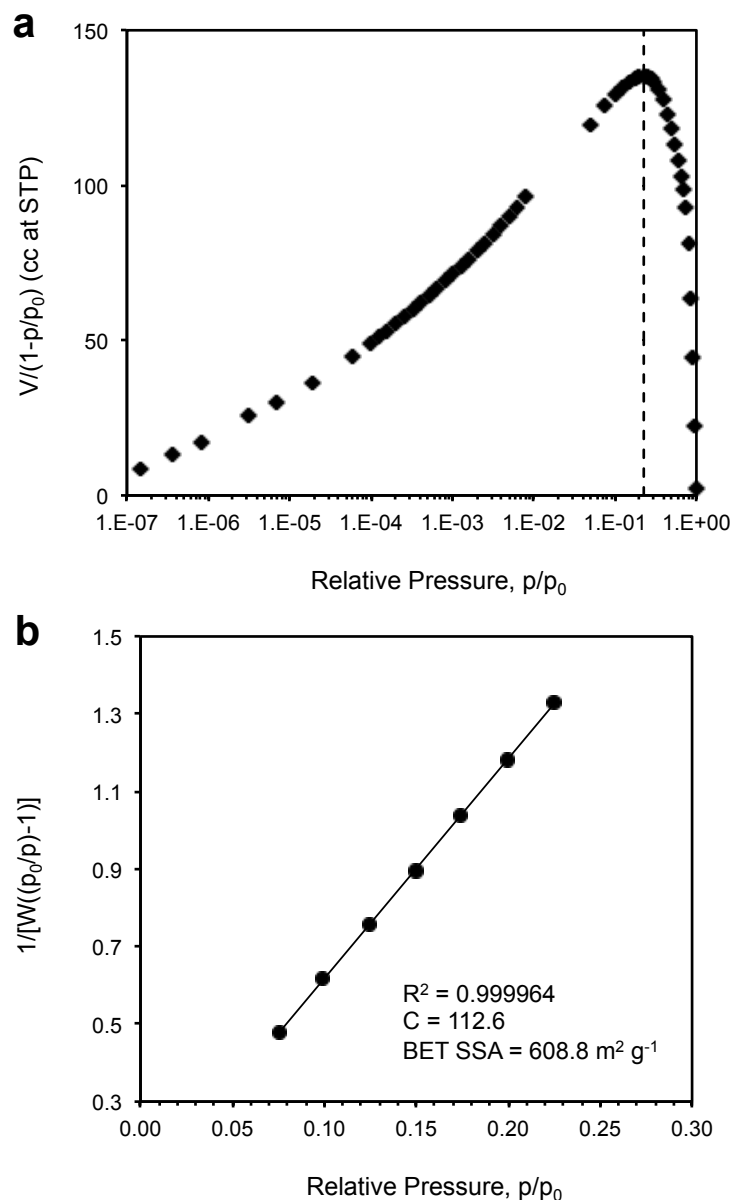


Figure S7. BET calculations and fittings for SU-MC: (a) $V/(1-p/p_0)$ vs. p/p_0 with the points in the range below $p/p_0 = 0.225$ satisfy the first consistency criterion for applying the BET theory; and (b) linear fitting of the BET equation, which satisfies the second consistency criterion for applying the BET theory⁴.

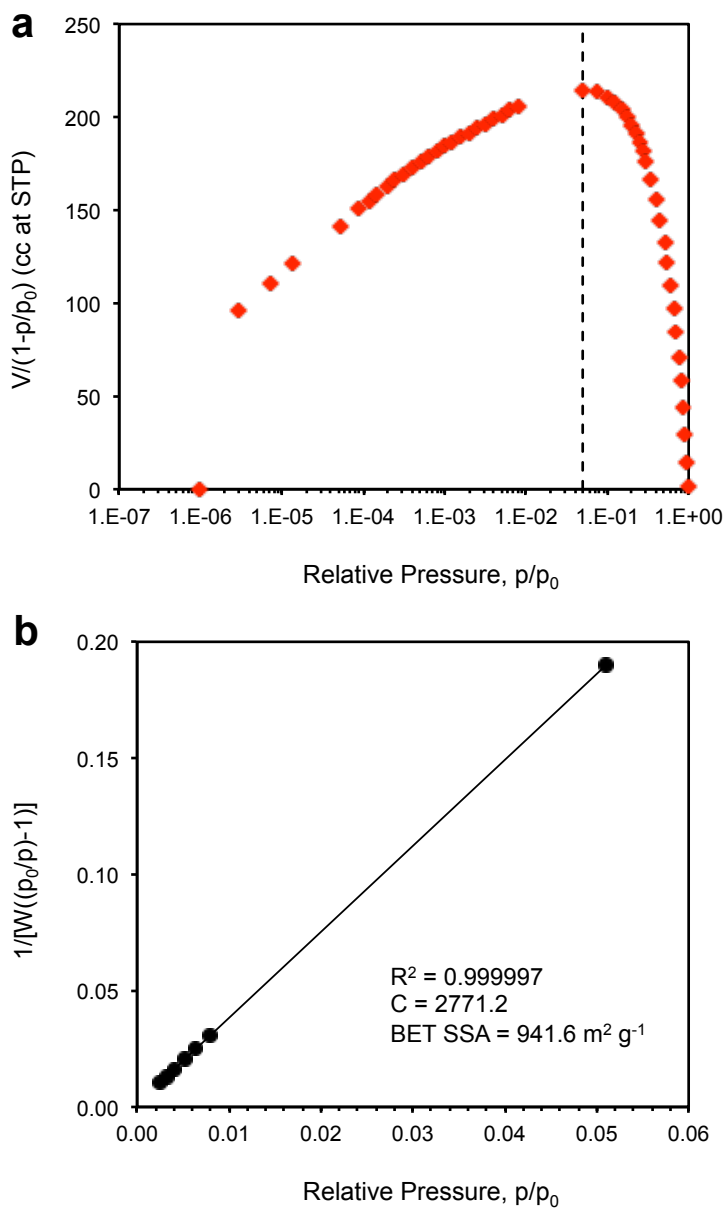


Figure S8. BET calculations and fittings for SU-MAC-500: (a) $V/(1-p/p_0)$ vs. p/p_0 with the points in the range below $p/p_0 = 0.051$ satisfy the first consistency criterion for applying the BET theory; and (b) linear fitting of the BET equation, which satisfies the second consistency criterion for applying the BET theory⁴.

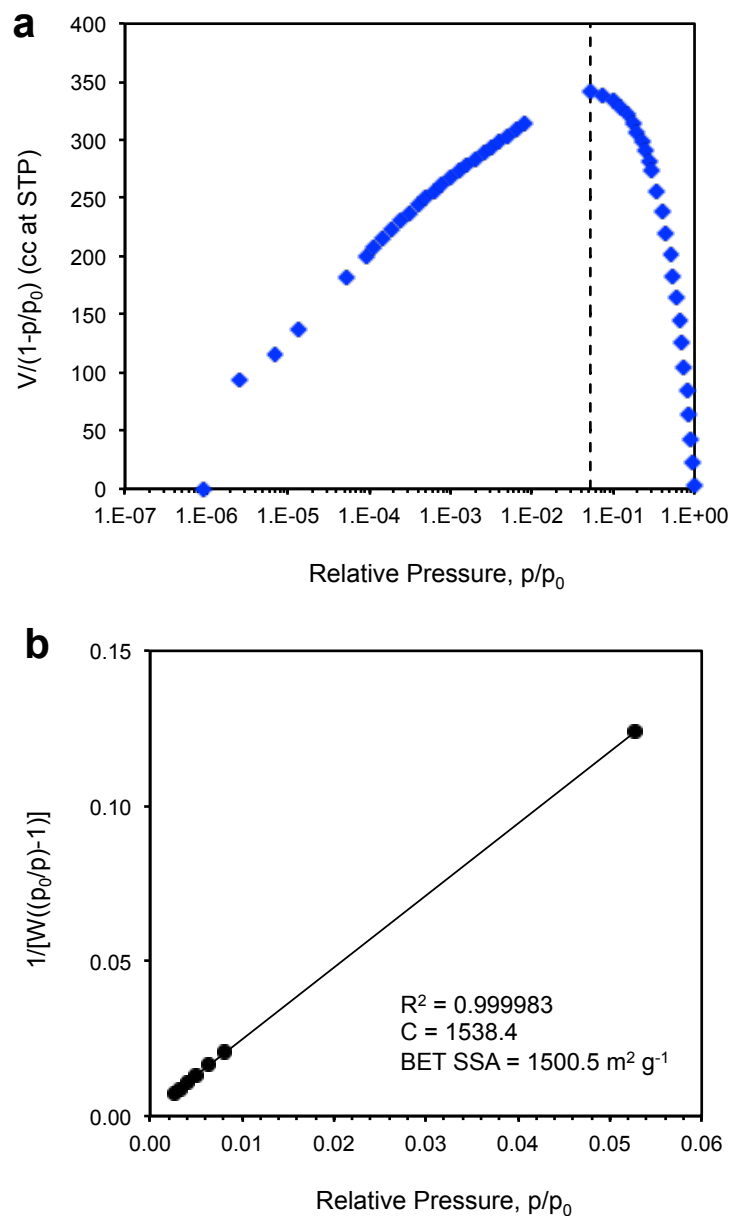


Figure S9. BET calculations and fittings for SU-MAC-600: (a) $V/(1-p/p_0)$ vs. p/p_0 with the points in the range below $p/p_0 = 0.053$ satisfy the first consistency criterion for applying the BET theory; and (b) linear fitting of the BET equation, which satisfies the second consistency criterion for applying the BET theory⁴.

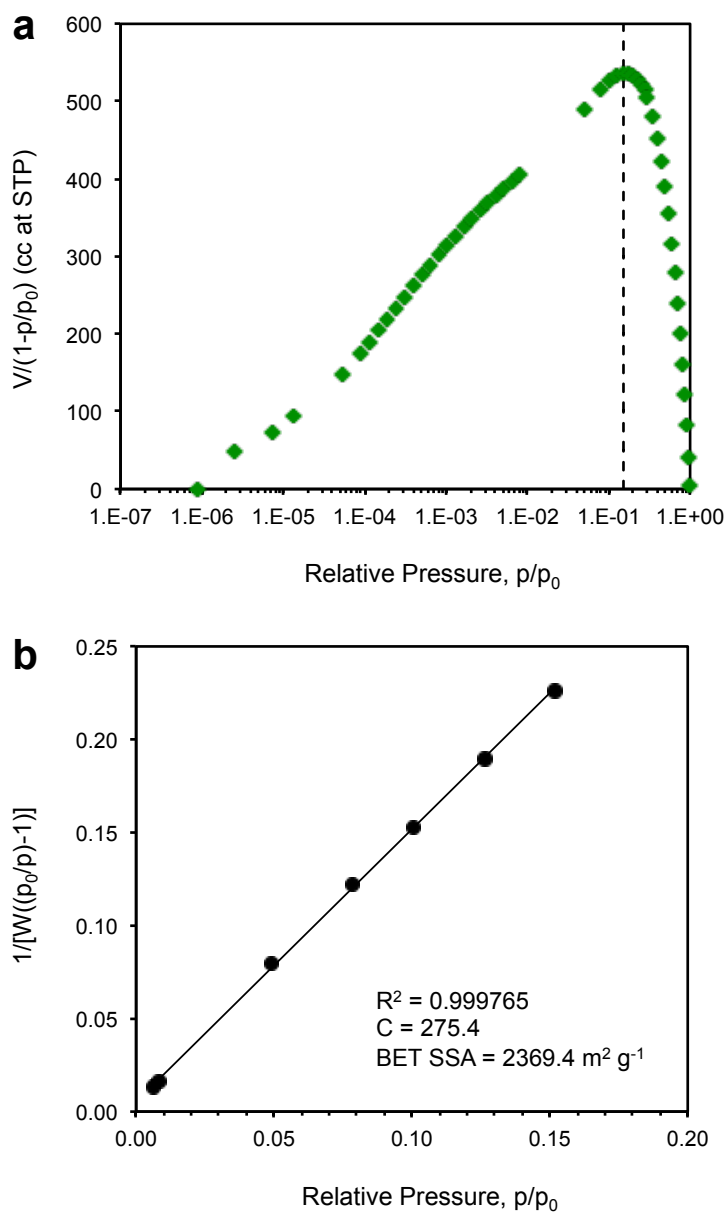


Figure S10. BET calculations and fittings for SU-MAC-800: (a) $V/(1-p/p_0)$ vs. p/p_0 with the points in the range below $p/p_0 = 0.152$ satisfy the first consistency criterion for applying the BET theory; and (b) linear fitting of the BET equation, which satisfies the second consistency criterion for applying the BET theory⁴.

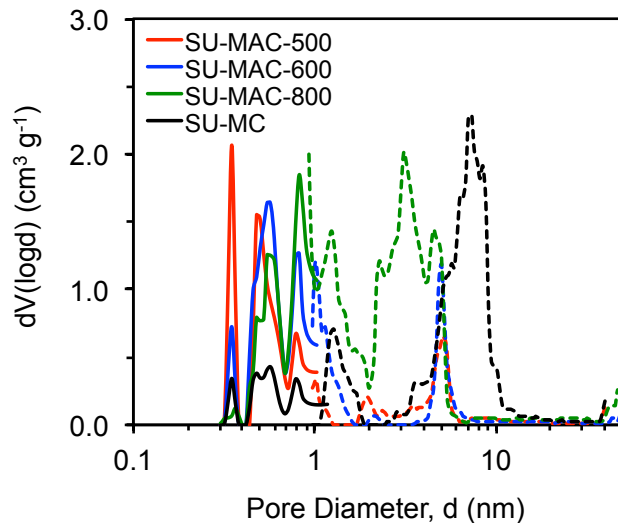


Figure S11. Pore size distributions (PSDs) of the SU-AC and SU-MAC samples, where the solid and broken lines represent the results from the CO₂ (273 K) and N₂ (77 K) adsorption tests, respectively

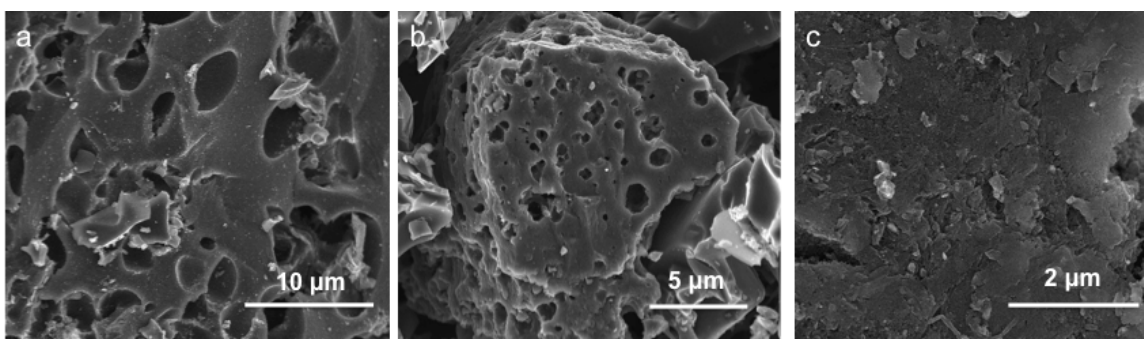


Figure S12. SEM images of (a) SU-MAC-500, (b) SU-MAC-600 and (c) SU-MAC-800, where SU-MAC-500 and SU-MAC-600 show considerable macroscopic porosity while SU-MAC-800 shows a shrinkage of the macropores due to high-temperature activation.

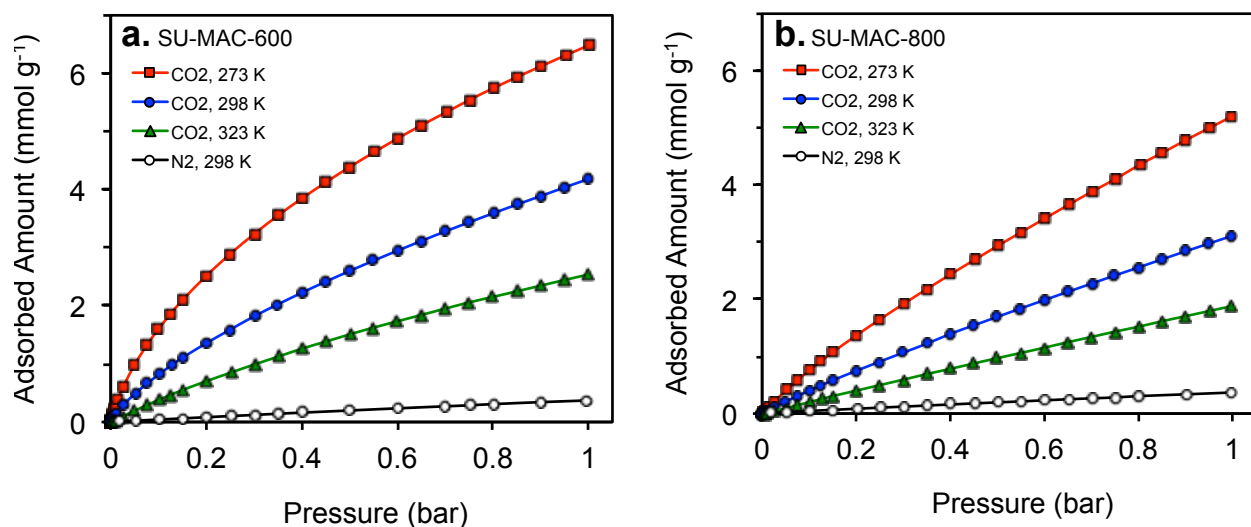


Figure S13. Gas adsorption performance of (a) SU-MAC-600 and (b) SU-MAC-800: CO₂ at 273, 298 and 323 K, and N₂ at 298 K. From SU-MAC-500 to SU-MAC-800, the CO₂ adsorption at 298 K shows a decreasing trend as the activation temperature increases, while all the three samples showed a similar N₂ adsorption capacity. The CO₂ isotherms become increasingly linear from SU-MAC-500 to SU-MAC-800, implying the weakened CO₂-sorbent interactions.

Calculation of Henry's law selectivity

To calculate the Henry's law selectivity of CO₂ over N₂, the CO₂ and N₂ isotherms at 273, 298 and 323 K were fitted to a virial-type thermal equation previously used to model gas adsorption on solid adsorbents⁵:

$$\ln p = \ln q + \frac{1}{T} \sum_{i=0}^m a_i q^i + \sum_{j=0}^n b_j q^j \quad (1)$$

where p , q and T are the pressure, gas adsorbed amount (in mmol g⁻¹) and temperature (in K), respectively. a_i and b_i are virial coefficients, which are the fitting parameters. m and n are the numbers of coefficients required for sufficient description of the isotherms. The Henry's constant (K_H) for each gas was then obtained using the virial coefficients of a_0 and b_0 , following Equation (2)⁶:

$$K_H = \exp(-b_0) \exp\left(-\frac{a_0}{T}\right) \quad (2)$$

The Henry's law selectivity of CO₂ over N₂ was then calculated using the ratio of the Henry's constants of CO₂ and N₂⁷:

$$S_{CO_2/N_2} = K_{H,CO_2}/K_{H,N_2} \quad (3)$$

The Henry's coefficients for samples SU-MAC-500, SU-MAC-600 and SU-MAC-800 are shown in Tables S1. The fittings of all isotherms in this study have R² greater than 0.99.

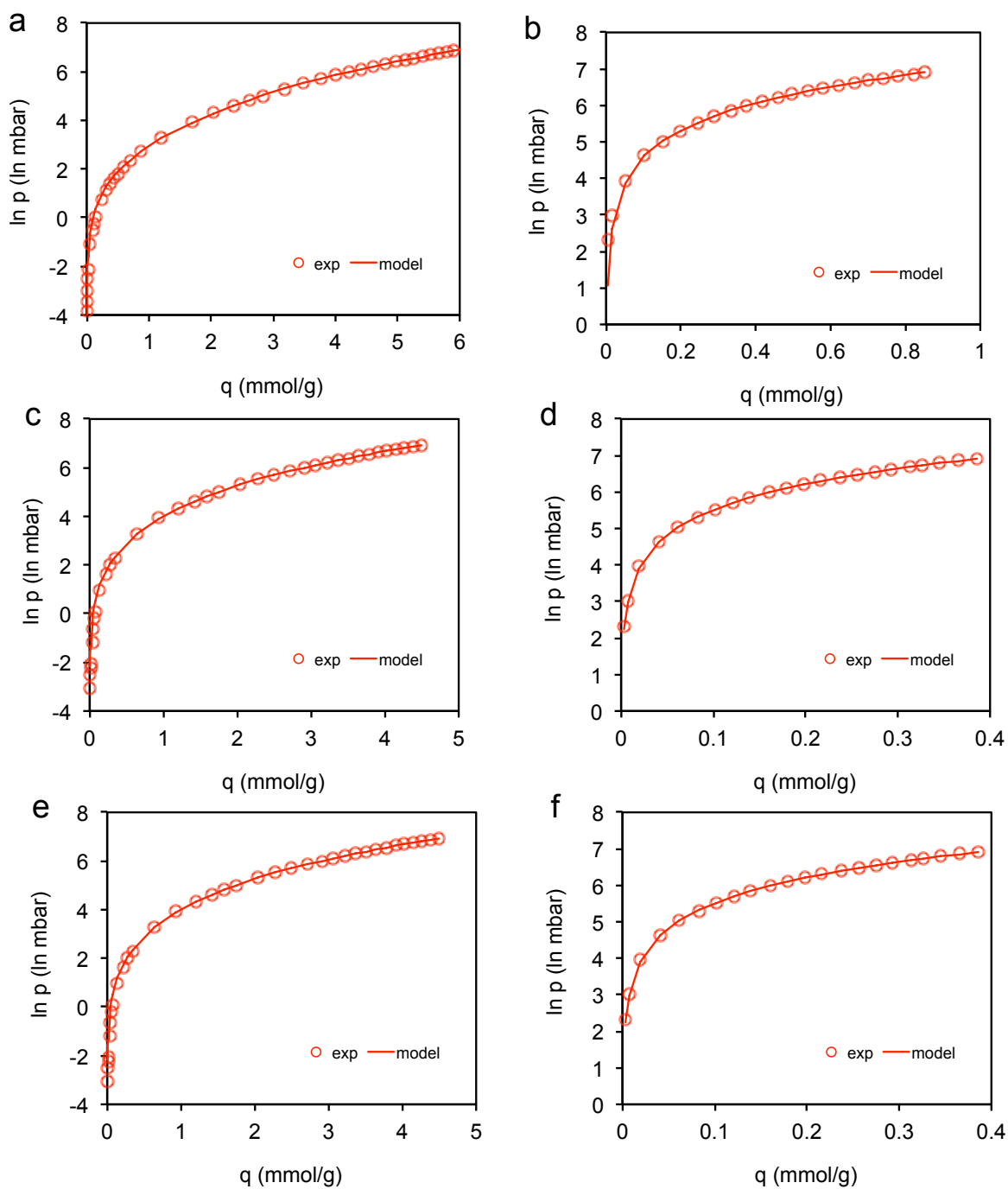


Figure S14. Virial fitting plots for the sample of SU-MAC-500: (a) (c) (e) CO₂ and (b) (d) (f) N₂ at (a) (b) 273 K, (c) (d) 298 K and (e) (f) 323 K.

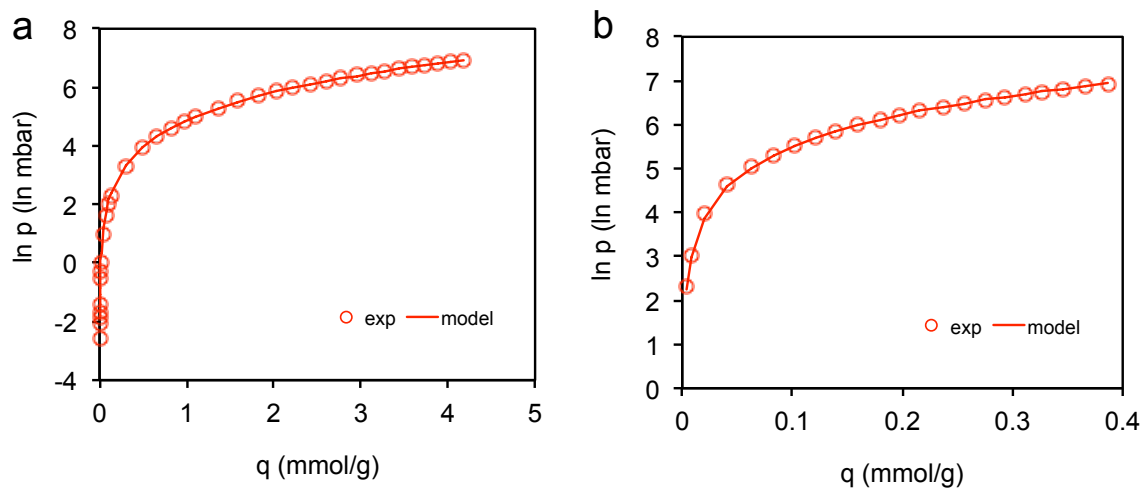


Figure S15. Virial fitting plots for the sample of SU-MAC-600: (a) CO_2 and (b) N_2 at 298 K.

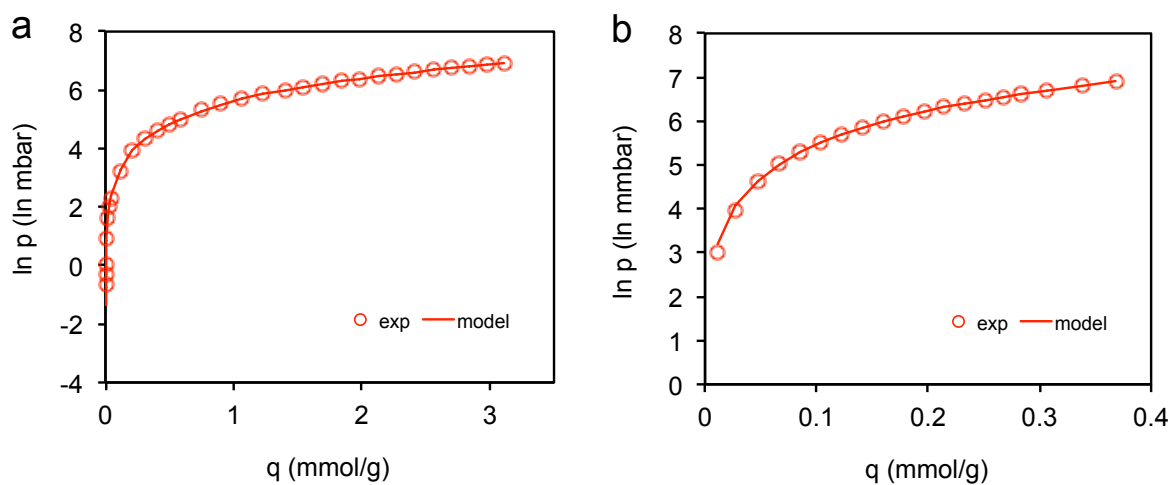


Figure S16. Virial fitting plots for the sample of SU-MAC-800: (a) CO_2 and (b) N_2 at 298 K.

Table S1. Henry's constants (K_H) and virial fitting results for the SU-MAC samples.

Sample	Gas, Temperature (K)	K_H (mmol g ⁻¹ mbar ⁻¹)	Fitting R ²	Error sum of squares
SU-MAC-500	CO ₂ , 273 K	113.521	>0.9999	4.329e-5
SU-MAC-500	N ₂ , 273 K	1.0635	0.9999	1.856e-4
SU-MAC-500	CO ₂ , 298 K	49.595	>0.9999	4.171e-5
SU-MAC-500	N ₂ , 298 K	0.3994	0.9999	2.995e-4
SU-MAC-500	CO ₂ , 323 K	12.304	>0.9999	1.099e-5
SU-MAC-500	N ₂ , 323 K	0.0635	0.992	9.628e-3
SU-MAC-600	CO ₂ , 298 K	12.796	>0.9999	1.352e-5
SU-MAC-600	N ₂ , 298 K	0.3966	0.9999	1.109e-4
SU-MAC-800	CO ₂ , 298 K	4.533	>0.9999	2.774e-5
SU-MAC-800	N ₂ , 298 K	0.5016	0.9998	2.555e-4

Ideal Adsorption Solution Theory (IAST)⁸

The selectivity (S_{IAST}) for gas mixtures of CO₂ and N₂ based upon the IAST theory is calculated as:

$$S_{IAST} = \frac{q_1/q_2}{p_1/p_2} \quad (4)$$

where q_1 and q_2 are the adsorbed amount of CO₂ and N₂ (in mmol g⁻¹), respectively; p_1 and p_2 are the partial pressure of CO₂ and N₂, respectively.

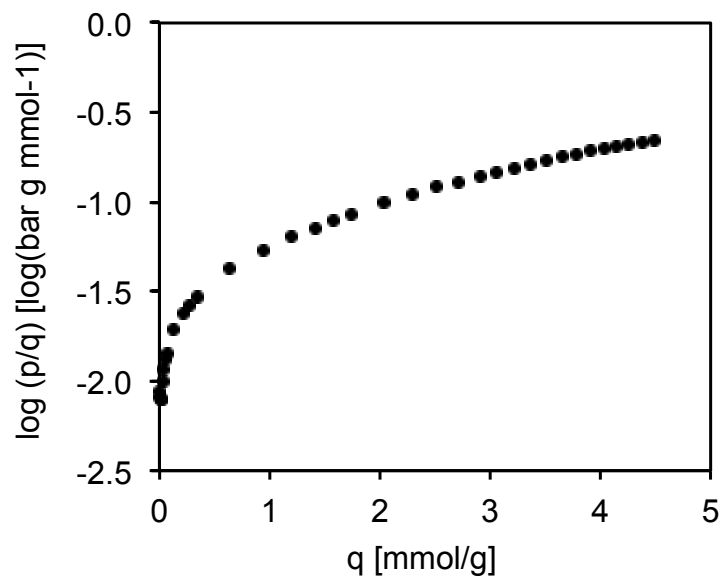


Figure S17. Virial plot of CO₂ adsorption at 298 K onto SU-MAC-500, where p and q represent the adsorption pressure and uptake, respectively.

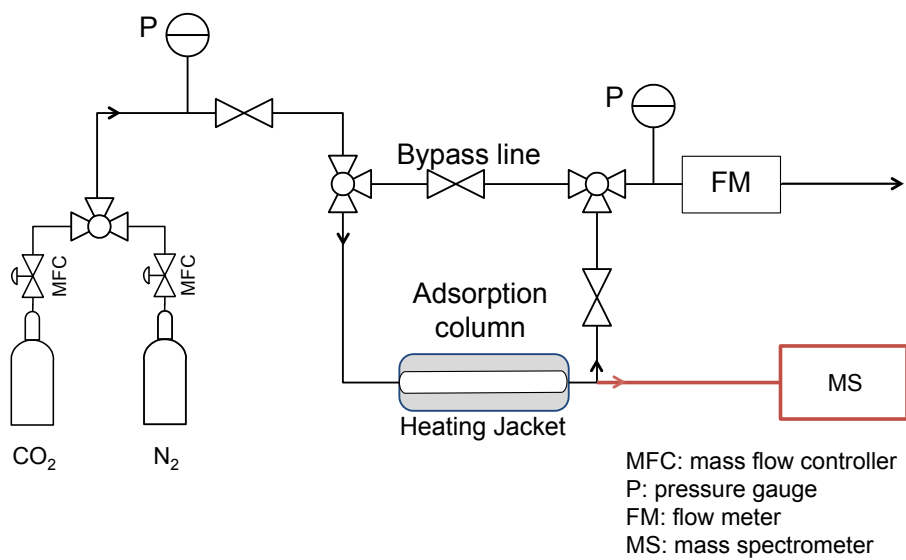


Figure S18. Experimental setup for dynamic column breakthrough experiments.

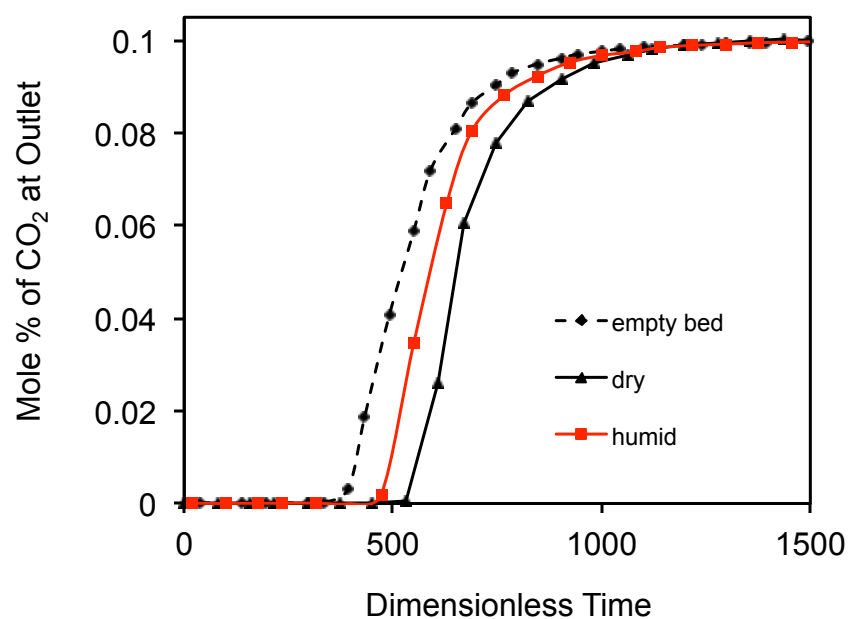


Figure S19. CO₂ mole fraction profiles detected at the outlet of the adsorption column for empty bed and SU-MAC-500 with a mixed gas input of 10%/90% CO₂/N₂ at 323 K and total pressure of 1 bar under dry and humid conditions (dimensionless time: $\tau = tu/\varepsilon L$).

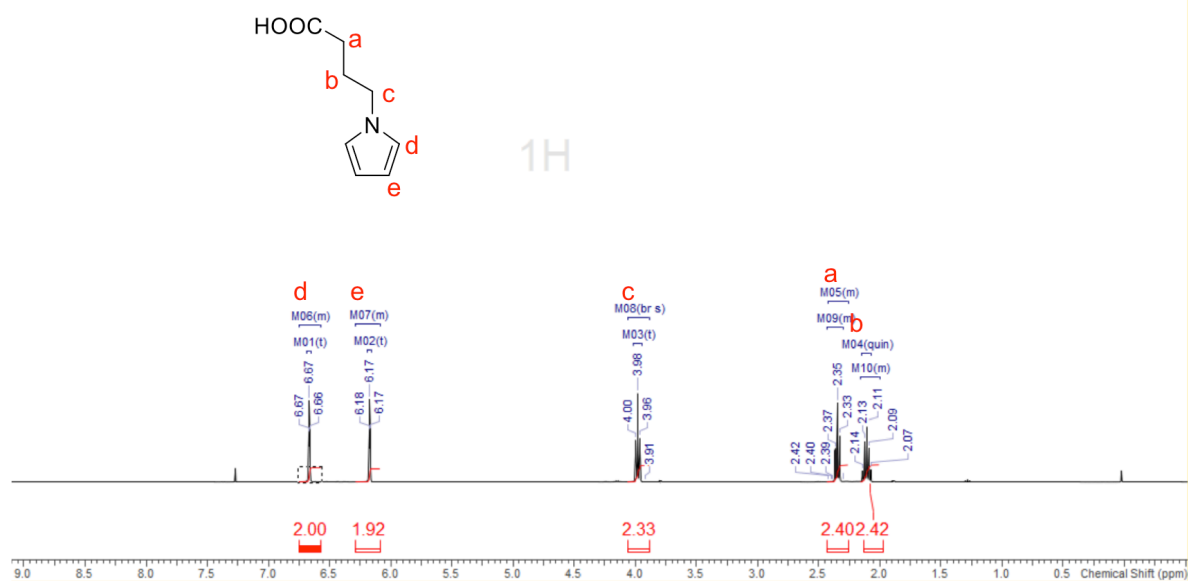


Figure S20. ¹H NMR spectrum of 4-(Pyrrol-1-yl)butanoic acid.

Table S2. Summary of textual properties and CO₂ capture performances of SU-MAC materials in comparison to literature reported physisorptive micro- and mesoporous carbons.

Material	Activation	Properties				CO ₂ Capacity at 298 K		Henry's law CO ₂ /N ₂ Selectivity ^d	IAST CO ₂ /N ₂ Selectivity ^e	Reference
		BET SSA ^a (m ² g ⁻¹)	V_micro ^b (cm ³ g ⁻¹)	V_ultramicro ^c (cm ³ g ⁻¹)	N wt%	(mmol g ⁻¹)				
						0.1 bar	1 bar			
HPC5b2-1100	Yes	2734	-	-	-	-	~3	-	-	Srinivas, <i>et al.</i> ⁹
MCs-800	No	974	0.10	-	24.2 (at%)	0.8	2.4	-	-	Lee, <i>et al.</i> ¹⁰
NPC-60-800	No	521	-	-	4.3	0.7	2.4	-	-	Gu, <i>et al.</i> ¹¹
sOMC	No	2255	-	-	-	0.3	2.0	9:1	10:1	Yuan, <i>et al.</i> ¹²
mPMF	No	930	0.21	-	39.6	-	2.0 ^f	-	-	Tan, <i>et al.</i> ¹³
H-NMC-2.5	No	537	0.17	-	13.1	1.0	2.8	37:1	35:1	Wei, <i>et al.</i> ¹⁴
A-NMC	Yes	1417	0.40	-	6.7	0.8	3.2	-	-	Wei, <i>et al.</i> ¹⁴
HCM_DAH-1	No	670	0.20	-	4.1	0.7	2.6	28:1	30:1	Hao, <i>et al.</i> ¹⁵
HCM-DAH-1- 900-3	Yes	2160	0.53	-	2.7	0.7	2.9	13:1	22:1	Hao, <i>et al.</i> ¹⁵
MCN/C	No	338	0.06	-	24.9	0.7	2.4	-	40:1	Deng, <i>et al.</i> ¹⁶
RFL-500	No	467	0.21	-	1.9	0.9	3.1	-	29:1	Hao, <i>et al.</i> ¹⁷
a-NDC6	Yes	1360	0.57	-	4.8	0.86	4.3	34:1	30:1	Chandra, <i>et al.</i> ¹⁸
CN-950		1979	-	-	4.32	1.4	4.3	30:1	12:1	Ma, <i>et al.</i> ¹⁹
CP-2-600	Yes	1700	0.74	0.21	10.14	0.76	3.9	-	10:1	Sevilla, <i>et al.</i> ²⁰
AS-2-600	Yes	1260	0.55	0.52	-	0.82	4.8	-	9:1	Sevilla, <i>et al.</i> ²¹
VR-93-M	Yes	2895	1.28	0.8	-	0.51	4.59	-	7:1	Wahby, <i>et al.</i> ²²

NPC-650	Yes	1561	0.65	-	4.11	0.56	3.1	13:1	8:1	Wang, <i>et al.</i> ²³
NG7	Yes	979.6	0.412	-	4.19	0.65	2.7	18:1	18:1	Kemp, <i>et al.</i> ²⁴
Bamboo-3-873	Yes	1846	-	0.36	-	0.93	4.5	-	16:1	Wei, <i>et al.</i> ²⁵
K4-700	Yes	1745	0.7	0.39	-		3.45	-	-	Hu, <i>et al.</i> ²⁶
ATS-2-700	Yes	1330	0.54		-	0.73	3.3	-	15:1	Sevilla, <i>et al.</i> ²⁷
CPC 550	Yes	1630	0.59	0.35	7.88	1.48	5.8	59:1	21:1	Ashourirad, <i>et al.</i> ²⁸
UC-650-2	Yes	1394	0.52	0.65 (<1 nm)	1.64	0.88	4.4	17:1	12:1	Bai, <i>et al.</i> ²⁹
SNS2-20	Yes	2100	0.93	0.35	-	1.6	4.48	-	36:1	Kim, <i>et al.</i> ³⁰
C-K-5	Yes	1160	0.52	0.45 (<1 nm)	-	1.15	4.7	-	-	Ludwinowicz, <i>et al.</i> ³¹
CG 700 2-1	Yes	1624	0.589	-	-	-	4.42	-	-	Travis, <i>et al.</i> ³²
NPC-2	Yes	1255.9	0.52	-	5.21	1.05	4.02	-	32:1	Wan, <i>et al.</i> ³³
NC-650-1	Yes	1483	0.57	0.61 (<1 nm)	4.56	1	4.26	29	23:1	Yang, <i>et al.</i> ³⁴
nZDC-700	No	950	0.35	-	-	1.1	3.5	-	76:1	Gadipelli, <i>et al.</i> ³⁵
SU-MAC-500	Yes	941	0.34	0.30	5.8	1.4	4.5	124 : 1	39:1	This work
SU-MAC-600	Yes	1500	0.50	0.27	4.0	0.8	4.2	32:1	22:1	This work
SU-MAC-800	Yes	2369	0.63	0.18	3.2	0.4	3.1	9:1	11:1	This work

a) *Brunauer–Emmett–Teller* (BET) specific surface area; b) cumulative micropore volume with diameter ≤ 2 nm; c) cumulative ultramicropore volume with diameter ≤ 0.7 nm; d) Henry's Law selectivity at 298 K; e) ideal adsorption solution theory (IAST) selectivity at 298 K for the mixture of 0.1/0.9 CO₂/N₂ at 1 bar.

Table S3. Summary CO₂ and N₂ adsorption capacities at 1 bar and different temperatures and Henry's law CO₂/N₂ selectivities at 298 K for the SU-MAC samples.

Sample	CO ₂ Capacity (mmol g ⁻¹)			N ₂ Capacity (mmol g ⁻¹)	Henry's law CO ₂ /N ₂ Selectivity
	273 K	298 K	323 K	298 K	
SU-MAC-500	6.03	4.50	3.06	0.39	124:1
SU-MAC-600	6.49	4.18	2.54	0.37	32:1
SU-MAC-800	5.20	3.11	1.88	0.37	9:1

Table S4. Summary of textual properties and chemical compositions of the SU-MC and SU-MAC samples.

Material	BET SSA ^a (m ² g ⁻¹)	V _{total} ^b (cm ³ g ⁻¹)	V _{micro} ^c (cm ³ g ⁻¹)	V _{ultramicro} ^d (cm ³ g ⁻¹)	V (d <0.5 nm) ^e (cm ³ g ⁻¹)	Total C ^f (wt%)	Total N ^f (wt%)	N-5 ^g (wt%)	N-6 ^g (wt%)
SU-MC	817	0.88	0.17	0.12 / 0.11	0.06	67.1	8.0	7.5	-
SU-MAC-500	941	0.47	0.34	0.30 / 0.30	0.14	56.8	5.77	5.01	0.76
SU-MAC-600	1500	0.70	0.56	0.27 / 0.25	0.10	57.0	4.0	3.69	0.31
SU-MAC-800	2369	1.34	0.68	0.18 / 0.09	0.05	65.4	3.19	2.66	0.54

a) *Brunauer–Emmett–Teller* (BET) specific surface area; b) total pore volume at $p/p_0 = 0.995$; c) cumulative micropore volume with diameter ≤ 2 nm determined by the NLDFT method based upon the N₂ isotherms; d) cumulative ultramicropore volume with diameter ≤ 0.7 nm determined by the NLDFT method based upon the CO₂ isotherms / N₂ isotherms; e) cumulative ultramicropore volume with diameter ≤ 0.5 nm determined by the NLDFT method based upon the CO₂ isotherms; f) The total C and N contents are determined using elemental analysis based on the complete and instantaneous oxidation of the sample by "flash combustion" at 1080 °C, which converts all organic and inorganic substances into combustion products. The analytes concentration were compared to an Atropine standard; g) Types of Nitrogen on the surface were determined using X-ray photoelectron spectroscopy (XPS). Peak deconvolution was performed to elucidate the types of nitrogen in the carbon samples by assuming the nitrogen in the bulk and on the surface are relatively the same.

References

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